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Comparative Analysis of Heat Transport in SiC Solid Foam and Al_2O_3 Granular Packings for Fixed-bed Reactors

Luca Turchetti[1], Giulia Monteleone[2], Maria Cristina Annesini[1]

[1]Department of Chemical Engineering, Materials & Environment
University of Rome "La Sapienza"
Via Eudossiana 18 - 00184 Rome, Italy
luca.turchetti@uniroma1.it

[2] ENEA, Italian Agency for New Technologies, Energy and Environment, Rome, Italy

The thermal features of different catalytic supports for fixed-bed reactors, namely a SiC open-cell solid foam and three types of alumina beads, were investigated experimentally. The results were analysed by means of a stationary one-equation bi-dimensional model and show that SiC foam packings are characterized by better radial heat transport because of their higher intrinsic thermal conductivity. However, from a structural point of view, the granular beds appear preferable in order to favor radial heat transport, mainly because of the lower void fraction and higher radial mixing of the gas phase.

1. Introduction

Conventional granular packings used in catalytic fixed-bed reactors suffer from heat transport limitations that may cause the formation of important temperature gradients in the reactor bed. Strong temperature inhomogeneities in the catalytic bed may become a major issue for fixed-bed reactors: the formation of hot spots may cause catalyst deactivation and/or safety issues, while, on the other hand, the presence of cold spots may lead to an incomplete exploitation of the catalyst loaded in the reactor. These issues are likely to be important especially in the case of strongly exothermic/endothermic processes carried out in externally cooled/heated reactors.

In the last decade, open-cell solid foams have received an increasing interest as a possible alternative to granular packings, because these systems possess an extremely interesting combination of desirable properties for a catalyst support, such as low resistance to fluid flow and high specific interfacial area. Furthermore, due to the monolithic structure, heat transport in solid foams is not affected by contact thermal resistances, as in the case of the discontinuous solid matrix of a granular bed; for this reason, the use of solid foam packings could enhance heat transport inside the reactor bed.

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In this work, the thermal features of a 20 ppi SiC foam packing are compared to those of three conventional beds of Alumina beads (3 mm and 5 mm square cylinders; 5 mm spheres). To that end, experimental tests were carried out in a laboratory scale, externally heated fixed-bed reactor. The reactor was fed with cold instrument air; different combinations of reactor packing, wall temperature and air flow rate were tested. In each test, the temperature inside the reactor was monitored at different axial and radial positions and stationary temperature profiles were collected.

The experimental data were analyzed with a stationary one-equation, two-dimensional model accounting for different heat transport mechanisms in the solid and fluid phases. The thermal contact resistance between the packing and the internal reactor wall was also considered.

The experimental results showed that, under the same operating conditions, the SiC foam packing is characterized by flatter radial temperature profiles and higher temperatures at the reactor axis than Al_2O_3 beds. On the other hand, the SiC foam packing also exhibited larger temperature drops in correspondence with the internal wall of the reactor. This finding shows that the monolithic SiC packing has a worse thermal contact with the reactor wall, but, in contrast, is characterized by a more efficient radial heat transport. A further analysis of the data by means of the mathematical model considered in this paper shown that the higher radial heat transport efficiency observed for the SiC packing was entirely due to its remarkably higher intrinsic thermal conductivity (more than 30 times higher than the Al_2O_3 beads), while, under a strictly structural point of view, the granular beds appear to be preferable to favor radial heat transport, mainly because of the lower void fraction and higher radial mixing of the gas phase.

2. Materials and Methods

2.1 Experimental apparatus

Experimental tests were performed in a stainless steel tubular reactor with an internal length L = 184.6 mm and radius R = 24.5 mm. The reactor was externally heated by means of 3 band heaters, each with a maximum power of 400 W, evenly spaced along the reactor length. A plate thermocouple was placed between the reactor external wall and each band heater for temperature control. The reactor was provided with four lateral access points, at a distance of 41 mm from each other, for the insertion of multi-point thermocouples. Each multi-point thermocouple provided for a temperature measurement at three different radial positions: at the reactor axis, close to the reactor internal wall and in an intermediate position. Two further thermocouples were placed close to the reactor inlet and outlet, in order to measure the gas temperature at these points.

2.2 Reactor packings

In experiments on granular catalyst supports, the reactor was packed with alumina particles of three types: square cylinders of 3 and 5 mm diameter and spheres of 5 mm diameter, with a void fraction (experimentally determined) of 33%, 38% and 41%, respectively.

In experiments on monolithic beds, the reactor was packed with 3 cylindrical samples of SSiC foam (Fraunhofer Institute for Ceramic Technologies and Systems IKTS, Dresden, Germany) with an average nominal pore density of 20 ppi, external void fraction of 0.91 and hollow struts with an internal void fraction of 30%. Each sample had an average diameter of 48 mm and a height of 54 mm.

2.3 Experimental runs

In each experimental run, instrument air was fed to the reactor with a prescribed mass flow rate F (5 Nm^3/h, 10 Nm^3/h and 15 Nm^3/h, controlled with a mass flow controller) and the reactor wall temperature was set to a different temperature T_w (150°C, 200°C and 400°C). Therefore, a total of 36 experimental runs, 9 on each packing type, were carried out.

After each run was started, the time course of the temperatures was monitored until a stationary codition was attained and the corresponding values were recorded.

2.4 Numerical procedures

Numerical integration of differential equations was performed with a commercial software package (gPROMS, Process System Enterprise, London, UK) by using a first-order backward finite difference method. The same software was used for the parameter estimation procedures.

3. Mathematical Model

A preliminary order-of-magnitude analysis showed that, under the operating conditions used in the experimental tests, the solid and gas temperature do not differ significantly, so that local thermal equilibrium can be assumed. This analysis was based on estimation of the solid-gas heat exchange coefficient by using the correlation proposed by Li and Finlayson (1977) for alumina pellets and Giani et al. (2005) for solid foams.

Therefore, the experimental data have been analysed with a stationary two-dimensional, one-equation model. The model relies on the following main assumptions: 1) the solid phase is isotropic; 2) the dominant axial heat transport mechanisms are conduction in the solid phase and convection in the gas phase (axial dispersion in the gas phase is neglected); 3) molecular conduction in the gas is negligible.

Under these hypotheses, the energy balance in the reactor can be written as

$$-Gc_f\frac{\partial T}{\partial z}+k_{es}\frac{\partial^2 T}{\partial z^2}+(k_{es}+k_d)\left(\frac{\partial^2 T}{\partial r^2}+\frac{1}{r}\frac{\partial T}{\partial r}\right)=0 \tag{1}$$

where r and z are the radial and axial variable, respectively, G is the gas mass flux, c_f the gas heat capacity, k_d the gas radial thermal dispersion coefficient and k_{es} the effective conductivity of the solid phase. Equation (1) can be integrated with the following boundary conditions

$$T\big|_{z=0} = T_{in} \quad ; \quad \frac{\partial T}{\partial z}\bigg|_{z=L} = 0 \quad ; \quad \frac{\partial T}{\partial r}\bigg|_{r=0} = 0 \quad ; \quad h_w(T_w - T\big|_{r=R}) = (k_{es} + k_d)\frac{\partial T}{\partial r}\bigg|_{r=R} \tag{2}$$

where T_{in} is the gas inlet temperature, and h_w the wall heat transfer coefficient.

3.1 Model parameters

All physical properties in Eq. (1) e (2) were assumed as uniform throughout the reactor and calculated at the average temperature between reactor inlet and outlet conditions (as experimentally determined).

For alumina beads, the effective solid thermal conductivity and the dispersion coefficient of the gas phase were calculated as proposed by Yagi and Kunii (1957), by assuming an intrinsic thermal conductivity for the beads of 3 W $m^{-1}K^{-1}$.

Different correlations can be found in the literature for the estimation of the effective thermal conductivity of SiC foams (Calmidi and Mahajan, 1999; Bhattacharya et al., 2002). For a 91% void fraction foam, such correlations predict an effective-to-intrinsic conductivity ratio that lies approximately between 2.9 and 3.1 %; here, an average value of 3% was assumed. Moreover, a further reduction of 30% was considered to account for the hollow structure of foam struts. A value of 100 W $m^{-1}K^{-1}$ was assumed for the intrinsic thermal conductivity of SiC struts (Fend et al., 2004). Thermal dispersion in the gas phase was neglected for solid foam beds, because a rough estimation of k_d for this type of packing (Calmidi and Mahajan, 2000) gives a value that is two orders of magnitude smaller than k_{es} .

The wall heat transfer coefficient was used as adjustable parameter to fit the mathematical model to the experimental data, both for granular and monolithic beds. It is worth noting that, for granular beds, correlations for this parameter are available in literature; such correlations, however, predict significantly different h_w values in the range of experimental conditions considered in this work. Correlations for the calculation of h_w for solid foams are presently not available in the literature.

4. Results

As an example of the results obtained, the steady state temperature profiles recorded in two experimental runs are presented in Fig. 1a and 1b as points in T Vs. r plots (different sets of points correspond to different axial positions). Fig. 1a and 1b report data obtained with SiC foams and 3mm Al_2O_3 cylinders, respectively, with the same operating conditions, namely F=5 Nm^3/h and T_w=200°C.

By comparing Fig. 1a and 1b, three important features (that have been observed in all experiments carried out with the same operating conditions) can be remarked: 1) the radial temperature gradient is higher for granular packings than for solid foams; 2) the difference T_w-$T|_{r=R}$ is smaller for granular packings than for solid foams; 3) the temperature at the reactor axis is higher for SiC foams.

Table 1: Optimal values of the wall heat transfer coefficient

T_w [°C]	F [Nm^3/h]	h_w [W m^{-2} K^{-1}]			
		SiC foam	Al_2O_3 cyl. 3mm	Al_2O_3 cyl. 5mm	Al_2O_3 sph. 5mm
150	5	32.0	62.5	53.0	66.0
150	10	48.0	79.0	75.5	85.0
150	15	62.5	92.0	84.5	100.0
200	5	38.5	63.0	64.0	65.0
200	10	52.5	88.0	81.0	82.0
200	15	64.5	92.0	101.0	124.0
400	5	43.0	63.5	70.0	72.0
400	10	64.0	105.0	94.0	110.0
400	15	76.5	111.0	103.0	120.0

5. Discussion

The higher radial temperature gradient observed for alumina packings (when compared to SiC foams) is consistent with their lower $k_{es}+k_d$ value and shows that this type of solid bed is characterized by worse radial heat transport. A further analysis shows that the better performance of SiC foams is entirely due to their intrinsic conductivity, which is one order of magnitude greater than the intrinsic conductivity of alumina beads. This conclusion can be supported by using the correlations proposed by Yagi and Kunii (1957) to estimate the sum $k_{es}+k_d$ for a granular SiC bed with the same geometrical properties of the Al_2O_3 packings used in this work: indeed, the predicted value exceeds that of SiC foams.

The lower difference $T_w-T|_{r=R}$ observed in experiments with alumina packings suggests that this type of solid bed has a better thermal contact with the internal reactor wall. This finding is more than reasonable, considering that incoherent granular beds can adapt their shape and better fill the space in proximity of the reactor wall.

For each experiment performed, the model presented in section 3 was fit to the experimental data by using h_w as adjustable parameter. The optimal values found for h_w are reported in Tab.1. The temperature profiles calculated by the model with the optimal h_w values are reported as lines in Fig. 1a and 1b. The agreement between the model and the experimental data is satisfactory. By comparing the values of h_w reported in Tab.1, the better thermal contact with the internal reactor wall, corresponding to a higher h_w, is confirmed.

In conclusion, among the catalyst supports considered in this work, SiC foams showed the best performance, because they allowed to obtain the most uniform temperature profiles and highest temperatures at the reactor axis. These results are due to the much higher intrinsic thermal conductivity of SiC foams, while, under a strictly structural point of view, granular packing would be preferable.

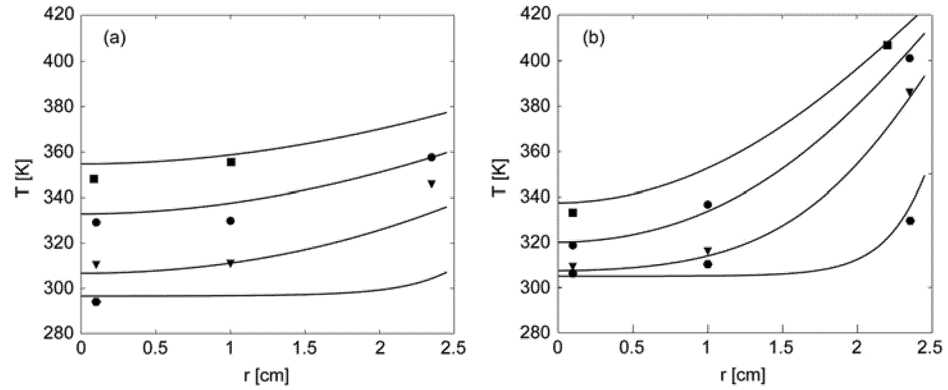

Figure 1.: Experimental (points) and calculated (lines) temperature profiles obtained for F=5Nm^3/h and T$_w$=200°C with two different reactor packings: SiC foams (a) and 3mm Al$_2$O$_3$ cylinders (b) .

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