Chemical Engineering Science 60 (2005) 1823–1835

Chemical Engineering Science

www.elsevier.com/locate/ces

Heat transfer in conductive monolith structures

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Received 27 April 2004; received in revised form 4 November 2004; accepted 29 November 2004
Available online 21 January 2005

Abstract

Honeycomb structures made of highly thermal conductive materials (e.g. certain metals) have been proposed as attractive catalyst supports with enhanced heat transfer properties. Recently prototypes of such materials have become available, enabling experimental investigations on their heat transfer properties, including packaging into tubes. This work is focused on the heat transfer performance of conductive monolith structures packaged into heat exchanger tubes. Several parameters such as monolith material structure and properties and packaging tolerances are investigated. Heat transfer coefficients on the order of $1000\,W/m^2\,K$ and higher were measured. The results are analyzed applying a detailed model based on a fundamental understanding of the relevant phenomena. It is demonstrated that it is necessary to consider the variations of the thermal expansion of the monolith and the tube over the length of the monoliths. By including thermal expansion, the model is in excellent agreement with the experimental results without the need of any fitting parameter. The results are used to develop some design guidelines. In addition some implications of the heat transfer performance for the relevant applications in multitubular reactors as well as some new potential application areas are discussed.

Keywords: Monolith; Honeycomb; Heat conduction; Heat transfer; Modeling; Catalyst support

1. Introduction

Recently, monolithic structures made out of materials with a high thermal conductivity have been proposed as an attractive alternative to conventional catalyst supports for the use in highly exothermic reactions (e.g. partial oxidations) employing multitubular reactors (Groppi and Tronconi, 2000, 2001; Boger and Menegola, 2005; Carmello et al., 2000; Tronconi et al., 2004). Significantly higher heat transfer rates compared to random catalyst packings (e.g. spheres or rings) were predicted, by changing the dominant heat transfer process from convection to conduction. Simulations (Groppi and Tronconi, 2000, 2001; Boger and Menegola, 2005) as well as experiments (Carmello et al., 2000; Tronconi et al., 2004) for highly exothermic reaction revealed significantly lower hot spot temperatures. This resulted in improved operating characteristics and the potential for significant economic benefits. A method and process to prepare suitable extruded metal monoliths with a high thermal conductivity was recently developed by Corning Incorporated (Cutler et al., 2001). A picture of a monolith made for example from copper is shown in Fig. 1. These copper monolithic catalyst supports were evaluated for a heterogeneously catalyzed CO oxidation reaction and showed no radial temperature gradients even at very high reactive heat loads (Tronconi et al., 2004), demonstrating the excellent radial heat removal due to the thermal conduction of the structure. The heat transfer from the monolith to the reactor resulting from the packaging was identified as bottleneck.

So far most of the investigations of the heat transfer in these conductive monolith structures was of theoretical nature and limited to the heat transport within the structures itself. To our knowledge, no detailed experimental and theoretical evaluation of the heat transfer of such monolith structures packaged into reactor or heat exchanger tubes is available. As the resistance due to the packaging was identified

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doi:10.1016/j.ces.2004.11.031

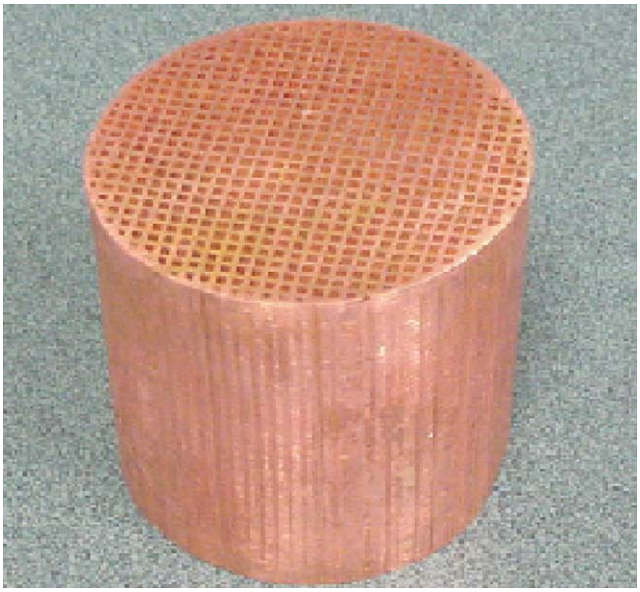

Fig. 1. Photo of copper based monolithic catalyst support with high thermal conductivity.

as the main bottleneck (Tronconi et al., 2004) the objective of the present paper is to further elucidate the effect and improve the overall performance. The results are based on an extensive experimental study combined with a detailed theoretical analysis. The extremely high heat transfer coefficients as well as the favorable ratio between heat transfer and pressure drop leads to additional areas for the use of such monolithic structures beyond the applications discussed so far (Groppi and Tronconi, 2000, 2001; Boger and Menegola, 2005; Carmello et al., 2000; Tronconi et al., 2004). Some of them will be addressed in the discussion section.

2. Theoretical background

In Fig. 2 a schematic of the cross-section of a monolith inserted into a tube is shown. The monolith is represented by a number of channels that are separated from each other by the channel walls of the monolith. No radial exchange of gas between the channels is feasible and therefore, no convective heat transfer in this direction occurs. On the other hand the walls in these extruded structures are connected throughout the entire diameter. Therefore high heat transfer rates can be achieved. Monolith structures as shown in Figs. 1 and 2 with square channels experience some anisotropy with respect to the radial heat flow. However, this is a second order effect and therefore is usually negligible. The rate of the conductive heat transfer is a function of the thermal conductivity of the wall material and of the solid fraction. Groppi and Tronconi (1996) developed a correlation to calculate the effective radial conductivity $k_{S,R}$ in such monolith structures based on geometric data (the void fraction ε) and the thermal conductivity k_S of the monolith walls.

$$k_{S,R} = k_S\left((1-\sqrt{\varepsilon}) + \frac{\sqrt{\varepsilon}}{(1-\sqrt{\varepsilon}+\frac{k_G}{k_S}\sqrt{\varepsilon})}\right)^{-1}. \quad (1)$$

Fig. 2 also shows in a qualitative manner some typical temperature profiles inside the monolith (see item 1). Additionally the temperature profiles of the gas inside the channels in case of an operation in which a hot gas is cooled (dashed lines, item 5) are illustrated. For most applications the flow inside the channels is laminar since the hydraulic diameter is small (in the order of a 1–3 mm). Because of the small dimensions compared to the monolith diameter the thermal exchange between gas and channel wall is usually described by a film model and using a heat transfer coefficient h_{GS} and the according Nusselt number Nu_c. Suitable engineering correlations for the Nusselt number inside monolith channels can be readily obtained from the literature (Vortruba et al., 1975) based on experimental investigations, including inlet and outlet effects as well as the thermally and hydraulically developing profiles.

$$h_{GS} = \frac{Nu_c\, k_G}{d_{h,c}}, \quad (2)$$

$$Nu_c = 0.571\left(Re_c\,\frac{d_{h,c}}{L}\right)^{2/3}. \quad (3)$$

In case of inserting the monoliths into a heat exchanger (or reactor) tube the heat needs to be transferred across the gap between the skin of the monolith and the inner surface of the tube (item 2 in Fig. 2). This heat transfer resistance is characterized by the size of the gap δ_{gap} and by the thermal conduction through the usually stagnant fluid film that fills this gap. Expressing the inverse resistance in form of a heat transfer coefficient we obtain $h_{gap} = \delta_{gap}/k_G$. In reality the geometric imperfections of monolith and tube as well as the not perfect centering of the monolith in the tube will result in gap size variations along the circumference of the tube. Therefore the gap size defined above should be considered as a kind of mean effective gap size.

In the tube itself the radial heat conduction occurs (step 3 in Fig. 2), which is dependent on the thickness of the tube and its thermal conductivity. The heat transfer between the outside surface of the tube and the external heat exchange medium (step 4 in Fig. 2) depends on the underlying hydrodynamics and the physical properties of the heat exchange medium. This heat transfer is usually described by means of a heat transfer coefficient h_{ext} and the according Nusselt number. Reasonable engineering correlations can be found in the literature (Jones, 2002).

$$Nu_{ext} = \frac{h_{ext} d_{h,ext}}{k_{HE}} = c\, Re_{ext}^{b}\, Pr_{ext}^{0.33}. \quad (4)$$

With $c=0.613$ and $b=0.47$ for $Re_{ext} < 1000$ and $c=0.384$ and $b=0.54$ for $Re_{ext} > 1000$. All the dimensionless groups