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Characterization of foam catalysts as packing for tubular reactors

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ABSTRACT

This study deals with the characterization of catalytically active foams as packing for continuous tubular reactors in terms of reactor performance and overall mass transfer. The tubular reactor was operated in a vertical direction such that gas and liquid entered the reactor in co-current upward flow. The hydrogenation reaction of α-methylstyrene was applied for characterization of the Pd/Al_2O_3 foam catalysts with a bed length of 50 cm and a diameter of 1.8 cm. Two pore densities for foams namely 30 and 45 PPI were investigated. The foam catalyst showed an overall mass transfer rate that ranged between 0.2 and 14 s^{-1} for 30 PPI and 0.3–18 s^{-1} for 45 PPI foam catalyst, whereas the energy dissipation was below 1 Wm^{-3}.

1. Introduction

The chemical industry is a major consumer of energy worldwide and yet the majority of chemical processes involve catalytic reactors. Therefore, the energy efficiency of chemical reactors is a very crucial issue. For the heterogeneously catalyzed multiphase reactions, several reactor technologies have been already applied such as bubble columns, trickle bed reactors, CSTRs or there are technologies under development for instance wall-coated monolith reactors, monolith reactors with dumped catalysts and last but not least microreactors. The application of bubble columns and CSTRs involves suspended catalysts that demand a subsequent separation of catalyst particles. The pelleted catalysts are usually applied as random packed bed in trickle bed reactors. Yet the disadvantages of the random packed beds are high pressure drop and formation of hot-spots. Microreactor is an innovative reactor concept but the fabrication of microractors is associated with high production costs and the pressure drop inside such reactors is relatively high. Usually a trickle regime is favored in conventional reactor design for continuous three phase reactions, where the gas side mass transfer is limiting due to low gas-liquid solubility. The advantage of the catalytically active foam packings is that, because of the high voidage (up to 97%) and low pressure drop, it is possible to run three phase reactions in an upward flow direction. A challenging issue of the design of trickle bed reactors is dealing with “hot spots” in the reactor, that causes a non-uniform temperature profile. Such hot spots are known to be undesired and to cause side products and shorten the life of the catalyst. Application of catalytically active foam packings in a co-current upward direction can increase the uniformity of the temperature profile due to very low energy dissipation, high void volume, and increased liquid holdup.

Measurement of overall volumetric mass transfer can be utilized as a criterion to evaluate the performance of a structured packing. Several studies used the hydrogenation of AMS using Pd/Al_2O_3 catalysts to evaluate the performance of various reactor types. Turek and Lange [1] investigated a trickle bed reactor with an inner diameter of 3.4 cm with liquid flow rates ranging between 0 and 1.5 L h^{-1} and gas flow rates from 0 to 100 L h^{-1}. Cini et al. [2] adopted the hydrogenation of AMS for tubular supported and Pd/γ-Al_2O_3 impregnated ceramic membranes. Kreutzer et al. [3] used this hydrogenation reaction to characterize the performance of a tubular reactor with wall-coated monolith packing. Purnama et al. [4] investigated the performance of a flow-through ceramic membrane reactor at temperatures ranging between 45 and 50 °C and pressures between 1 and 40 bar, and compared the performance of this reactor with such other reactor types as trickle bed and bubble column reactors. Haase [5] characterized wall-coated and composite (packed with pellets) minichannels for gas-liquid-solid-reactions in various multiphase flow regimes. Langsch [6] studied mass transfer in miniaturized packed bed reactors at different multiphase flow regimes using the hydrogenation of AMS to cumene. Tourvieille et al. [7] investigated minichannels packed with foam catalysts by applying a pulsing flow regime and obtained overall mass transfer values that ranged from 0.25 s^{-1} to 1.9 s^{-1}.

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Nomenclature

a_{GL}	Gas liquid interfacial area [m^2]
a_{LS}	Liquid solid interfacial area (m^2)
a_{ov}	Gas liquid solid interfacial area (m^2)
c_j	Concentration of species j (mol m^{-3})
$E_{A,obs}$	Observed activation energy (kJ mol^{-1} K^{-1})
$E_{A,int}$	Intrinsic activation energy (kJ mol^{-1} K^{-1})
$E_{A,diff}$	Diffusion activation energy (heat of adsorption) (kJ mol^{-1} K^{-1})
H	Henry coefficient [–]
k_L	Liquid side mass transfer coefficient (m^2 s^{-1})
k_{LS}	Liquid-solid mass transfer coefficient (m^2 s^{-1})
k_{int}	Intrinsic reaction rate of AMS hydrogenation (s^{-1})
k_{obs}	Observed reaction rate of AMS hydrogenation (s^{-1})
n_H	Moles of reacted hydrogen (mol)
p_{tot}	Total reactor pressure (Pa)
r_H	Observed reaction rate (mol m^{-3} s^{-1})
T_R	Reactor temperature (K)
V_H	Volume of hydrogen in reactor (m3)
V_R	Reactor volume (m^3)

Greek letters

$\varepsilon_{\Delta G}$	Gas holdup for reacted hydrogen (–)
η	Effectiveness factor (–)
ρ_{Pd}	Mass of Pd per reactor volume (g m^{-3})
τ	Residence time (s)

The present study deals firstly with characterization of the reactor performance foam catalysts as tubular packings. Subsequently, the overall mass transfer of 30 PPI and 45 PPI foams were measured and compared with the data from literature. The overall mass transfer is referred to the external mass transfer which is mainly influenced by the foam structure, whereas the internal mass transfer in the washcoat also plays a role. Thus, internal mass transfer was considered in the mathematical model used to evaluate the observed reaction rates. Although there is some effect from internal mass transfer, the influence of the internal mass transfer was although available however, the major concentration of the palladium was detected in the vicinity of the washcoat surface as shown in a previous paper [10] by applying EDX analysis. Therefore, the influence of the internal mass transfer for different washcoat thicknesses between 17.8 μm (for 30 PPI) and 6 μm (for 45 PPI foams) plays a less important role.

2. Experimental

2.1. Catalyst preparation

The foam catalysts with pore densities of 30 PPI and 45 PPI were prepared using the methods in a previous publication by Lali et al. [8] and Leon et al. [9].

The preparation steps of a 30 PPI foam catalyst are shown in Fig. 1, where the image number 0 represents an uncoated aluminum foam, number 1 was an anodized foam, number 2 was washcoated anodized foam with alumina slurry, number 3 was impregnated with palladium acetate and number 4 was reduced foam catalyst. The Image 5 in Fig. 1 shows a spent foam catalyst.

2.2. Pretreatment of α-Methylstyrene

The purities of the α-methylstyrene and cumene supplied by SIGMA-ALDRICH were 99% and 98%, respectively. In order to prevent the AMS from polymerizing, 15 ppm of the inhibitor *tert*-butylcatechol (TBC) was added by the supplier. Earlier experiments showed that the TBC can build residues on the catalyst and decrease the catalytic activity drastically. The authors have several views about the deactivation of Pd/Al_2O_3 catalyst by TBC. While some studies [10] confirm a negative impact of TBC on catalytic activity, Meille et al. [11] found neither a negative impact on a Pd/Al_2O_3 catalyst nor any reaction inhibition. Nevertheless, our own preliminary experiments confirm the inhibitory effect of TBC. The

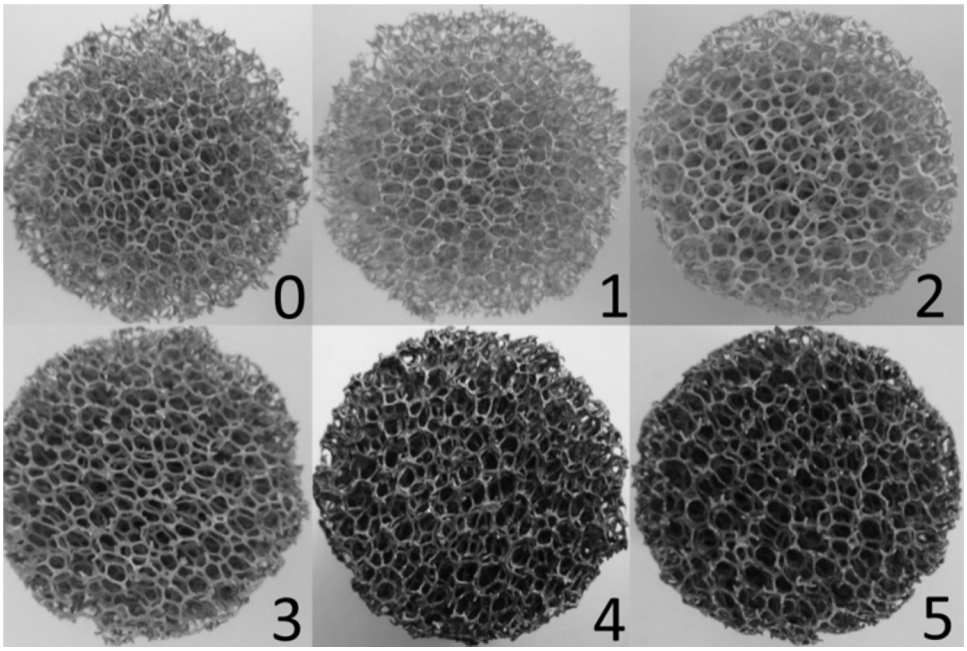

Fig. 1. Pictures of foam samples after different preparation steps 0) pristine aluminum foam 1) anodized foam 2) coated with alumina 3) impregnated with palladium acetate 4) reduced Pd/Al_2O_3 foam catalyst 5) spent foam catalyst.