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High-Temperature Organic-Fluid Fouling Unit *

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A new type of fouling unit is developed for high-temperature (500 °C) and high-pressure (70 atmosphere) fouling experiments by modifying a commercial autoclave. Key modifications are the installation of a helical impeller in a flow tube and a fouling probe in the autoclave to simulate the fluid dynamics and heat transfer of typical heat-exchange equipment. A calibration technique is described, and fouling results are presented for experimental runs with indene and kerosene. The results are compared with those obtained using other types of fouling test units. Other potential applications of the fouling unit, such as corrosion and micro-scale reaction experiments, are discussed.

INTRODUCTION

Mechanisms for chemical-reaction fouling are complex and often involve several processes. Some previous investigators (Crittenden et al. [1]; Panchal et al. [2]; and Fryer et al. [3]) have developed analytical models to explain the experimental results; however, the present knowledge is inadequate to predict fouling rates from first principles. Experimental data obtained with prototype fluids at industrial conditions is essential.

This paper describes a fouling unit for high-temperature, high-pressure applications. The unit was developed as a part of the cooperative work between Argonne National Laboratory (ANL), Heat Transfer Research, Inc. (HTRI), and Chevron Research and Technology Company (CRTC).

FOULING MEASUREMENTS

The four common objectives for conducting fouling measurements are to determine: (1) the rate of fouling at process conditions to establish cleaning cycles, (2) threshold conditions for initiation of fouling, (3) the effectiveness of new chemical and physical mitigation methods, and (4) the change in the fouling rate due to changes in the chemical composition and/or operating conditions. Fouling is commonly measured as the change in the overall heat-transfer coefficient; but often other measurements are used. Knudsen [4] and Melo et al. [5] reviewed commercial and laboratory monitors used to measure the rate of fouling for low-temperature applications. Marner and Henslee [6] reviewed available fouling probes for high-temperature gas streams. Fouling in the refining industry occurs at high-temperature (>250 °C) and high-pressure conditions (> 20 atm.); therefore, these fouling units can not be used. Fouling units used for high-temperature organic-fluid fouling experiments are summarized in Table 1.

Table 1. Devices for Fouling Measurements.

Fouling Unit	Measurement Principle	Typical Reference
Jet Fouling Test Oxidation Tester (JFTOT)	Mass collection of solid by a filter	Hazlett et al. [7]
Thermal Fouling Test	Change in fluid temperature for given heat rate	Dickakian [8]
Closed-flow loop	Change in heat-transfer coefficient measured by a fouling probe	Panchal and Watkinson [2], Crittenden [9]
Laboratory Fouling Test Apparatus (LFTA)	Autoclave with rotating cylinder around a heating probe	Eaton and Lux [10]

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The JFTOT (Hazlett et al. [7]) is used to determine fluid stability in the formation of insoluble resins, and the Thermal Fouling Test unit (Dickakian, [8]) is used to characterize fouling of petroleum fluids and to evaluate the effectiveness of different chemical additives. These two units are designed to provide qualitative results; hence, they can not be used to determine threshold conditions. Closed-flow loops are commonly used to investigate the fouling mechanisms, determine threshold conditions, and develop new mitigation methods. Their major limitations are that they are costly to build and maintain, difficult to keep contaminant free, and are not easily portable. Closed-flow-loop fouling monitors used to investigate organic-fluid fouling are either in-tube or annulus-flow heat transfer monitors (Panchal and Watkinson, [11]). In either case, the test fluid is pumped from a fluid reservoir through the fouling monitor, a network of pipes, and back to the reservoir. Overall, the fouling simulations for these types of systems are complicated by their designs.

DESIGN DEVELOPMENT

Four major criteria considered in the development of the fouling unit are as follows: (1) low cost, (2) compact design for ease of maintenance, (3) high-temperature (>300 °C) and high-pressure (10 to 70 atm.) capabilities, and (4) known fluid dynamics.

One way to design a fouling unit would be to enclose a fouling probe in an autoclave. The fouling unit developed by Eaton and Lux [10] would meet the first three criteria described above; however, the fluid dynamics of their fouling probe is not easily applied to heat-exchanger conditions. This limitation is corrected by configuring a flow tube over a probe that provides an annular passage. A diagram of the autoclave is shown in Figure 1, and a photographic view of the internal assembly is shown in Figure 2. Inside the pressure vessel, a stainless steel flow tube is connected to the thermowell and aligned vertically. A helical impeller, in the upper portion of the tube, drives the fluid upwards. The fouling probe is similar to those used by other investigators (Knudsen [4]). The probe contains a 76.2 mm heated section and three interior thermocouples positioned near the wall to monitor the rate of fouling. The probe can be easily removed for examination and deposit sampling.

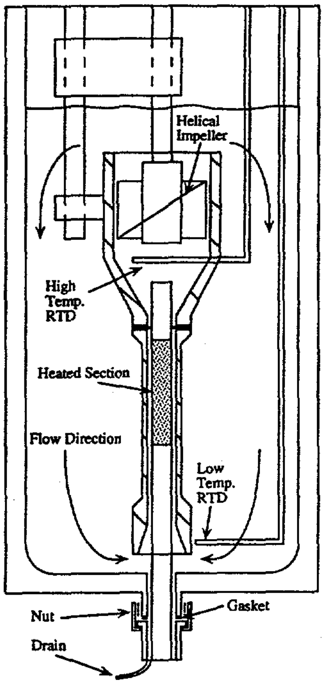

Figure 1: Schematic diagram of the autoclave interior.

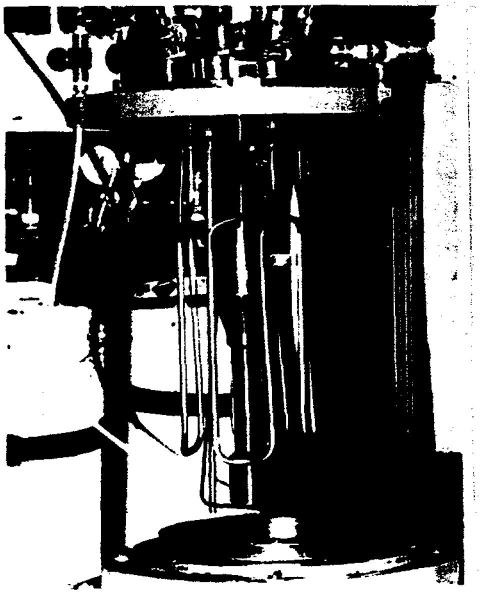

Figure 2: Photograph of the autoclave interior.

The fouling unit meets all of the four objectives for fouling measurements described above. The unit can be used for experiments at conditions which cover many refinery processes. Another major advantage is that it can be easily cleaned, which typically takes two to four hours.

FLOW CALIBRATION

The major limitation of the fouling unit is that conventional flowmeters can not be easily installed to measure the flow rate directly inside the flow tube. Therefore, the flow rate is calculated on the basis of heat and mass balances. A differential resistance temperature device (RTD) is installed across the flow tube to measure the change in fluid temperature (Figure 1). The flow rate is inversely proportional to the increase in the fluid temperature through the tube.

The flow calibration results for kerosene, water, and acetone are shown in Figure 3. Calibration tests were conducted at 3 to 5 atm. pressure, 50 °C, and 65 to 230 kW/m^2 of power on the fouling probe. A linear relation between the rotation rate of the impeller and the flow rate was observed, and the flow-calibration data fall within 10% of the linear regression.

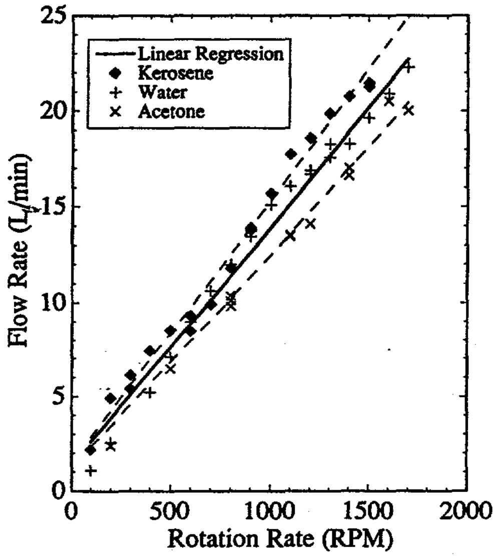

Figure 3: Flow calibration plot.

HEAT TRANSFER CALIBRATION

The major elements of the heat transfer calibration are to determine the wall resistance, develop a predictive method for the heat transfer coefficient, and compare results with a literature correlation for annular flow (e.g. Knudsen and Katz, [12]). In order to calculate the wall resistance, a modified Wilson plot, shown in Figure 4, is used. With this method, an appropriate ratio of the physical properties and the fluid velocity is used as an independent variable. Use of three test fluids with widely different physical properties increases the reliability of the calibration. A linear regression of the data gives an intercept of 0.052 m^2 K/kW for the wall resistance, R_w. This corresponds well within 10% of the theoretically calculated wall resistance of 0.056 m^2 K/kW. An important point, not considered by the previous investigators, is that the wall resistance changes during the fouling test as the wall temperature increases. As a result, the calculated fouling resistance is generally lower than the actual value. Therefore, changes in the thermal conductivity should be accounted for in calculating the fouling resistance.

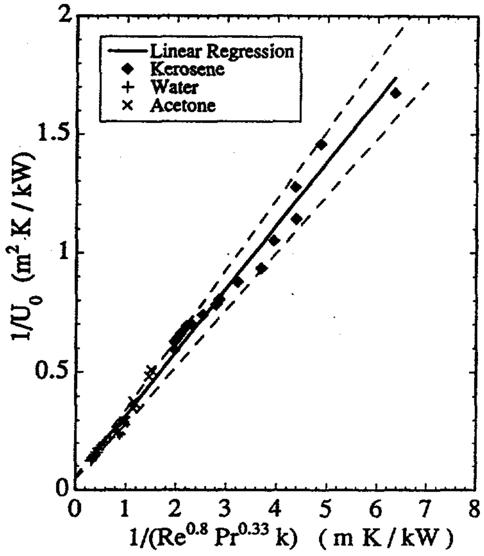

Figure 4: Modified Wilson plot

Theoretical values of the heat transfer coefficient are calculated using the correlation given by Knudsen & Katz [12] for annular flow. The constant in the literature correlation is 0.02. Using this method, the experimental heat transfer coefficients are generally 30 to 50% greater than the theoretical values for the whole range of test conditions. Deviations of this magnitude are expected due to entrance effects for a low length-to-diameter ratio of 12. Therefore, a new value for the constant, equal to 0.03, was used to predict heat transfer coefficients in the apparatus; following Nusselt [13] and Boelter, Young, and Iverson [14], who recommended using correction factors to account for entrance effects. A comparison between the revised correlation and the experimental data is shown in Figure 5. Most of the data points lie within the measurement accuracy of 10%. The heat transfer results show that the fluid dynamics and thermal conditions are reproducible in the fouling unit, thereby qualifying it for fouling experiments.

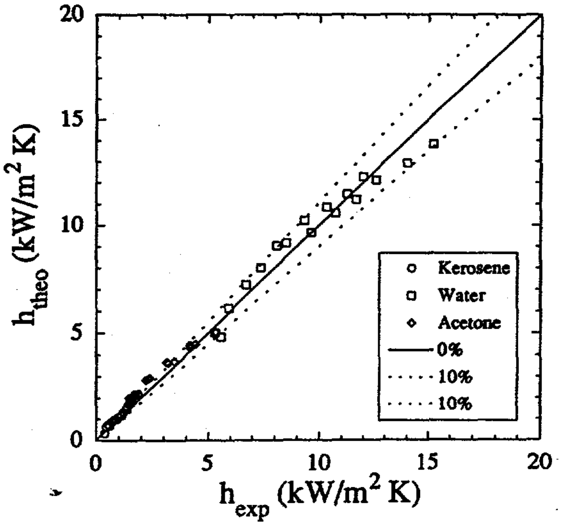

Figure 5: Comparison between experimental and predicted heat transfer coefficients.

FOULING

The first set of experiments performed in the autoclave are duplicates of those previously done in the organic-fluid fouling loops at Argonne and the University of British Columbia (Panchal and Watkinson, [2]). Five liters of test fluid containing 10% indene in kerosene, prepared by weight and aerated for 4 hours under 4 atmospheres of pressure, is used for the fouling experiments described below.

Results from three of the indene/kerosene experimental runs are shown in Figures 6, 7, and 8.

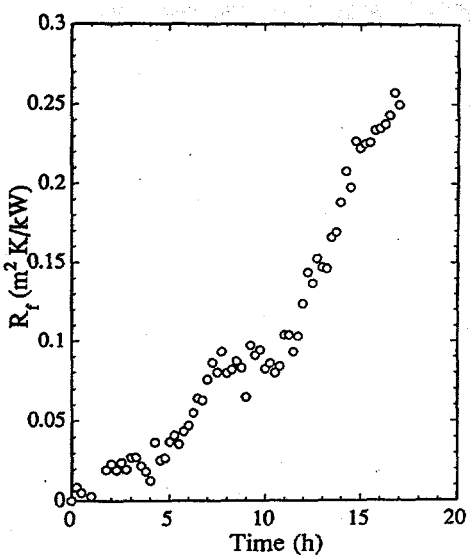

Figure 6: Fouling curve for 10% Indene in Kerosene. Test conditions are as follows: 0.6 m/s, 100 kW/m^2, 82°C, and 4 atm.

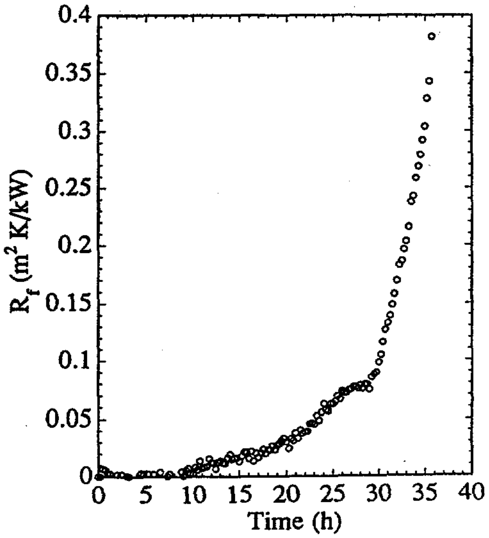

Figure 7: Fouling curve for 10% Indene in Kerosene. Test conditions are as follows: 1 m/s , 194 kW/m^2, 82°C, and 4 atm.

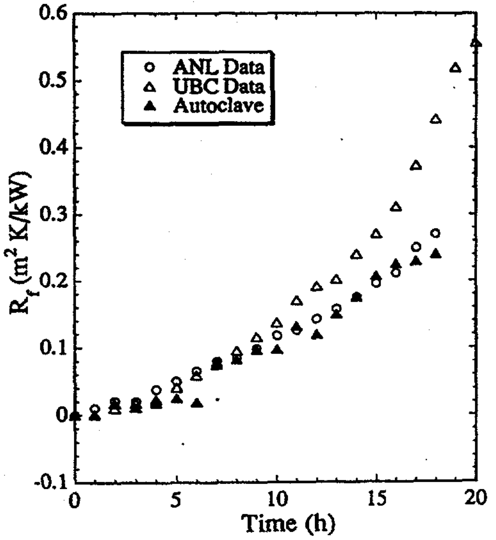

Figure 8: Comparisons between the autoclave and UBC and fouling loop apparatuses. Test conditions are as follows: 0.7 m/s, 214 kW/m², 82°C, and 4 atm.

These plots illustrate the increase in the fouling resistance typical of autoxidation of olefins shown by the previous experiments with flow loops (Panchal and Watkinson, [2]). As seen in Figures 6 and 7, when the fluid Reynolds number through the annulus was increased from 6,100 to 10,400, the fouling rate dropped significantly. However, after an extended period of testing, an accelerated rate of fouling occurred due to build up of high concentrations of fouling precursor hydroperoxides.

A comparison between the data obtained using the fouling unit and the earlier data with closed flow loops by Panchal and Watkinson [2] with in-tube and annular-flow units is shown in Figure 8. As shown in Table 2, the wall temperature, chemical composition, and the flow conditions were comparable for the three sets of data. The results show that the fouling curve obtained with the autoclave apparatus is comparable to the other two sets of data. The results can be further validated by comparing the heat transfer coefficients from the three sets of experiments at the beginning before fouling started. On the basis of these comparisons, it can be concluded that the autoclave apparatus produces heat transfer and fouling results comparable to the flow loop apparatus used in the previous investigations.

Table 2. Test conditions for indene/kerosene fouling experiments.

	Autoclave Unit	Argonne Flow Loop	UBC Annular-Flow Loop
Indene, wt %	10	10	10
Pressure, atm	4	4	4
Fluid T., °C	82	82	84
Surface T., °C	188	188	188
Reynolds number	6,100	10,000	11,000
Heat-Transfer Coefficient, m² K/kW	1.3	1.7	1.4

OTHER APPLICATIONS

The discussion above was focused on laboratory experiments, in which a batch of fluid is used for fouling experiments. There is a major technical issue of how to apply the experimental data obtained with closed-flow-loop experiments to industrial processes. The physical and chemical changes that occur during the test period must be taken into account when applying the laboratory data to industrial conditions. The present unit can be easily modified for field experiments. Depending on the required residence time, the flow rate of a given process stream can be controlled. One of the major advantages of the fouling unit for field experiments is that the safety requirements are easily met at relatively low costs.

The major limitation of corrosion experiments is that they are conducted without fouling and prototype heat transfer conditions. The present fouling unit can be used to investigate the interactive effects of fouling and corrosion at prototype temperatures, pressures, and fluid-dynamic conditions. For corrosion/fouling experiments, a thin metallic sleeve can be shrunk-fit on the probe. If necessary, the sleeve can be removed for surface analysis.

Many chemical reactions occur with a finite heat of reaction that must be removed or added depending upon whether the reaction is exothermic or endothermic. The fouling unit can be used for endothermic, catalytic, or non-catalytic reactions to develop a kinetic model and determine the effects of fluid dynamics of flow reactors. The reaction rate can be easily correlated with the thermal energy supplied to the probe. The unit is ideally suited for investigating threshold conditions for catalyst fouling, in which the heated section would be coated with an appropriate catalyst and directly measured by monitoring the wall temperature. A probe for exothermic reactions, similar to that developed for gas side fouling (Marner and MacDavid, [15]), would be used.

CONCLUSIONS

A new fouling unit was developed for high-temperature and high-pressure fouling experiments. The unit not only serves as a research tool, but can be used in field testing to obtain fouling data to determine threshold fouling conditions, effectiveness of new and improved mitigation methods, and changes in the fouling characteristics due to different chemical composition and/or operating conditions. In addition to fouling experiments, the unit can be used for corrosion and chemical-reaction experiments.

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NOMENCLATURE

d_o, d_i	outside and inside annulus diameters (m)
C_p	heat capacity (kJ/kg °C)
D_e	equivalent diameter ($= d_o - d_i$) (m)
h	heat-transfer coefficient (kW / m^2 K)
k	thermal conductivity (kW / m K)
Pr	Prandtl number ($= C_p \mu / k$)
R_f	fouling resistance
Re	Reynolds number ($= D_e V \rho / \mu$)
t	time (s, hr)
T	temperature (°C)
U_0	overall heat transfer coefficient (kW / m^2 K)
V	velocity (m/s)
μ	viscosity (kg/m s)
ρ	density (kg/m^3)

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