Highly conductive "packed foams": A new concept for the intensification of strongly endo- and exo-thermic catalytic processes in compact tubular reactors

Carlo Giorgio Visconti, Gianpiero Groppi *, Enrico Tronconi

Politecnico di Milano, Department of Energy, Laboratory of Catalysis and Catalytic Processes, Via La Masa 34, 20156 Milano, Italy

We report a preliminary assessment of the heat transfer properties of innovative structured tubular reactors based on the conductive "packed foams" concept. In such reactors the catalyst is loaded in the form of small pellets packed in the voids of a highly conductive open-cell foam. This enables a catalyst inventory which is significantly higher than that of reactors loaded with the same structured substrate washcoated with catalytic material, while still exploiting the potential of enhanced heat removal through a highly conductive structured skeleton. By comparing the heat transfer performances of the packed foams with those of conventional packed beds of pellets and with those of bare (naked) open cell-foams, we show that the presence of the conductive foam within the packed bed of pellets grants optimal heat transfer performances as a result of a synergy between the enhanced conductive heat transfer in the solid structure of the foam and the effective heat transfer at the wall-bed boundary typical of packed beds. The former mechanism, which is flow independent, controls the effective radial conductivity. The latter one enhances the wall heat transfer coefficient and contributes to increase the overall heat transfer coefficient, particularly at high flow rates. This opens new perspectives for the intensification of highly exothermic/endothermic catalytic processes which require greater catalyst inventories in the reactor than those achievable with washcoated open-cell foams.

Keywords: Packed foams, Highly conductive structured reactors, Process intensification, Heat transfer

1. Introduction

It is widely recognized that spatially structured reactors are at the very heart of process intensification [1,2] and it has been shown that significant enhancement of radial heat transfer rates in multi-tubular catalytic reactors with external cooling can be achieved if the random packings of catalyst pellets are replaced by structured catalysts with thermally connected, highly conductive foams [3–6]. Nevertheless, some issues still exist that prevent the application of this technology at the industrial scale.

For example, in the case of non-adiabatic reactors loaded with highly conductive washcoated open-cell foams (or sponges) or honeycomb monoliths, due to the thin washcoat layer (usually less than 200 μm thick [7]), the catalyst inventory (hold-up) may be insufficient, thus limiting the potential advantages associated with the adoption of conductive structured reactors. Indeed, the fact that the overall load of catalytically active phase in washcoated substrates is usually much less than the amount of catalyst in a packed bed of bulk pellets limits the productivity per reactor volume of the reactions under total or partial kinetic control which can be met in the process industry [1,8]. A very clear example in this sense is offered by the highly exothermic Fischer-Tropsch synthesis, which would definitely benefit from the adoption of highly conductive structured substrate (such as highly conductive honeycombs), but requires catalyst inventories much higher than those typical of washcoated reactors in order to fully exploit the concept [9,10]. Similar results have been reported for the methanol synthesis over highly conductive open-cell foams [7].

Also, catalyst loading and unloading in the reactor are particularly critical in washcoated reactor technologies. This is due to the fact that the methods for loading, packaging, sealing and unloading structured catalysts in the synthesis reactors are different from those well established for randomly packed catalysts, and cannot be directly derived from the experience made in stationary environmental installations [1]. In addition to that, the adhesion of the active layer to the substrate is a challenging aspect, which requires both the optimization of the preparation methods [11,12] and the careful handling of the washcoated sample. Last but not

Article history:
Received 4 December 2015
Received in revised form 12 February 2016
Accepted 21 February 2016
Available online 26 March 2016

* Corresponding author.
E-mail address: gianpiero.groppi@polimi.it (G. Groppi).

Published Journal Article available at: http://dx.doi.org/10.1016/j.cattod.2016.02.060

Nomenclature

$\widehat{c_p}$	Gas specific heat (J/kg/K)
D	Internal tube diameter (m)
d_c	Foam cell diameter (m)
d_p	Particle diameter (m)
h_w	Wall heat transfer coefficients (W/m^2/K)
$k_{e,ax}$	Effective axial conductivity (W/m/K)
$k_{e,r}$	Effective radial conductivity (W/m/K)
k_f	Gas conductivity (W/m/K)
k_s	Solid conductivity (W/m/K)
L	Bed length (m)
$Nu_{w,dc} = h_w d_c/k_f$	Wall nusselt number, foam
$Nu_{w,dp} = h_w d_p/k_f$	Wall nusselt number, packed bed
r	radial coordinate (m)
$Re_{dc} = \rho_f v d_c/\mu_f$	Reynolds number, foam
$Re_{dp} = \rho_f v d_p/\mu_f$	Reynolds number, packed bed
T	Temperature (K)
T_{TC}	Thermostatic chamber temperature (K)
T_w	Tube wall temperature (K)
U	Overall heat transfer coefficient (W/m^2/K)
v	Empty tube velocity (m/s)
x	Axial coordinate (m)
ε_T	Foam total porosity
ε_H	Foam hydrodynamic porosity
ε_{PB}	Packed bed porosity
μ_f	Gas viscosity (kg/m/s)
ρ_f	Gas density (kg/m^3)

least the possibility of regenerating and replacing spent active phase is particularly critical in washcoated reactors and requires the development of dedicated technologies.

To overcome those issues the adoption of "packed structured" reactors has been recently proposed by the group of Kapteijn at TU Delft (packed cross flow structures [8]) and by our group at Politecnico di Milano together with colleagues at Eni (packed honeycomb monoliths [13] and packed foams [14]). In such reactors, the catalyst is loaded in the form of small pellets randomly packed in the voids of structured substrates.

In the case of cross flow structures, which are particularly beneficial with gas-liquid processes because of the enforced radial liquid flow, it has been shown [8] that the presence of a packing boosts the overall heat transfer coefficient, while not sacrificing much in catalyst inventory compared to the randomly packed bed. Also, the pressure drop is approximately half that of the randomly packed bed due to the voidage at the internal walls of the packed channel network [8].

Herein we investigate for the first time the concept applied to conductive open-cell metal foams in single phase tubular reactors (Fig. 1). Notably, the heat transfer mechanism within conductive foams is totally different from that characteristic of cross flow structures. As we have shown in previous papers [2,7], indeed, while the conductive (static) contribution of the connected solid is by far the controlling term determining the effective radial thermal conductivity of metallic foam structures in tubular reactors [3], the wall heat transfer coefficient is controlled primarily by a conductive mechanism in the fluid phase [15]. The effective radial conductivity is well approximated by the theoretical model of Lemlich [16] (Eq. (1)), which states that the ratio of the effective conductivity ($k_{e,r}$) to the bulk conductivity of the material (k_s) is equal to one third of the relative volumetric density of the foam (1-ε_T) [3]. The wall heat transfer coefficient (h_w, Eq. (2)) is essentially proportional to the gas conductivity (k_f) and to the reciprocal of the cell size (d_c) [15]. Eq. 2 also shows that a positive effect associated with a convective

Table 1
Geometrical and morphological properties of the investigated packings.

Sample	Nominal pore density (PPI)	1-ε_H (–)	1-ε_T (–)	d_c (mm)	d_p (mm)
FeCrAlY foam	10	0.060	0.051	5.09	–
Al-6101 foam	40	0.055	0.055	2.00	–
Al_2O_3 pellets	–	–	–	–	0.3

contribution to the wall heat transfer coefficient is present at the wall, but this is of minor importance at least for Reynolds number (Re_{dc}) below 255.

$$k_{e,r}/k_s = (1-\varepsilon_T)/3 \quad (1)$$

$$h_w = k_f/d_c\left(7.18 + 0.029 Re_{dc}^{0.8}\right) 4 < Re_{dc} < 255 \quad (2)$$

That said, no information is available in the literature to date on the heat transport properties of packed foams. Accordingly, in order to elucidate what are the dominant contributions to heat transfer in packed foams and what are the interactions between the packed pellets and the structured substrate, in this work, for the first time, we preliminarily compare by experiments and by modeling the heat transfer performances of packed foams with those of packed beds of micro-pellets and of bare open-cell foams.

2. Experimental

2.1. Material and methods

2.1.1. Packed foams

In order to assess the heat transfer performances of conductive packed sponges, open-foam samples made of FeCrAlY (purchased from Porvair) and 6101 T6 aluminum (purchased from ERG Aerospace) alloys (named samples A and E in our previous works [3,14]), representative of metallic materials with very different thermal conductivities (about 16 W m^{-1} K^{-1} for FeCrAlY and 218 W m^{-1} K^{-1} for Al-6101 alloy at room temperature), were selected. The geometrical and morphological properties of these materials are given in Table 1, while details on the experimental techniques used to evaluate such properties can be found elsewhere [3]. Notably, the FeCrAlY foam has hollow struts, as demonstrated by different values of the total (ε_T) and the hydrodynamic (ε_H) porosities (with $\varepsilon_T > \varepsilon_H$), while the Al-6101 foam has solid struts. Nevertheless, the hydrodynamic porosity of the two samples is very similar, which results in a similar space available to pack the micro-pellets.

Cylindrical samples with a diameter of 28 mm (+0.01, −0.00 mm) and a length of 25 mm (FeCrAlY sample) or 50 mm (Al-6101 sample), precisely cut out of larger panels (as provided by Porvair and ERG AEROSPACE, respectively) by electro-erosion, were used in the experiments. Two axial through holes of 3.28 mm diameter were then drilled in each sample at two different radial positions, namely at the center-line and 9 mm from the center. These holes allowed for the tight insertion of two stainless steel thermowells (O.D. 1/8") protecting sliding K-type thermocouples for the measurement of the internal axial temperature profiles during the heat transfer experiments (cf. Section 2.2).

Once loaded the foams in the tubular reactor and put the thermowells in place, the foam bed was randomly packed with γ-Al_2O_3 pellets with a diameter of 0.3 mm. These pellets, kept in position by layers of quartz wool located up- and down-stream the bed, were observed to be free-flowing in the foam matrix during both the loading and the unloading procedures. The amount of pellets loaded in each foam was found to be reproducible and to correspond exactly to the amount of pellets in a packed-bed with the same volume of the voids (cells) of the foam and ε_{PB} = 0.37. This clearly indicates that, likely due to the very large d_c/d_p ratio, the small pellets can uniformly fill the voids of the foam structure

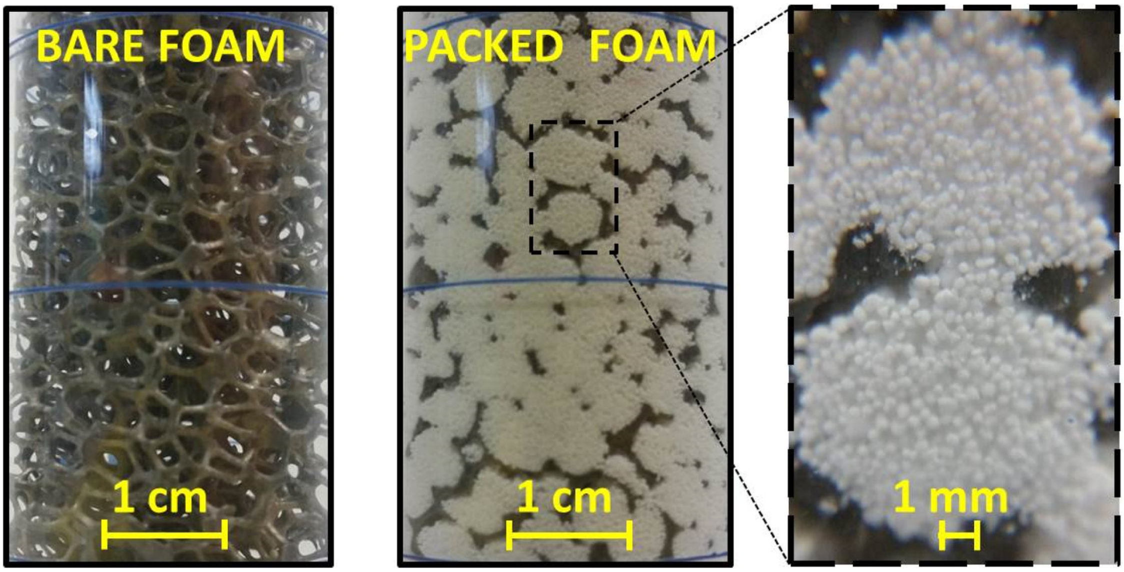

Fig. 1. Bare and packed foams.

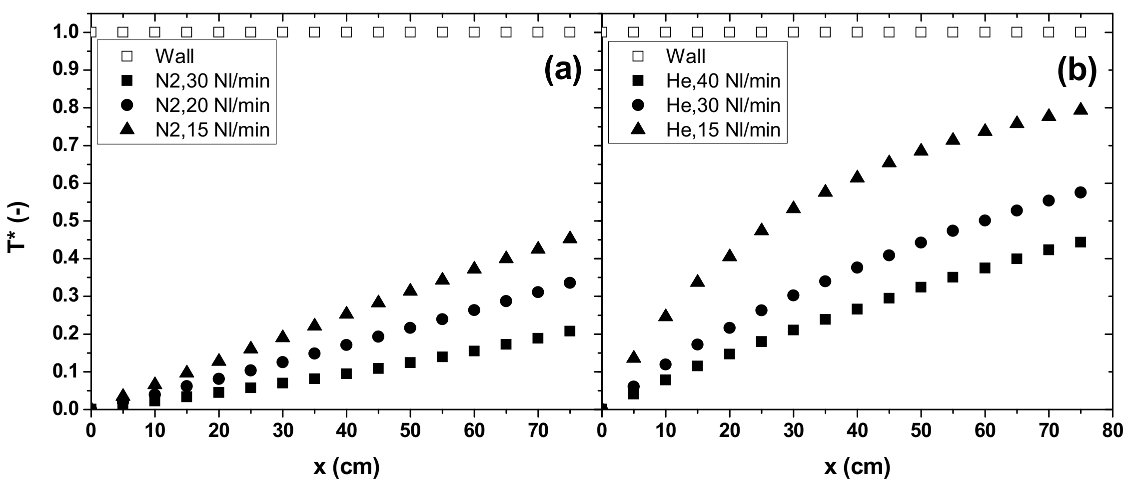

Fig. 2. Dimensionless temperature profiles measured during experiments with the randomly packed bed of 0.3 mm Al_2O_3 spheres under flows of (a) N_2 or (b) He, T_{TC} = 400 °C.

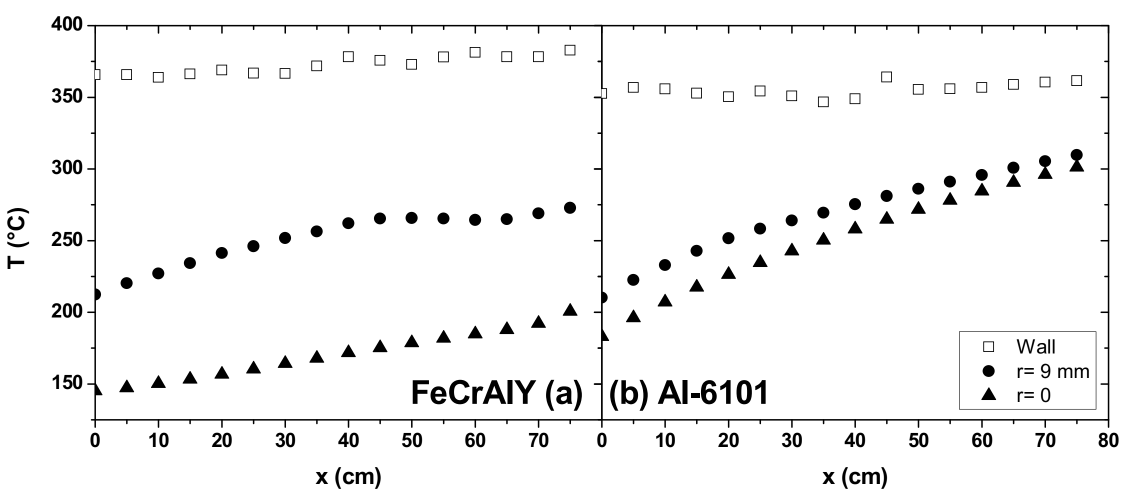

Fig. 3. Temperature profiles measured during experiments with the packed foams. T_{TC} = 400 °C, 60 Nl/min N_2: (a) FeCrAlY; (b) Al-6101.

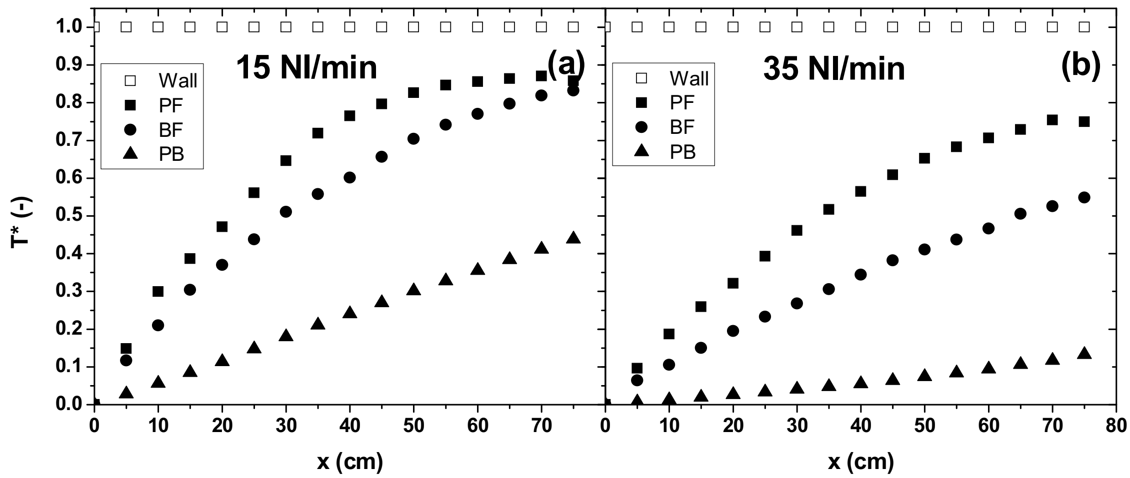

Fig. 4. Dimensionless temperature profiles (wall and centerline) measured with the packed-bed, Al-6101 packed foam and Al-6101 bare foam fed with N_2 at (a) 15 Nl/min and (b) 35 Nl/min. T_{TC} = 300 °C.

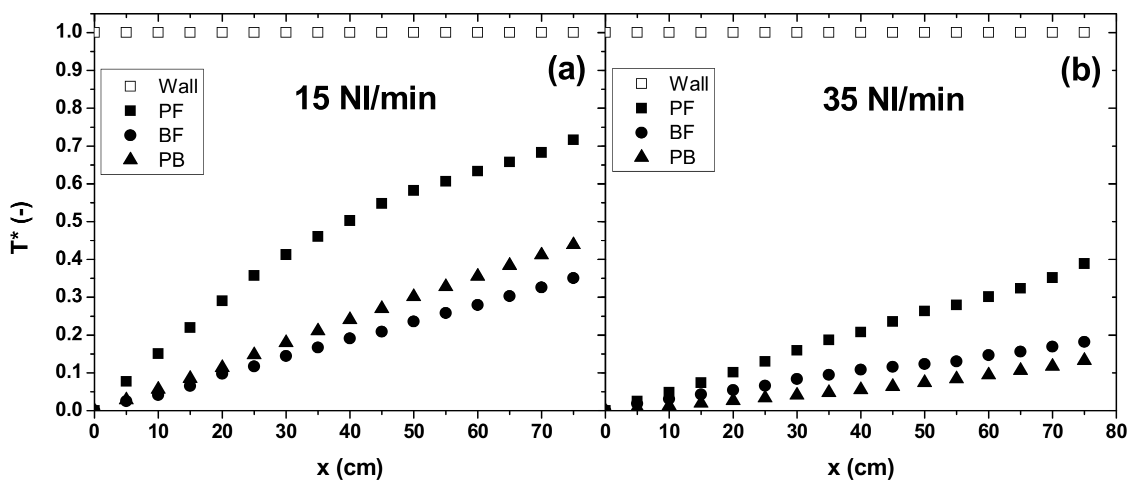

Fig. 5. Dimensionless temperature profiles (wall and centerline) measured with the packed-bed, FeCrAlY packed foam and FeCrAlY bare foam fed with N_2 at (a) 15 Nl/min and (b) 35 Nl/min. T_{TC} = 300 °C.

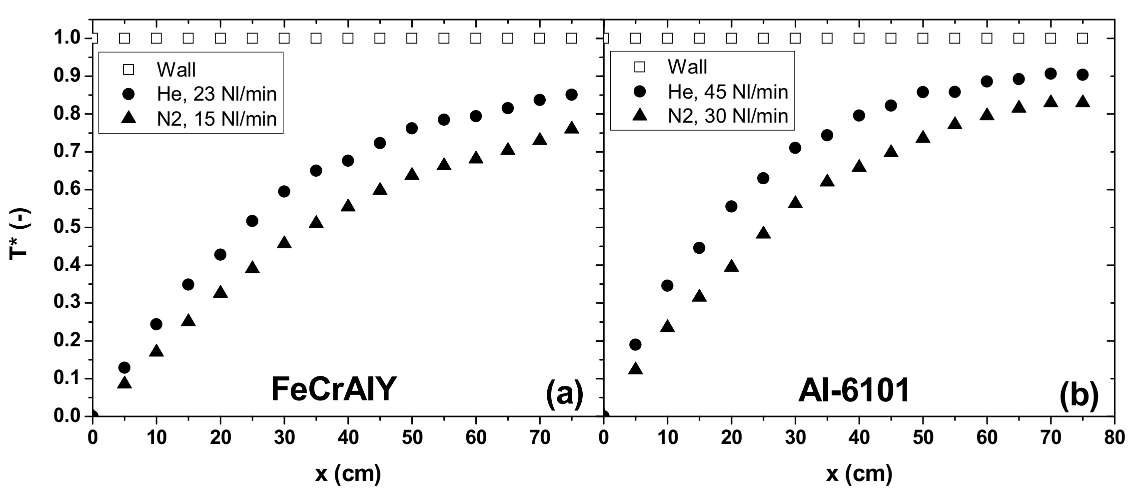

Fig. 6. Dimensionless temperature profiles (wall and centerline) measured with (a) FeCrAlY and (b) Al-6101 packed foams, both fed with N_2 and He at the same flowing heat capacity. T_{TC} = 400 °C for tests with FeCrAlY packed foams and T_{TC} = 500 °C for tests with Al-6101 packed foams.

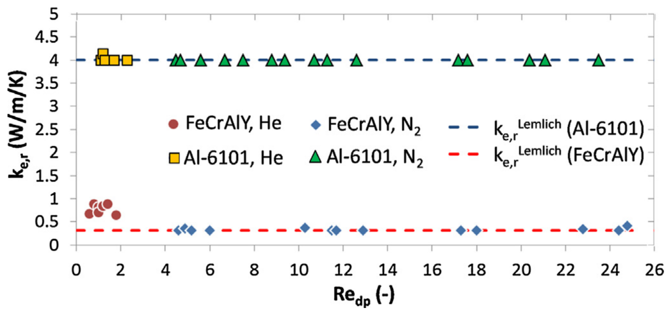

Fig. 7. Estimates of $k_{e,r}$ for FeCrAlY and Al-6101 pack foams and comparison with the predictions obtained with the correlation by Lemlich [16].

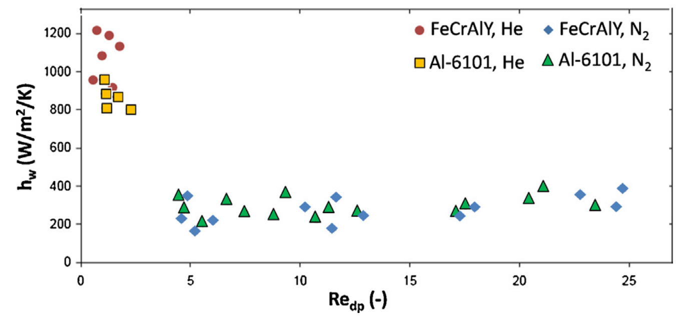

Fig. 8. Estimates of h_w for FeCrAlY and Al-6101 packed foams.

and negligible effects on the packing effectiveness are given by the presence of the solid struts of the foam. The visual inspection of the packed foam loaded in a transparent tube with 28 mm I.D. confirmed these results (Fig. 1).

Notably, the weight of pellets loaded in each packed foam (about 760 kg/m^3), which is very similar to that of a packed bed with the same volume (800 kg/m^3), is approximately one order of magnitude higher than those achievable by washcoating the same open-cell foams with 100 μm thick γ-Al_2O_3 layers (calculated using the expression given in [7]), further confirming that packed foams are, at least in principle, a valid alternative to washcoated foams when the small catalyst inventory is an issue.

2.1.2. Packed bed

The heat transfer performances of the packed foams (PF) were compared to those of a randomly packed bed (PB) of pellets with average diameter equal to 0.3 mm. In particular, the same Al_2O_3 pellets used to perform experiments with the "packed foam" were used and similar experiments were carried out (see Section 2.2). However, due to some limitations in our set-up, only one K-type thermocouple sliding in a thermowell with a O.D. of 1/8" located at the tube centerline was used in this case to measure the T-profile.

2.1.3. Bare open-cell foams

The heat transfer performances of the packed foams were also compared to those of the same foams, tested without the packing of micro-pellets. In the following these samples will be referred to as bare foams (BF). Notably, results obtained with the bare foams have been already reported and discussed in our previous works [3,15]. Accordingly, in the following of this paper they will be only recalled for comparison purposes.

2.2. Heat transfer set-up and experiments

Flow heat transfer runs were performed in a test rig consisting of a horizontal tube (I.D. = 28 mm) inserted into a thermostatic oven with air recirculation (Mazzali Thermotest) and loaded either with the foam samples, or the packed bed and the packed foam configurations. Due to the fact that the foams O.D. and the reactor I.D. are all 28 mm, the structured substrates were loaded in the reactor with interference fit both in the case of experiments with PF and in those with BF.

The test tube was equipped with an external thermowell (also containing a sliding K-type thermocouple) in tight contact so to measure the outer skin temperature profile, and with external fins to improve the external heat transfer. The optimal contact of the fins with the test tube was ensured by a thin copper foil rolled around the tube, except in the section upstream of the bed, which was shielded for insulation. Upstream from the test samples an additional cylindrical FeCrAlY foam was loaded, used as a static mixer to minimize entry length effects and to ensure uniform gas flow and temperature distribution. Both upstream and downstream the packed (foam) bed, layers of quartz wool 10–20 mm long were also used so to keep fixed the packed pellets.

During the experiments, carried out heating the oven at different temperatures (between 200 and 500 °C), a highly conductive gas (99.999 vol% He, $k_{f,NTP} = 0.147\ Wm^{-1}K^{-1}$) and a poorly conductive gas (99.995 vol% N_2, $k_{f,NTP} = 0.026\ Wm^{-1}K^{-1}$) at different flow rates (10–60 $Nlmin^{-1}$) were fed at atmospheric pressure to the reactor. Steady-state axial T-profiles inside the packing and at the pipe skin were measured with a resolution of 5 mm. Each temperature profile was taken twice during each run to check its reproducibility, corresponding to a local variation of less than 1 °C between the two measurements in the same point.

More details on the experimental apparatus and methods, as well as more details on the experimental errors associated with the temperature measurements, can be found in our previous papers [3,15].

2.3. Mathematical analysis of the experimental data

Following the approach already presented in [3,15], a classical steady-state non-reactive 2D two-parameters pseudo-homogeneous model (Eqs. (3)–(5)) was used to analyse the results:

Enthalpy balance

$$\rho v\widehat{c_p}\partial T/\partial x = k_{e,ax}\partial^2 T/\partial x^2 + k_{e,r}\left(\partial^2 T/\partial r^2 + 1/r\partial T/\partial r\right) \quad (3)$$

Boundary conditions

$$k_{e,ax}\partial T/\partial x = \rho v\widehat{c_p}\left(T(0,r) - T_{-\infty}\right)@x = 0;\ \partial T/\partial x = 0@x = L; \quad (4)$$

$$\partial T/\partial r = 0@r = 0;\ k_{e,r}\partial T/\partial r = h_w\left(T_w(x) - T(x,R)\right)@r = D/2; \quad (5)$$

The three model parameters, i.e. the effective axial and radial thermal conductivities of the bed ($k_{e,ax}$ and $k_{e,r}$) and the wall heat transfer coefficient (h_w), were estimated in each experiment by fitting the experimental T-profiles. In order to reduce statistical correlation between parameter estimates, based on the results of a previous study [3], $k_{e,r}$ was constrained to be greater than the contribution associated with static conduction through the solid connected matrix. This latter was calculated according to the Lemlich correlation (Eq. (1)).In all the experiments, the effective axial conductivity was found to have a minor effect on the heat transfer properties. Accordingly, in the following the discussion will be focused on the values of the effective radial conductivity and of the wall heat transfer coefficient.

In order to enable a direct comparison of the heat transfer performances of the different packings, the overall heat transfer coefficient, U, was also calculated as shown in Eq. (6) [17].

$$1/U = 1/h_w + D/(6.13k_{e,r}) \quad (6)$$

3. Results

3.1. Random packed-bed

3.1.1. Measured temperature profiles

In Fig. 2, some of the axial temperature profiles measured in the packed bed center-line during experiments with He and N_2 fed at different flow rates and with the thermostatic chamber set at 400 °C are shown as examples. Temperatures are plotted in a dimensionless form, T*(x), calculated according to the following expression:

$$T^*(x) = (T(x) - T(0)) / (T_W(x) - T(0)) \quad (7)$$

which corresponds to the normalized ratio of the actual temperature increase to the total available driving force.

Fig. 2(a) and (b) show that the higher the thermal conductivity of the flowing gas, the faster is its heating rate and the higher its average temperature at a given volumetric flow rate. As a matter of fact, T* at the reactor outlet measured in the test with 15 Nl/min of He is almost twice that measured in the test with 15 Nl/min of N_2. Along similar lines, Fig. 2(a) and (b) show that the higher is the gas flow rate, the lower is the gas heating at given flowing gas.

Notably, the different flowing heat capacities (i.e. the products of the gas flowrates and their molar specific heats) cannot explain the observed differences in the heating profiles of N_2 and He. This is well evident when comparing, for example, the temperature profile measured with 20 Nl/min of N_2 (Fig. 2(a)) with that measured with 30 Nl/min of He (Fig. 2(b)). Regardless the similar flowing heat capacities, the temperature profile on the packed bed axis during the experiment with He is significantly above that obtained by feeding N_2. This clearly proves that the thermal conductivity of the gas phase largely affects the heat transfer in a packed bed of micro-pellets as already reported in the literature [18–20].

3.2. Packed-foams

3.2.1. Measured temperature profiles

In Fig. 3(a) and (b) the axial temperature profiles measured at different radial positions in two packed foams, obtained by loading 0.3 mm Al_2O_3 pellets within the FeCrAlY and the Al-6101 foams, are shown. Data refer to an experiment carried out with 60 Nl/min of N_2 and the thermostatic chamber set at 400 °C. Two main differences are observed when comparing the two figures. First, the average temperature within the bed in the experiment carried out with the Al-6101 foam is much higher than that measured with the FeCrAlY foam. Second, the radial temperature profile within the Al-6101 packed foam is flatter than that obtained within the FeCrAlY packed foam. Considering that the two foams have very similar void fractions and that there are no differences between the two packed-beds filling the foams, observed differences in terms of temperature profiles can be attributed uniquely to the different thermal conductivity of the structured substrate, that in the case of the Al-6101 foam is about one order of magnitude higher than in the case of the FeCrAlY foam. Accordingly, we can conclude that, in line with the results obtained with bare foams [3], the adoption of a highly conductive structured substrate can significantly improve the radial heat transfer rate within a tubular reactor, thus making the radial temperature profile flatter.

In order to better evidence the contributions of the packing to the radial heat transfer of packed foams, in Fig. 4(a),(b) dimensionless temperature profiles measured at the centerline with the Al-6101 packed foam at different N_2 flowrates (15 and 35 Nl/min) are compared with those obtained at equal flowrates with the same foam loaded in the reactor without the packing (BF: bare foam) and with the packed bed (PB). It is evident that the use of the conductive internal significantly improves the heating rate of the flowing gas. Actually also the bare foam exhibits superior heat transfer performances than the packed bed and a synergy is evident when combining foam and particles, the packed foam showing the maximum heating rate. On the other hand when using poorly conductive FeCrAlY foams, the heat transfer performances of the bare foam are comparable to those of the packed bed (Fig. 5(a),(b)). However also in this case the packed foam exhibits a synergetic effect, the gas heating rate being significantly higher than that observed for both BF and PB.

Fig. 6(a) shows the results of heat transfer experiments carried out with the FeCrAlY packed foam fed with He and N_2 at flow rates modulated so as to have a constant flowing heat capacity. A similar plot is presented in Fig. 6(b) for the Al-6101 packed foam. These results evidence that, for both low conductive and highly conductive foams, the higher is the fluid thermal conductivity, the more effective is the radial heat transfer. Indeed, experiments carried out with He grant a faster increase of dimensionless temperature at the tube centerline.

It is worth noticing in this regard that the fluid thermal conductivity is known to affect the wall heat transfer coefficient in the case of bare foams [15,21], as well as the radial effective conductivity and the wall heat transfer coefficient in the case of packed beds [18–20]. Accordingly a quantitative analysis of the experimental data with the heat transfer model is needed to distinguish which of the k_f-dependent parameters are controlling the overall heat transfer rates in the packed foam.

3.2.2. Estimates of heat transfer parameters

3.2.2.1. Effective radial conductivity. In Fig. 7, the estimates of the effective radial conductivity obtained by regression of temperature profiles measured in each experiment carried out with packed FeCrAlY and Al-6101 foams are plotted as a function of the Reynolds number, Re_{dp}, defined using the particle diameter (d_p) as the characteristic length. The contribution to the radial effective conductivity given by the conduction within the structured substrate, estimated according to the Lemlich equation (Eq. (1)), is also plotted for comparison purposes.

The $k_{e,r}$ estimates shown in Fig. 7 clearly evidence that the effective radial conductivity of packed foams strongly depends on the thermal conductivity of the solid phase, the values of $k_{e,r}$ obtained for the highly conductive Al-6101 foam being one order of magnitude higher than those estimated for the less conductive FeCrAlY foam, when tests are performed with N_2 flowing gas. On the contrary, regardless the thermal conductivity of the foam substrate, the effective radial conductivity of packed foams does not significantly depend on flow rate, at least in the investigated range of low Reynolds number (Re_{dp} = 0-25). Along similar lines, a high thermal conductivity of the fluid phase does not bring appreciable benefits to the effective radial conductivity of packed foams made with highly conductive substrates. In fact, the estimates of $k_{e,r}$ in the tests with He are indeed substantially undistinguishable from the estimates in the tests with N_2 in the case of the highly conductive Al-6101 foam. This is not the case of packed foams with a less conductive substrate. Estimates of the effective radial conductivity deriving from tests with the packed FeCrAlY foam, indeed, show that the adoption of a highly conductive gas (He) can double the effective radial conductivity with respect to those obtained in tests with a low conductive gas (N_2).

Interestingly, $k_{e,r}$ predicted by the Lemlich law (Eq. (1)) are well in line with the estimates of $k_{e,r}$ for the highly conductive Al-6101 packed foam and the less conductive FeCrAlY packed foam fed with a low conductive gas. These results unequivocally demonstrate that, exactly like in bare open-cell foams [3], solid conduction within the structured substrate controls the effective radial conductivity of highly conductive packed-foams. Solid conduction is also the dominant contribution to effective radial conductivity in

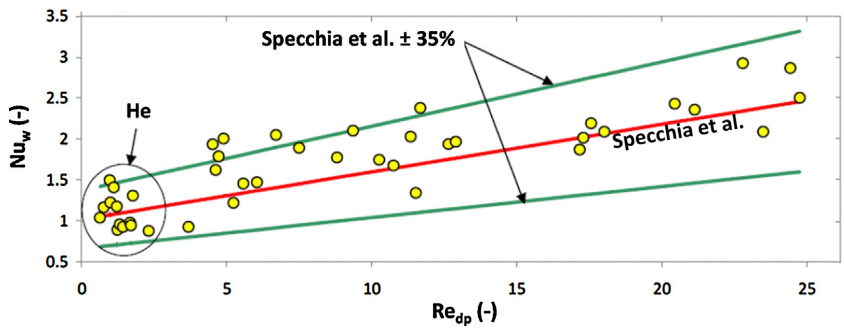

Fig. 9. Nu_w, vs. Re_{dp} for packed foams. Comparison between estimated values and theoretical predictions with the equation for packed beds proposed in Ref. [19].

the case of low conductive foams, fed with a low conductive fluid as N_2. In this case, however, the adoption of a highly conductive fluid can give an appreciable additional contribution to the effective radial conductivity, possibly via mechanisms associated with the flow through the packed particles.

3.2.2.2. Wall heat transfer coefficient. In Fig. 8, the estimates of the wall heat transfer coefficient are plotted against the particle Reynolds number, Re_{dp}. No significant effect of the foam type is observed. On the contrary, a major effect of the gas type is evident, the h_w values obtained in tests with He being close to 1000 $Wm^{-2}K^{-1}$, i.e. much higher than those obtained in N_2 tests, which are always below 400 $Wm^{-2}K^{-1}$, despite Re_{dp} for N_2 is higher than Re_{dp} for He. This is expected since it has been reported that the wall heat transfer coefficient strongly depends on the gas fluid conductivity k_f, for both packed beds [18,19] and bare foams [15,21]. Estimates of h_w from He tests are quite dispersed, possibly due to the relatively low sensitivity of the overall heat transfer resistance on the wall coefficient when using He as flowing gas. On the other hand estimates from N_2 data show a positive trend on increasing Re_{dp} suggesting a possible role of the convective contribution characteristics of packed beds.

To better clarify this point Nusselt numbers, $Nu_{w,dp}$, defined with the particle size diameter as the characteristic length, have been calculated from h_w estimates and compared with the predictions of the correlation proposed by Specchia and coworkers for packed bed of pellets [19]:

$$Nu_{w,dp} = h_w d_p/k_f = 2\varepsilon_{PB} + (1-\varepsilon_{PB})\Big/\left(0.0024\left(D/dp\right)^{1.58} + k_f/3k_s\right) + 0.0835 Re_{dp}^{0.91} \quad (8)$$

As shown in Fig. 9 our estimates well match the predictions of the literature correlation, in fact most of our points deviate less than ±35% from the latter. This suggests that packed foams behave as packed bed of pellets at the bed-wall boundary, taking advantage of the contribution of local convective mixing to reduce the heat transfer resistance at the wall.

Such a result may have a dramatic impact on the design of conductive foams as internals for catalytic reactors, because it suggests that the heat transfer in packed foams would not be limited by the lack of intimate contact or the close proximity of the foam with the tube wall, which on the contrary has been shown to have a major effect on the performances of bare foams [21–23].

The above results can be summarized by the simplified scheme in Fig. 10 depicting the heat transfer in packed foams according to an electrical analogy. Here, the overall resistance between the centerline and the wall is schematically represented as an in series combination of in parallel resistances, namely the internal resistance of the bare foam in parallel with the bulk resistance of the packed bed (R^{BF}_{bulk} and R^{PB}_{bulk}), followed by the wall resistance of

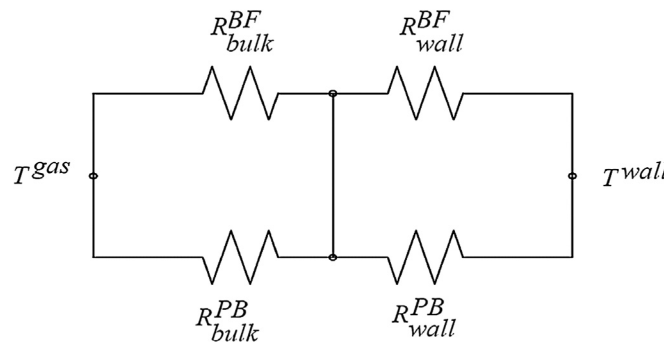

Fig. 10. Circuit of equivalent resistances used to describe the heat transfer mechanisms within packed foams.

the bare foam in parallel with that of the packed bed (R^{BF}_{wall} and R^{PB}_{wall}). Of course, the real situation is more complex, as each one of such resistances actually summarizes a number of contributions from different transport mechanisms, including e.g. convective heat transfer, conduction through the contact between particles and between particles and foam struts, and radiation in the case of R^{BF}_{bulk}. As previously discussed, at the low Reynolds numbers (Re_{dp} and Re_{dc}) herein investigated highly conductive foam internals provide a preferential path to radial heat dissipation via the connected solid structure of the foam $\left(R^{BF}_{bulk} \ll R^{PB}_{bulk}\right)$. On the other hand, at the boundary between bed and tube the local convective mixing and the distributed contact associated with the small packed particles may guarantee a low-resistance heat transport path even in the presence of a loose contact between foam and tube $\left(R^{PB}_{wall} \ll R^{BF}_{wall}\right)$.

3.3. Comparison of heat transfer in packed-foams, packed-beds and open-cell foams

To complete the assessment of the heat transfer performances of packed-foams we have compared the performances of this packing with those of more conventional packed-beds and open-cell foams. To this scope the overall heat transfer coefficients, U, were calculated for each bed configuration using Eq. (6), considering both FeCrAlY and Al-6101 foams, and estimates of $k_{e,r}$ and h_w obtained with both N_2 and He flows. Results are reported in Fig. 11(a) and (b), where lines connect groups of points obtained at different flow rates and oven temperatures with the three packing configurations (PB, BF and PF) in the case of FeCrAlY (Fig. 11(a)) and Al-6101 (Fig. 11(b)) internals.

At the relatively low flow rates herein investigated, which correspond to very low Re_{dp} due to the small size of the particles used for packing purposes, heat transfer in packed beds is rather poor with overall heat transfer coefficients of about 50 $Wm^{-2}K^{-1}$ and 100 $Wm^{-2}K^{-1}$ obtained when using N_2 and He, respectively. Insertion of the poorly conductive FeCrAlY foam (Fig. 11(a)), which

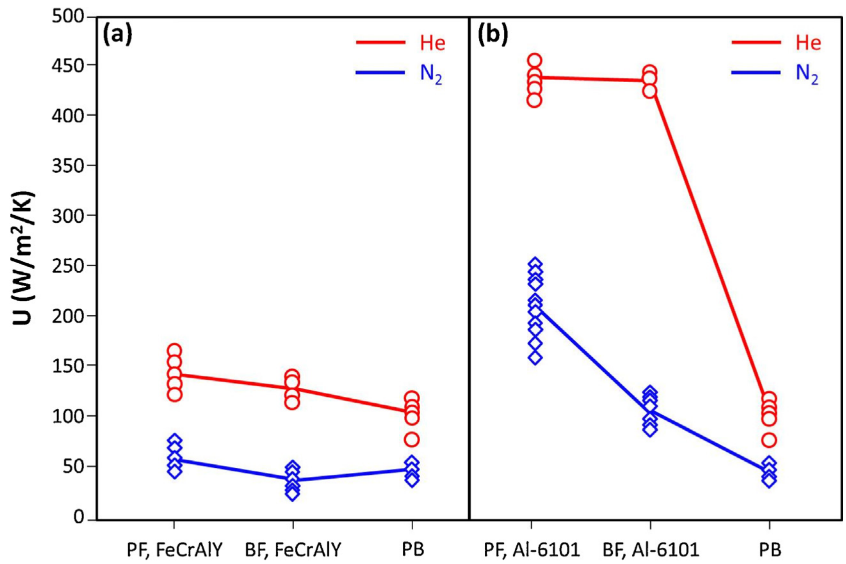

Fig. 11. Heat transfer performances of different packings: packed foams vs bare foams vs packed bed. (a) FeCrAlY foam, (b) Al-6101 foam. Values of U for bare foams have been calculated starting from data reported in Refs. [3,15].

in the bare configuration is characterized by U values comparable with those of packed bed, results in a small improvement in the packed foam configuration. On the other hand the use of highly conductive Al-6101 foam internals greatly enhances the heat transfer rate (Fig. 11(b)). Just by using the Al bare foams overall heat transfer coefficients of about 100 $Wm^{-2} K^{-1}$ and 400 $Wm^{-2} K^{-1}$ are obtained when flowing N_2 and He, respectively. The packed foam configuration allows for a further improvement, especially when using the low conductive gas (N_2). In this case the dominant resistance is at the foam-wall interface, and it is markedly reduced by the presence of the packed particles resulting in an overall heat transfer coefficient of 200 $Wm^{-2} K^{-1}$. A smaller but significant effect is also evident when using He. In this case the wall heat transfer coefficient is still increased by using the packed foam configuration, but its impact on the overall heat transfer coefficient is lower due to the high conductivity of He, which guarantees low resistance at the wall also in the case of bare foams.

4. Conclusions

Highly conductive packed-foams have been recently claimed by our group as an innovative solution to increase the catalyst inventory in structured tubular reactors, while granting at the same time enhanced heat transfer performances within the reactor. This is of particular interest in view of intensifying non-adiabatic reactors running reactions under total or partial kinetic control, which often require catalyst inventories much higher than those feasible with washcoated open-cell foams. Indeed the catalyst amount which can be packed in an open-cell foam is much greater than the amount which can be loaded by washcoating the same foam. In addition, the adoption of packed-foams instead of washcoated foams would intrinsically solve major issues related to catalyst loading and unloading in the reactor, as well as the replacement of the spent active phase. Last but not least, when adopted as retrofitting technology, packed foams would allow to exploit the same pelletized catalysts already operating (possibly modified in their size), which are often the result of long and costly development work, and which have properties well-tailored to the specific process needs.

In this work we have preliminarily assessed the heat transfer properties of conductive packed-foams loaded with micro-pellets with the appropriate size. By comparing the performances of packed-foams with those of the corresponding bare foams and the corresponding packed-bed tested at the same process conditions, we have demonstrated that packed-foams can synergically combine the different heat transfer mechanisms of packed bed of pellets and highly conductive open cell foams. This allows to exploit the most effective heat transfer mechanism available, i.e. conduction within the highly conductive structured substrate in the bulk of the bed and local convective mixing of the packed bed at the boundary between bed and tube wall. According to this rationale, the overall heat transfer coefficients largely exceeding those reported in this work can be achieved by an optimized design of the packed bed and metallic foam system, including e.g. the adoption of conductive foam matrices with greater relative densities and their operation at higher Reynolds numbers.

Notably, the presence of small pellets packed into a structured foam results in increased pressure drops, which may become unsustainable in the case of long reactor tubes. This can be particularly critical if fouling or carbon deposition occur under reaction conditions. For this reason, packed foam reactors are particularly interesting for compact scale applications (i.e. short tubes) where process intensification means finding the best trade-off between catalyst hold-up, heat transfer and pressure drop.

Acknowledgments

The authors gratefully acknowledge funding by the Italian Ministry of Education, University and Research, Rome (MIUR, Progetti di Ricerca Scientifica di Rilevante Interesse Nazionale, prot. 2010XFT2BB) within the project IFOAMS ("Intensification of Catalytic Processes for Clean Energy, Low-Emission Transport and Sustainable Chemistry using Open-Cell Foams as Novel Advanced Structured Materials").

References

[1] E. Tronconi, G. Groppi, C.G. Visconti, Curr. Opin. Chem. Eng. 5 (2014) 55–67.
[2] J. Gascon, J.R. van Ommen, J.A. Moulijn, F. Kapteijn, Catal. Sci. Technol. 5 (2015) 807–817.
[3] E. Bianchi, T. Heidig, C.G. Visconti, G. Groppi, H. Freund, E. Tronconi, Chem. Eng. J. 198–199 (2012) 512–528.
[4] P. Mülheims, B. Kraushaar-Czarnetzki, *2011*, Ind. Eng. Chem. Res. 50 (*2011*) 9925–9935.
[5] I. Gräf, A.K. Rühl, B. Kraushaar-Czarnetzki, Chem. Eng. J. 244 (2014) 234–242.

[6] Y. Li, Q. Zhang, R. Chai, G. Zhao, Y. Liu, Y. Lu, F. Cao, AIChE J. 61 (2015) 4323–4331.
[7] A. Montebelli, C.G. Visconti, G. Groppi, E. Tronconi, S. Kohler, Chem. Eng. J. 255 (2014) 257–265.
[8] D. Vervloet, F. Kapteijn, J. Nijenhuis, J.R. van Ommen, Catal. Today 216 (2013) 111–116.
[9] C.G. Visconti, E. Tronconi, G. Groppi, L. Lietti, M. Iovane, S. Rossini, R. Zennaro, Chem. Eng. J. 171 (2011) 1294–1307.
[10] C.G. Visconti, E. Tronconi, L. Lietti, G. Groppi, P. Forzatti, C. Cristiani, R. Zennaro, S. Rossini, Appl. Catal. A: Gen. 370 (2009) 93–101.
[11] V. Meille, App. Catal. A: Gen. 315 (2006) 1–17.
[12] A. Montebelli, C.G. Visconti, G. Groppi, E. Tronconi, C. Cristiani, C. Ferreira, S. Kohler, Catal. Sci. Technol. 4 (2014) 2846–2870.
[13] M. Iovane, R. Zennaro, G. Groppi, L. Lietti, E. Tronconi, C.G. Visconti, S. Rossini, E. Mignone, inventors; Eni S.p.A., applicant. Reactor for exothermic and endothermic catalytic reactions. Patent application WO/2010/130399. 2010 May 10.
[14] G. Groppi, E. Tronconi, C.G. Visconti, R. Zennaro, A. Tasso, inventors; Eni S.p.A., applicant. Packed-bed tubular reactor for heterogeneous exothermic or endothermic catalytic reactions. Patent application WO/2015/033266. 2014 September 2.
[15] E. Bianchi, T. Heidig, C.G. Visconti, G. Groppi, H. Freund, E. Tronconi, Catal. Today 216 (2013) 121–134.
[16] R. Lemlich, J. Colloid Interf. Sci. 64 (1978) 107–110.
[17] J.E. Crider, A.S. Foss, AIChE J. 11 (1965) 1012–1019.
[18] C.H. Li, B.A. Finlayson, Chem. Eng. Sci. 32 (1977) 1055–1066.
[19] V. Specchia, G. Baldi, S. Sicardi, Chem. Eng. Comm. 4 (1980) 361–380.
[20] A.G. Dixon, AIChE J. 31 (1985) 826–834.
[21] E. Bianchi, G. Groppi, W. Schwieger, E. Tronconi, H. Freund, Chem. Eng. J. 264 (2015) 268–279.
[22] P. Mülheims, S. Marz, S.P. Müller, B. Kraushaar-Czarnetzki, Chem. Ing. Tech. 83 (2011) 286–294.
[23] C. Hutter, D. Büchi, V. Zuber, Ph. Rudolf von rohr, Chem. Eng. Sci. 66 (2011) 3806–3814.