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Hydrodynamics and mass transfer in carbon-nanofiber/graphite-felt composite under two phase flow conditions

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ABSTRACT

A carbon nanofiber (CNF)/graphite felt composite was synthesized by growing CNFs on the surface of graphite fibers and was used as the packing of a fixed bed reactor under two phase flow conditions. The pressure drop, axial dispersion and mass transfer in the liquid were studied by experiment and by piston dispersion exchange (PDE) model. It was shown that the pressure drop and total liquid up could be predicted by the slit model in an acceptable accuracy. The axial dispersion in the liquid phase in the composite and the mass transfer between the dynamic and static liquid are higher than in the packed bed of solid particles owing to the porous and fluffy CNF layer on the carbon felt fiber.

1. Introduction

Carbon nanofibers (CNFs) have received extensive academic and industrial interests for their tunable bulk and surface structures and novel chemical and physical properties. They have large external surface area, considerable resistance to acids and bases, and strong mechanical strength. Among all possible applications of CNFs, the most perspective one seems to be the catalyst support. It has been widely reported that CNF-supported catalysts display unusual behaviors compared with traditional supports such as silica, alumina, and active carbon [1–3]. But the small dimensions of CNFs inhibit their use as catalyst support in the form of powders in chemical industry. When CNF powders are packed in fixed bed reactors, the pressure drop along the bed will be extremely high; while in fluidized bed reactors, the separation of CNFs from the fluid will be very difficult. In order to overcome the drawbacks of their small dimensions, CNFs are immobilized in a substrate with a macroscopic shape to form a CNF composite. Different materials have been used to immobilize CNFs, such as metallic filters [4], metallic sinter-locked microfibers [5,6], solid carbon foams [7], cordierite monolith [8], solid particles [9], graphite fiber felt [10,11], etc.

For traditional fixed bed reactors packed with solid particles, such as spherical particles and cylindrical extrudates, the pressure drop is relatively high due to the small porosity (0.3–0.6) of the packings. On the other hand, the pressure drop of structured catalysts, such as foam [12], monolith [13], cloth [14–16], fibers [17], etc., is small because of their large porosities and ordered structures.

It has been reported that the structured fibrous catalysts have low pressure drops and excellent mass/heat transfer properties and have potential applications for both gas and liquid phase reactions [18]. Therefore, we grew CNFs on graphite fiber felt to form a CNF/felt composite as a structured catalyst support. The hydrodynamics of structured catalysts are very important for their catalytic performance. However, as a new composite material, the CNF/felt composite has not been systematically studied. Only recently did we relate the pressure drop for single-phase flow to the structural properties (porosity and fiber diameter) of the composite [19].

In the present work, the hydrodynamics and mass transfer in the CNF/felt composite under two phase flow conditions are investigated. The pressure drop and total liquid hold up are determined, and the axial dispersion and the mass transfer in the liquid are studied by residence time distribution experiments and with the piston-dispersion-exchange (PDE) model.

2. Experimental

2.1. Synthesis of CNF composite

Graphite fiber felt, which has a porosity of 0.93 and a fiber diameter of 15.8 μm, was first tailored into a desired shape and dimension (cylinder, Φ35 mm × 10 mm) and then impregnated

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Nomenclature

Bo	Bodenstein number
c_d	concentration of tracer in dynamic phase (mol/m^3)
c_{d1}	concentration of tracer in dynamic phase at the exit of the bed (mol/m^3)
c_s	concentration of tracer in static phase (mol/m^3)
$c(t)$	concentration of tracer at the exit at time t (mol/L)
c_0	moles of tracer/volume of the packed bed (mol/m^3)
C_d	concentration of tracer in dynamic phase
C_s	concentration of tracer in static phase
D_{ax}	axial dispersion coefficient (m^2/s)
d_f	diameter of fibers (m)
d_f'	diameter of the static liquid covered fiber (m)
d_{f0}	diameter of graphite fibers (m)
d_p	particle diameter or equivalent diameter of fibers (m)
E_1, E_2	Ergun constants
$f(t)$	residence time distribution function (s^{-1})
g	gravitational acceleration constant (m/s^2)
Ga_G	Galileo number for gas ($= gd_p^3\varepsilon^3\rho_G^2/(\mu_G^2(1-\varepsilon)^3)$)
Ga_L	Galileo number for liquid ($= gd_p^3\varepsilon^3\rho_L^2/(\mu_L^2(1-\varepsilon)^3)$)
k_La	mass transfer coefficient between dynamic and static liquid (s^{-1})
L	length of the fixed bed (m)
N_α	number of mass transfer units ($=k_LaL/\phi u_d$)
ΔP	pressure drop (Pa)
Pe	Peclet number ($=u_dL/D_{ax}$)
Q_L	flow rate of liquid phase (m^3/s)
Re_G	Reynolds number for gas ($= \rho_G u_G d_p/(\mu_G(1-\varepsilon))$)
Re_L	Reynolds number for liquid ($= \rho_L u_L d_p/(\mu_L(1-\varepsilon))$)
S	cross-sectional area of the packed bed (m^2)
t	time (s)
u_d	velocity of liquid in dynamic phase (m/s)
u_G	superficial velocity of gas (m/s)
u_L	superficial velocity of liquid (m/s)
V_R	volume of fixed bed (m^3)
x	distance down the fixed bed (m)
z	distance down the fixed bed

Greek letters

δ	Dirac function
ε	porosity of the CNF composite
ε_0	porosity of the graphite felt
ε_L	total liquid holdup calculated from slit model
ε_{CNF}	liquid holdup of small pores within CNF layers
ε_{Ld}	dynamic liquid holdup
ε_{Ls}	static liquid holdup
ε_{Lt}	total liquid holdup
θ	dimensionless time
μ_G	viscosity of gas (kg/(m s))
μ_L	viscosity of liquid (kg/(m s))
ρ_G	density of gas (kg/m^3)
ρ_L	density of liquid (kg/m^3)
τ	average residence time (s)
ϕ	dynamic liquid fraction
ψ_G	dimensionless force on gas phase
ψ_L	dimensionless force on liquid phase

Subscripts

d	liquid in dynamic phase
G	gas phase
L	liquid phase
s	liquid in static phase

with an ethanol solution of nickel nitrate to support nickel. The loading of Ni was about 3 wt%. The graphite fiber felt was then dried in air at 120 °C for 12 h and then placed in a vertical quartz tube reactor. The nickel compound was reduced in a gas flow of hydrogen and argon with a molar ratio of 1:3 and a total flow rate of 360 mL/min at 400 °C for 3 h. Then the temperature was increased to 640 °C and the gas flow was replaced by hydrogen and ethane with a molar ratio of 1:1 and a total flow rate of 180 mL/min to grow CNFs. The growth time was 6 h. The loading of CNFs on the graphite felt, which was defined as the weight ratio of the grown CNFs to the original graphite felt, was determined as 0.97 by weighting the graphite fiber felt and the composite.

2.2. Characterization of the composite

The BET specific surface area of the composite was measured by N_2 adsorption–desorption at 77 K on ASAP 2010 (Micromeretics), and the pore size distribution and porosity of the composite was measured by mercury porosimetry on AutoPore IV 9500 (Micromeretics). The morphology of the composite was characterized by scanning electron microscopy (SEM, JSM-6360LV). In the composite, the surface of the graphite fibers was found to be covered uniformly by a layer of CNFs with a thickness of about 10 μm and the diameters of the loaded CNFs were 50–100 nm. Because of the CNFs, the BET specific surface area of the composite increased from less than one to 59 m^2/g, and the diameter of the graphite fibers expanded from 15.8 to 35.3 μm.

2.3. Measurement of the pressure drops and total liquid holdup

The setup for two-phase flow experiments is shown in Fig. 1. The composite, which was wrapped with PTFE tape to avoid bypass of the fluid, was packed in a plexiglass tube with an internal diameter of 35 mm. Gas and cyclohexane were conducted to flow downward cocurrently through the composite. The pressure drop of the composite was measured by a manometer and the flow rate adjusted by a rotameter. In order to measure the total liquid holdup under specified operating conditions, the inlet and outlet valves were closed simultaneously and then the whole column was unloaded and weighed. The weight of held liquid was then calculated by subtracting the weights of the empty column and the dry packing.

2.4. Determination of the flow characteristics of liquid phase

The experimental setup for the RTD measurement is the same for the pressure drop measurement shown in Fig. 1. Liquid (cyclo-

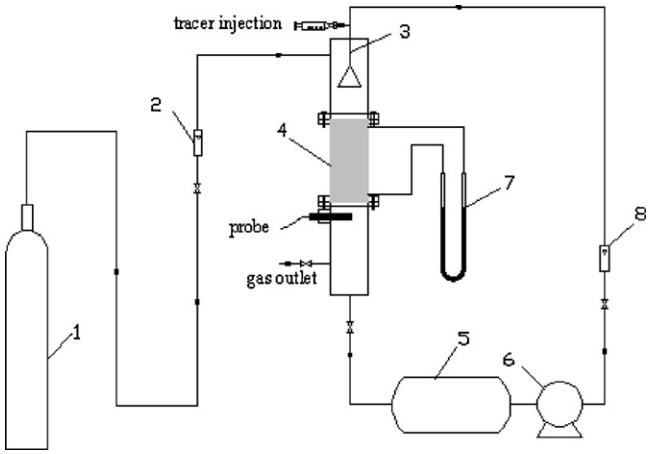

Fig. 1. Experimental setup: 1, gas cylinder, 2, rotameter; 3, nozzle; 4, packing of CNF composite; 5, liquid reservoir; 6, pump; 7, manometer; 8, rotameter.