NOVEL JET-LOOP REACTOR FOR GAS-SOLID CATALYZED KINETIC STUDIES USING COMMERCIAL SIZE PARTICLES

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Abstract

Measurement of catalytic kinetics using a well-characterized experimental reactor system that can be operated over the required range of reaction mixture compositions, pressures, and temperatures using commercial catalyst particle is an essential requirement for kinetic model discrimination and parameter estimation. A laboratory-scale reactor system based upon a novel jet loop reactor concept is described for direct measurement of reaction kinetic data that can accommodate commercial-size catalyst particles for various types of gas-phase chemistries. Key features of the reactor include the absence of any moving parts, use of various materials of construction, isothermal operation, and nearly ideal back-mixing for the gas phase. The oxidation reaction kinetics of SO_2 to SO_3 over commercial-size V_2O_5-based catalysts are studied to illustrate the system capabilities, and also to demonstrate an application for an important process technology where identification of catalytic materials have higher activity at lower temperatures for reduction of SO_2 emissions is an ongoing environmentally-driven objective.

Keywords

Laboratory reactor, Jet-loop, Reaction kinetics, Scale-up, SO_2 oxidation, Heterogeneous catalysis.

Introduction

Robust kinetic data for gas-solid and gas-liquid solid catalyzed systems and the associated reaction kinetic models are an essential requirement for development of multiscale reaction engineering models that are used for design of new processes or for troubleshooting and optimization of existing processes (Duduković and Mills, 2014, 2015). For generation of kinetic data, laboratory-scale reactors that operate isothermally with a fluid flow pattern that approaches perfect back-mixing are preferred over those where the flow pattern approaches ideal plug-flow since the data analysis and operation is simplified (Berty, 1984; Berty 1999). Among various laboratory reactor designs with nearly ideal fluid back-mixing, recycle reactors are preferred for catalyst testing and kinetic studies. Illustrations of various recycle reactor design concepts from the literature are shown in Figure 1.

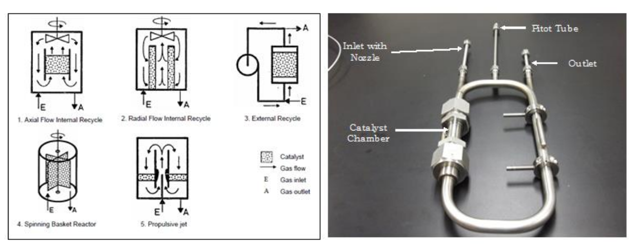

Figure 1. Various types of recycle reactors and photograph of the new jet-loop reactor.

The primary objective of this paper is to describe the development, modeling and application of a new

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experimental reactor that is based upon a propulsive jet concept for determination of gas-solid catalyzed reaction kinetics for commercial-size particles. The oxidation of SO_2 to SO_3 over V_2O_5-based catalysts as realized in commercial-scale processes for the manufacture of H_2SO_4 is used to demonstrate system operation and to illustrate key reactor operational features for determination of the particle-scale reaction kinetics.

Materials and Methods

A photograph of the new jet loop reactor design is shown in Figure 1. This configuration is constructed of Type 316 stainless steel, although other versions have been constructed from other advanced metal alloys or quartz for inertness. The particular catalyst chamber shown here can accommodate several commercial size catalysts whose largest dimension is *ca.* 16 mm.

The jet-loop reactor design evolved through a series of prototypes by performing various experiments under both cold-flow and hot conditions in which the total gas flow rate, gas temperature, reactor total pressure, catalyst particle size, nozzle size, and nozzle location were systematically varied to assess the effect of these variables on the gas recycle rate. The results from these hydrodynamic studies were used to help identify the preferred nozzle parameters and other related mechanical design aspects of the loop reactor.

The oxidation of SO_2 to SO_3 over V_2O_5-based catalysts, which is the basis for all H_2SO_4 processes, was selected as a test reaction due to both its commercial importance and challenging reaction characteristics, such as exothermicity and corrosive nature of the reaction mixture. Details of the automated reactor system and on-line analytical systems will be given in the manuscript.

Results and Discussion

CFD simulation. The hydrodynamics of the proposed jet loop reactor design were first evaluated using a computational fluid dynamics (CFD) model for single-phase turbulent flow through a packed bed using COMSOL Multiphysics 5.0 and subsequently compared to experimental data. Both 2-D and 3-D models were created based upon the k-ε turbulence model. The simulation results were compared to existing correlations for recycle reactors, as well as the experimental data acquired from a prototype laboratory-scale jet loop reactor using N_2 gas under cold-flow conditions.

Figure 2 compares typical experimental and model-predicted values for the gas recycle ratio as a function of the reactor inlet gas flow rate over the range of 0.75 to 2.5 l/min. The results are in reasonable agreement and show that recycle ratios from *ca.* 6 to 10 were achieved under these particular conditions, which were large enough for a moderate range of SO_2 conversions in this work.

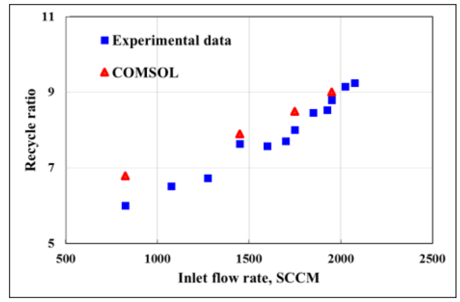

Figure 2. Comparison of predicted and experimental recycle ratio vs inlet gas flow rate.

Kinetic data. Examples of SO_2 conversion *vs* gas flow rate data are provided in Figure 3. The data in Figure 3a are based upon temperatures between 390 to 410ºC using a dry feed gas consisting of ca. 5% SO_2, 15% O_2 and 82.5% (bal) N_2 at flow rates between 100 sccm to 900 sccm. The data in Figure 3b include 20% H_2O as steam to simulate wet acid conditions. The resulting rate data were fitted to a rate model that accounts for reversible reaction behavior. More complex rate equations were also evaluated for goodness-of-fit using robust parameter estimation methods and account for inhibition effects.

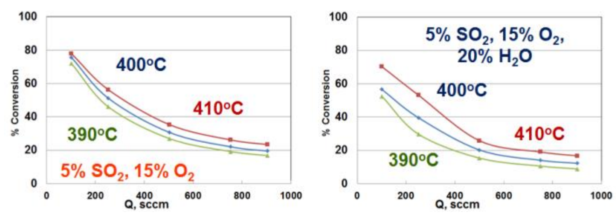

Figure 3a. % SO_2 conversion vs. flow rate (dry gas condition); Figure 3b. % SO_2 conversion vs flow rate (wet acid condition).

These and other detailed results demonstrate the utility of the proposed jet loop reactor as a relatively simple but effective reactor type for studying the kinetics of gas-solid catalyzed reactions for either fundamental kinetic studies, or for evaluating various short to longer-term measures of performance for commercial catalysts.

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