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Role of gas phase in the deposition dynamics of fine particles in trickle-bed reactors

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Abstract

Experiments were conducted to study the role of gas velocity in the capture of fine particles from non-aqueous suspensions circulated in co-current down-flow trickle flow reactors. The rate of filtration and pressure drop in the trickle bed were investigated using surfactant-stabilized kaolin-containing kerosene suspensions. It was determined that the filter coefficient was sensitive to liquid holdup and specific deposit. The initial collection efficiencies were compared with predictions based on existing theories. Agreement was generally not good with the exception for the limit of low superficial gas velocity. A general correlation establishing the relationship between the filtration rate and the liquid holdup in trickle beds was proposed to reconcile the experimental data with existing filtration theories.

Keywords: Trickle bed; Pressure drop; Filtration rate; Deposition; Plugging; Hydro-treating

1. Introduction

Canadian Athabasca bitumen is known to consist of an extra-heavy hydrocarbon matrix interpenetrated by dispersed clay fine solids. This type of bitumen requires specific upgrading steps for conversion into readily usable light and sweet hydrocarbon fractions. A residual fraction of bitumen-borne fines, albeit small, is often entrained during primary upgrading by the coker-effluent gasoil cuts into the downstream distillation and hydrotreating trickle-bed units. To minimize damage to the hydrotreating catalyst, preventive filters are usually installed prior to the trickle beds to intercept and remove the larger fine particles, ca. 25 μm. However, smaller fines succeed in worming into the downstream catalytic trickle bed. As a matter of fact, even low concentration of fine solids, in the range of several parts per million in the hydrotreater feed, may cause, over months of operation, substantial deposition throughout the catalyst bed. Such an accumulation of fine solids brings about, as a least upset, a pressure drop buildup until the commercial plant may necessitate a shut down and that the catalyst is replaced. When this replacement occurs before catalyst activity has been adequately taken advantage of, operating cost issues emerge.

There is likelihood that removal of these fine particles by the catalyst packing in hydrotreating reactors is analogous to deep-bed (or granular) filtration, where the liquid phase flows through a granular bed while enabling fine particles to deposit on the surface of the collecting packings (Tien, 1989). This process has received extensive literature coverage for aqueous suspensions due to its significance in water purification. Studies on the deposition of fine particles from non-aqueous liquids are, on the contrary, scantier (Chowdiah et al., 1982; Narayan et al., 1997).

Often, non-filterable fine solid impurities such as iron sulfides, sodium chloride, coke, organic precipitates, and sediments, either naturally occurring in the liquid feed or formed in situ, get partially trapped in the trickle-bed hydrotreaters (Narayan et al., 1997; Wang et al., 1999; Gray et al., 2002). Depending on the importance of the gas–liquid interactions in the trickle bed, the presence of a flowing gas phase may drastically affect liquid holdup and liquid interstitial velocity thereby altering the course of deposition within the porous medium. Since classical granular filtration theories deal primarily with

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gas-free single-phase liquid flows, earlier methods for relating the pressure drop to the particulate concentration in deep-bed filtration cannot be transposed to two-phase flows without precautionary adaptations. For instance, Ortiz-Arroyo and Larachi (2005) provided filter coefficient correlations adapted for two-phase trickle flow, which they obtained *numerically* by solving an equation describing the two-dimensional trajectories of fines for both monolayer and multilayer deposition in inclined slits used to approximate the actual tortuous porous media. Iliuta et al. (2003), on the other hand, postulated that single-phase flow trajectory analysis in granular filters, such as the one proposed by Rajagopalan and Tien (1976), was also applicable to trickle-bed filtration for the estimation of the filter coefficients. Until now, however, no experimental proof has been elaborated to prop up such assumptions.

When deposition of fine particles takes place in porous media, the structure of the collector as well as the surface state undergo continuing changes. These changes, in return, are foreseen to affect the course of filtration and deposition rate. In addition, despite the actual problem ascribed to fine particles adverse interference in hydrotreating processes and in spite of the large body of literature on pressure drop behavior due to fines deposition, a very few experimental research studies have focused on gas–liquid flow filtration. Narayan et al. (1997) found that the structure of the particle deposits from a non-aqueous suspension in single-phase liquid flow was sensitive to the prevailing flow conditions, via a suspension Reynolds number. In trickle beds, only the pioneering work by Gray et al. (2002) reported valuable experimental pressure drop data for two-phase flows. However, quantitative information with respect to other key filtration variables such as inlet and outlet particle concentrations in the flowing suspension as well as the filtration rates is lacking.

The purpose of this study was to investigate mainly the influence of superficial gas velocity on the filtration rate in the trickle flow regime. In order to understand the role of the gas phase on the capture of fine particles from non-aqueous media, the superficial liquid velocity was kept constant ($U_L = 0.001\,\mathrm{m/s}$) at varying gas flow rates. The rate of filtration and the pressure drop in the packed bed were monitored using suspensions of surfactant-stabilized kaolin particles dispersed in a kerosene carrier. Finally, a simple correlation was developed for the representation of the filtration rate and was tested against experimental data. This work provides a contribution to the understanding of fines deposition under well-defined operating conditions, to serve as a basis for modeling, optimization, and scale-up of trickle-bed hydrotreaters experiencing filtration conditions.

2. Experimental materials and methods

To understand the role of trickle flow conditions on deposition and plugging in packed-bed hydrotreaters, a simplified experimental system was built and synthetic suspensions were prepared to perform laboratory-scale tests. Kaolin was selected because it is a major component in the clay minerals encountered in the Athabasca oil sand (Tanabe and Gray, 1997). Kerosene was chosen because of its chemical similarity with the hydrocarbon streams, in addition to its stability and relatively low vapor pressure in ambient conditions. For the deposition to occur, the attractive forces between the fine particles and the collector must be strong enough to resist the impeding shear-induced forces arising in the flowing films both at the liquid–collector interface and due to the gas–liquid interface. Consequently, the size of kaolin particles should not be too large. In the other hand, it is important that the suspension remains stable, i.e., does not suffer flocculation and subsequent sedimentation, at least during a time period compatible with the duration of the experiments, typically 70 on-stream hours. In actual industrial suspensions, the hydrocarbon-borne polymeric asphaltene macromolecules are known to adsorb on the surface of the fine particles thereby enhancing the stability of fines suspensions (Wang et al., 1999). To mimic such stability, we have tested different surfactants in order to obtain stable model suspensions of kaolin in kerosene at ambient temperature.

2.1. Design and analysis of model suspension

The suspension used for the experiments was prepared in the following manner. Various surfactants were tested to determine their ability to stabilize a suspension of kaolin. Most surfactants were not soluble in kerosene and/or did not exhibit an effect on the stability of the kaolin suspension. Finally, the surfactant solution was obtained by adding acetyltrimethylammonium bromide ($C_{19}H_{42}NBr$) to kerosene. This solution was mixed for 30 min in an ultrasonic bath to dissolve the surfactant. For all the experiments, a 5% w/w surfactant/kaolin ratio was used. Kaolin was preventively heated during 1 h in a furnace to get rid of any moisture before it was added to the mixture into a 500 mL vessel. The mixture was stirred for 24 h before to be left at rest.

A microscopy video-image system (Olympus SZ-PT- Spot Insight) was used to control and measure the distribution of fine particle diameters in the final suspension. Microscopic images (Fig. 1) of the suspension were observed on a TV monitor, and recorded using a video recorder. Images were displayed on the monitor and analyzed by means of spot advanced software. Fig. 2 gives the size distribution of fines in the influent suspension. The microscopic images of the suspension (Fig. 1) show that without surfactant, flocculation of kaolin occurs and the size distribution of fines is quite large. The mean diameter is approximately 55 μm. With only 5% w/w surfactant, microscopy images reveal that the kaolin suspension is stable and does not flocculate yielding kerosene–surfactant–kaolin suspensions with a mean diameter of approximately 8 μm.

2.2. Process design

The main elements of the experimental setup are schematically represented in Fig. 3. The reactor (I.D. = 5.7 cm) was packed with 2.7 mm spherical γ-alumina catalyst to complete a total bed height of 92 cm with a porosity 32%. The whole