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Novel catalyst structures with enhanced heat transfer characteristics

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ABSTRACT

Highly exothermic and highly endothermic reactions require catalyst beds with good heat transfer characteristics. A novel catalyst structure, microfibrous entrapped catalyst (MFEC) structure, made of high thermal conductive metals can significantly improve heat transfer efficiency, compared with traditional packed beds (PB). First, the thermal parameters of metal MFEC were determined experimentally. In a stagnant gas, the radial effective thermal conductivity of Cu MFEC was 56-fold of that of alumina PB, while the inside wall heat transfer coefficient was 10 times of that of alumina PB. Compared to PB, even those made of pure copper particles, conductive metal MFEC also provides much more effective thermal conductivity and higher inside wall heat transfer coefficient in a flowing gas testing. In addition, an application of Cu MFEC in Fischer–Tropsch synthesis (FTS) demonstrated an improvement in temperature distribution inside the catalyst bed and an increase in product selectivity. Furthermore, unlike monolith catalyst structures and metallic foams, the MFEC structure is compatible with pre-manufactured catalyst particles, very flexible and ease to be corrugated. Contrast to corrugated packing with a poor conductive contribution to heat transport, MFEC with a good self-dependent thermal conductivity does not require the recycle of gas or liquid to increase the convective term of heat transfer. Therefore, the conductive metal MFEC structures serve as a great catalyst structure to enhance the intra-bed heat transfer for highly exothermic or highly endothermic reactions, reducing temperature excursions in the reactors.

1. Introduction

Because of the poor effective thermal conductivity of typical catalyst beds, heat transfer imposes a size limitation on the reactors for highly exothermic and highly endothermic heterogeneous reactions, which generate or require a large amount of heat on the surface of catalytic particles. For example, for Fischer–Tropsch synthesis (FTS), which has a reaction heat of −165 kJ/mol of CH_2 [1] and an adiabatic temperature rise of 1600 °C [2], Van Vuuren [3] summarized that the maximum diameter for tubular fixed beds with catalyst granules is 80 mm. Increasingly, endeavor to enhance heat transfer inside the reactor is made for those highly exothermic and highly endothermic reactions/processes. Generally, fluidized reactors [4,5], slurry reactors [6,7], metal monolith catalyst structures [8,9], metallic foams, and corrugated packing with open/close cross flow structure [10,11] are used to improve heat transfer efficiency inside the reactor. Though some successful applications of those methods experimentally and industrially exist, they still carry some disadvantages. For instance, the catalyst density of fluidized reactors and slurry reactors is relatively low [12]. Monolith reactor [13,14] structures and metallic foams [15] need a washcoating process to load catalytic component, which is not suitable for pre-manufactured catalysts. Corrugated packing, compatible with both washcoating and pre-manufactured catalyst particles, has been proven to have a poor conductive contribution to heat transport [16] so that a gas or liquid recycle is usually applied to improve the convective component to achieve an enhanced intra-bed heat transfer. In this paper, based on the study of thermal parameter measurements and an application in FTS process, microfibrous entrapped catalyst (MFEC), a novel enhanced heat transfer catalyst structure, is introduced to provide an alternative key to solve these problems.

The MFEC structure was developed by Auburn University and is now commercially available at IntraMicron Inc., AL. As shown in Fig. 1, MFEC is a microstructured catalyst made of sintered micron-sized metal, glass, or polymer fibers with small catalyst particles entrapped inside [17–20]. It was found that MFEC demonstrates high void volume and acceptably uniform particle distribution in the media. This high void volume significantly reduces pressure drop compared to packed beds of similar-size particles. Intra-particle mass transfer and heat transfer are enhanced due to the presence of small particles in this material versus typical extrudates used in industrial fixed bed reactors. Ultra-high contact efficiency results from using small particles entrapped in a sintered fiber matrix [21]. Several investigations have been carried out to improve the understanding of the functions of microfibrous media.

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Nomenclature

C_p	heat capacity (J/kg K)
D	diameter (m)
h	inside heat transfer coefficient (W/m^2 K)
k	thermal conductivity (W/m K)
L	length of the bed
Pr	Prandtl number
r	radial position (m)
Re	Reynolds number
S	surface area (m^2)
t	time (s)
T	temperature (K)
v	superficial velocity (m/s)
V	volume (m^3)
x	mass fraction
y	volume fraction
z	axial position (m)

Greek symbols

ε	voidage
μ	gas viscosity (kg/m s)
ρ	density (kg/m^3)
φ	shape factor

Subscripts

Al	alumina
e	effective
f	fluid
i	*i*th particle
m	metal
p	particle
r	radial
s	solid
w	wall
z	axial

Kalluri et al. [23] studied the effects of microfibrous media on mitigating bed channeling. Yang et al. [24] and Duggirala et al. [25] investigated the effects on external mass transfer in desulfurization by both experiments and CFD modeling. Zhu et al. [26] studied the electrical conductivity of the metal microfibrous sheet in fuel cell. Ryan Sothen [27] discussed MFEC's pressure drop and effective removal of harmful airborne contaminants in air filtration systems.

However, the thermal property of this structure has not been addressed before. Like metal monolith structures, MFEC can be made of highly conductive metals, such as copper, brass, or nickel, to improve the intra-bed heat transfer efficiency in a fixed bed reactor. Such a reactor with conductive metal MFEC may be able to avoid hot or cold spots in the catalyst bed and achieve a uniform temperature profile or fine temperature control. Therefore, there is great potential to apply MFEC for highly exothermic and highly endothermic reactions/processes, especially those having narrow operational temperature windows due to product selectivity requirements and catalyst deactivation issues. To study these applications, the thermal parameters of MFEC are critical and need more research effort to understand them.

The objective of this paper is to give an experimental study of effective thermal conductivity and inside wall heat transfer coefficient for copper, nickel, and stainless steel (SS) MFECs. To understand the improvement of thermal conductivity from sintered fiber structure, a Cu MFEC sample was compared with packed beds (PBs) made of copper and alumina extrudates. Both transient and steady-state measurements were carried out on seven samples. One-phase pseudo-homogeneous approaches were used to analyze radial effective thermal conductivity, axial effective thermal conductivity, and wall heat transfer coefficient for metal MFECs and PBs. Furthermore, Cu MFEC entrapping with 15%Co/Al_2O_3 catalyst particles was employed to study the application of MFEC in FTS reaction. The temperature distribution and product selectivity of Cu MFEC were compared with that of PB catalyst.

2. Materials and methods

2.1. Preparation of MFEC

A wet-lay method to prepare MFEC based on traditional high-speed and low-cost paper-making techniques was developed by Auburn University. The detailed process can be found in Refs. [19–22]. Recently, a new method had been developed by them to prepare the MFEC for pre-manufactured catalyst particles (patent in process [20]), which cannot be applied in metal monolith catalyst structures and metallic foam structures. This new method arises a great potential for MFEC in many heterogeneous catalyst applications with original optimized catalyst recipe, instead of seeking of new recipe for washcoating support. In this study, all metal MFECs were made of 4-μm-diameter and 3-mm-length, and 12-μm-diameter and 3-mm-length metal fibers (IntraMicron, Auburn, AL, USA). After being sintered, the MFEC sheet (Fig. 1) was punched to disks sized to stack into the reactor tube. In order to ensure good contact and avoid dead space between the tube wall and MFEC, the diameter of the MFEC disk was 105% of the ID of the tube.

2.2. Thermal conductivity measurement

As shown in Fig. 2, test materials were loaded in the middle section of a 1.5″ (38.1 mm) OD copper tube, which was immersed in a water bath kept at constant temperature during every measurement. Fine thermocouples (Omega, 1/32″, 0.79 mm) were utilized to measure the temperature profiles of the test materials. For transient tests, as Waddams [29] did, there was no gas passing through the test tube, and the tube was filled with stagnant N_2 at ambient pressure. At $t = 0$, the test apparatus was put into the water bath, where it was heated up from room temperature to the water bath temperature. The heating curve on the midplane was recorded by a data logger (Omega, OM-DAQPRO-5300).

For steady-state tests, a N_2 gas stream at room temperature was fed to the test tube immersed in the water bath and heated up along with the testing materials. The temperature profiles inside the materials were measured after the outlet N_2 stream reached a steady temperature. The locations of the thermocouples are show in Fig. 2.

2.3. Samples for thermal conductivity measurement

Copper, nickel, and SS MFECs entrapping 180- to 250-μm alumina particles (Alfa, pore volume 1.14 cc/g, surface area 245 m^2/g) were prepared for thermal conductivity measurements. Packed beds made of copper powder (Alfa, 180–250 μm, pore volume 0.22 cc/g) and alumina particles of the same size were also evaluated for comparative purposes. The properties of all samples are