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NOVEL SUPPORTS FOR COAL LIQUEFACTION CATALYSTS

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NOVEL SUPPORTS FOR COAL LIQUEFACTION CATALYSTS

DOE/PC/88942-5

Henry W. Haynes, Jr.

ABSTRACT

Cold model studies of the liquid fluidized bed microreactor are essentially complete. This report documents experimental data obtained for the optimum (60° blade angle) axial flow impeller in Gas/Liquid/-Solid service. Dimensionless correlations are presented for the critical stirrer speeds corresponding to incipient fluidization, S_1 , and particle carryover, S_2 . Under some circumstances bed expansion ceases prior to particle carryover due to cavitation in the impeller. A correlation is developed for S_3 , the stirrer speed at which cavitation is first observed, and by comparing S_2 and S_3 it is possible to establish whether the bed can be fully expanded.

Construction of the stainless steel vessel and internals was completed during the last quarter, and assembly of the CCLM unit is underway.

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INTRODUCTION

Catalyst deactivation is a severe problem in coal liquefaction applications. Generally speaking, catalysts deactivate with process time as a consequence of coke formation, metals and ash deposition and sintering. The evidence is suggestive of two types of coke deposition. During the early stages of liquefaction the chief culprit may be categorized as "Type I" coke formation. Many investigators have observed that coke builds up rapidly to levels sometimes exceeding 20 percent by weight, and that this buildup is accompanied by a sharp decrease in catalyst activity. After this initial period, the coke content of the catalyst remains nearly constant, and a scan of a "coke equilibrated" catalyst particle indicates that carbon is deposited uniformly throughout the pellet cross-section. Type I coke deposition is accompanied by a substantial reduction of pore volume in the micropore region and clogging of these fine pores is undoubtedly a major contributor to the deactivation. In some situations carbonaceous deposits will accumulate at the external surface of the catalyst particle. This "Type II" carbon deposition has been observed on spent coal liquefaction catalysts (1), and on catalysts that had seen service in the hydrotreatment of heavy coal liquids (2, 3). It appears that the critical factor is the presence of benzene (or toluene) insolubles. It is speculated that the Type II carbon deposits served to plug external pores and prevent access to the particle interior.

Metals and ash deposits also accumulate at the external surface of the catalyst particle as time progresses, and this leads to deactivation of the catalyst most probably via a pore plugging mechanism. The commonly deposited metals include Fe, Ti, Ca, B, Mg, Na, and Si.

Concentrations of up to (and sometimes exceeding) five weight percent are common. The analysis of spent catalysts from the Wilsonville facility revealed that the penetration depth can be quite substantial (4).

Sintering is reported to be a cause of deactivation in the H-Coal process (5). The effect is especially pronounced when low rank coals are processed, and the degree of sintering appears to be related to the presence of water in the reaction mixture. Thomas and Sample report a loss of catalytic metals--Mo, Ni and Co--from spent H-coal catalysts (6). Similar phenomena have been reported in commercial hydrodesulfurization processes.

The objectives of this research are first to construct a Catalytic Coal Liquefaction Microreactor (CCLM) and then to use this unit to screen catalysts prepared from novel and/or modified supports for coal liquefaction applications. Run lengths will be sufficient to give a measure of catalyst deactivation rate. Modified supports will include aluminas treated with alkali and alkaline earth metals and halogens. Novel supports will include carbons, single component oxides and composite oxides. Throughout this work, efforts will be made to correlate coking tendency with catalyst acidity measurements. While the emphasis of this investigation is not directed towards pore structure, the importance of this variable is not underestimated. Pore structures of all catalyst samples, both fresh and spent, will be monitored routinely.

ACCOMPLISHMENTS

Our efforts during the last quarter were devoted almost entirely to wrapping up the cold model study of the CCLM and to construction of the actual unit. Correlations were developed for the critical stirrer speeds in Gas/Liquid/Solid service for the optimum axial flow impeller. Laboratory work has identified a coal paste composition that can be conveniently extruded into the reactor.

Cold Model Studies of the CCLM (R. Borgialli, T. Zhang):

In the previous report (7) correlations were presented for the critical stirrer speeds corresponding to incipient fluidization, S_1 , and particle carryover, S_2 , for a radial flow impeller in Liquid/Solid systems. The radial flow design was found to be unsuitable for Gas/-Liquid/Solid fluidization, however, and initial testing of an axial flow impeller was described for this application. Of the several variations tested, an axial flow impeller having a 60° blade angle performed best. The development of dimensionless correlations for the 60° axial flow impeller is described below.

A drawing of the reactor assembly is presented in Figure 1. Feed enters from the bottom through the hole drilled along the axis, and product is collected through a 3/16 in. diameter drawoff tube as indicated on the drawing. The liquid level can be adjusted by varying the length of the drawoff tube. The catalyst basket, which consists of a cylindrical tube with wire screen ends, is held in place by spring clips welded to the wall at 120° spacings. These are visible in the scale drawing of the basket, Figure 2. A drawing of the 60° axial impeller is provided in Figure 3.

Figure 1. Reactor assembly

Figure 3. 60° axial impeller (actual size)

Figure 2. Catalyst basket (actual size)

Results from cold model studies on the arrangement of Figure 1 are compiled in Table 1. Experiments were conducted with glass ($\rho_p = 2.57$ g/cc) and steel ($\rho_p = 8.25 - 8.42$ g/cc) spheres of several sizes and with aluminum ($\rho_p = 2.70$ g/cc) cylinders. When no entry appears under the particle length, L_p , heading of Table 1, the particles are spheres. Liquids employed in the study included vegetable oil ($\mu_\ell = 57$ cp), SAE 5W-30 motor oil ($\mu_\ell = 120$ cp) and SAE 30 motor oil ($\mu_\ell = 245$ cp). Gas density was varied by using air ($\rho_g = 1.29 \times 10^{-3}$ g/cc) and helium ($\rho_g = 0.179 \times 10^{-3}$ g/cc). Gas flowrate, Q_g , was varied over a wide range. Liquid height, H_ℓ , and rest bed height, H_b , were also varied. All experimentation was conducted at ambient conditions.

For Gas/Liquid/Solid fluidization, dimensional analysis identifies a total of 8 dimensionless correlating variables. Upon taking the characteristic dimension of the assembly as the impeller diameter, $d_i = 9.5$ mm, these groups may be written as:

$$Re_1 = \frac{S_1 \rho_\ell d_i^2}{\mu_\ell} = \text{rotational Reynolds number.}$$

$$Ga = \frac{d_{p,eff}^3 (\rho_p - \rho_\ell) \rho_\ell g}{\mu_\ell^2} = \text{Galileo number}$$

$$Fr = \frac{Q_g}{d_i^2 g d_{p,eff}} = \text{Froude number}$$

$$(\rho_p - \rho_\ell)/\rho_\ell = \text{buoyancy factor}$$

$$\rho_g/\rho_\ell = \text{gas density factor}$$

$$d_{p,eff}/d_i = \text{particle size factor}$$

$$H_b/d_i = \text{bed height factor}$$

$$H_\ell/d_i = \text{liquid height factor}$$

Table 1: Gas/Liquid/Solid Fluidization Results for 60° Axial Impeller

Q_g (std cc/mm)	H_b (mm)	H_t (mm)	d_p (mm)	L_p (mm)	ρ_p (g/cc)	ρ_l (g/cc)	$\rho_g \times 10^3$ (g/std.cc)	μ_l (cp)	S_1 (rpm)	S_2 (rpm)
0.	10	65	3.	—	2.569	0.875	—	57.	394	1279
0.	10	65	4.	—	2.569	0.875	—	57.	541	1500
0.	10	65	5.	—	2.569	0.875	—	57.	689	1648
0.	10	65	6.	—	2.569	0.875	—	57.	836	1795
0.	10	65	3.1	—	8.424	0.875	—	57.	910	—
0.	10	65	4.67	—	8.254	0.875	—	57.	1500	—
0.	6	65	3.	—	2.569	0.875	—	57.	394	1500
0.	6	65	4.	—	2.569	0.875	—	57.	541	1648
0.	20	65	5.	—	2.569	0.875	—	57.	615	984
0.	20	65	6.	—	2.569	0.875	—	57.	763	1058
0.	10	65	2.32	2.416	2.70	0.875	—	57.	320	836
0.	10	65	2.32	6.950	2.70	0.875	—	57.	468	1205
0.	10	65	3.17	3.859	2.70	0.875	—	57.	468	1279
0.	10	65	3.17	6.360	2.70	0.875	—	57.	541	1426
2.337	10	65	4.	—	2.569	0.875	1.293	57.	394	1426
7.011	10	65	4.	—	2.569	0.875	1.293	57.	468	1500
11.685	10	65	4.	—	2.569	0.875	1.293	57.	468	1648
21.033	10	65	4.	—	2.569	0.875	1.293	57.	541	1721
2.337	10	65	6.	—	2.569	0.875	1.293	57.	910	—
11.685	10	65	6.	—	2.569	0.875	1.293	57.	910	—
21.033	10	65	6.	—	2.569	0.875	1.293	57.	984	—
2.788	10	65	4.	—	2.569	0.875	0.1786	57.	394	1574
8.364	10	65	4.	—	2.569	0.875	0.1786	57.	468	1574
13.940	10	65	4.	—	2.569	0.875	0.1786	57.	541	1648
25.092	10	65	4.	—	2.569	0.875	0.1786	57.	615	1721

Table 1: (cont.) Gas/Liquid/Solid Fluidization Results for 60° Axial Impeller

Q_g (std cc/mm)	H_b (mm)	H_t (mm)	d_p (mm)	L_p (mm)	ρ_p (g/cc)	ρ_t (g/cc)	$\rho_g \times 10^3$ (g/std.cc)	μ_t (cp)	S_1 (rpm)	S_2 (rpm)
2.788	10	65	6.	—	2.569	0.875	0.1786	57.	836	—
13.940	10	65	6.	—	2.569	0.875	0.1786	57.	836	—
25.092	10	65	6.	—	2.569	0.875	0.1786	57.	910	—
0.	10	65	3.	—	2.569	0.867	—	120.	246	689
0.	10	65	4.	—	2.569	0.867	—	120.	320	910
0.	10	65	5.	—	2.569	0.867	—	120.	468	1205
0.	10	65	6.	—	2.569	0.867	—	120.	615	1500
0.	10	65	3.10	—	8.424	0.867	—	120.	615	—
0.	10	65	4.67	—	8.254	0.867	—	120.	1205	—
0.	6	65	3.	—	2.569	0.867	—	120.	246	1058
0.	6	65	4.	—	2.569	0.867	—	120.	320	1500
0.	20	65	4.	—	2.569	0.867	—	120.	320	615
0.	20	65	5.	—	2.569	0.867	—	120.	468	836
0.	25	65	6.	—	2.569	0.867	—	120.	615	763
0.	10	65	2.32	2.416	2.70	0.867	—	120.	173	615
0.	10	65	2.32	6.950	2.70	0.867	—	120.	320	1058
0.	10	65	3.17	3.859	2.70	0.867	—	120.	246	984
0.	10	65	3.17	6.360	2.70	0.867	—	120.	320	1131
2.337	10	65	4.	—	2.569	0.867	1.293	120.	320	984
7.011	10	65	4.	—	2.569	0.867	1.293	120.	320	1058
11.685	10	65	4.	—	2.569	0.867	1.293	120.	394	1205
21.033	10	65	4.	—	2.569	0.867	1.293	120.	468	1500
2.337	10	65	6.	—	2.569	0.867	1.293	120.	615	1500
11.685	10	65	6.	—	2.569	0.867	1.293	120.	652	1574
21.033	10	65	6.	—	2.569	0.867	1.293	120.	689	1648

Table 1: (cont.) Gas/Liquid/Solid Fluidization Results for 60° Axial Impeller

Q_s (std cc/mm)	H_b (mm)	H_t (mm)	d_p (mm)	L_p (mm)	ρ_p (g/cc)	ρ_t (g/cc)	$\rho_g \times 10^3$ (g/std.cc)	μ_t (cp)	S_1 (rpm)	S_2 (rpm)
2.788	10	65	4.	—	2.569	0.867	0.1786	120.	320	1058
8.364	10	65	4.	—	2.569	0.867	0.1786	120.	394	1205
13.940	10	65	4.	—	2.569	0.867	0.1786	120.	431	1205
25.092	10	65	4.	—	2.569	0.867	0.1786	120.	468	1279
2.788	10	65	6.	—	2.569	0.867	0.1786	120.	615	1648
13.940	10	65	6.	—	2.569	0.867	0.1786	120.	689	1795
25.092	10	65	6.	—	2.569	0.867	0.1786	120.	763	1943
0.	10	65	3.	—	2.569	0.845	—	245.	90	394
0.	10	65	4.	—	2.569	0.845	—	245.	135	468
0.	10	65	5.	—	2.569	0.845	—	245.	246	541
0.	10	65	6.	—	2.569	0.845	—	245.	320	836
0.	10	65	3.1	—	8.424	0.845	—	245.	394	—
0.	10	65	4.67	—	8.254	0.845	—	245.	763	—
0.	6	65	3.	—	2.569	0.845	—	245.	98	689
0.	6	65	4.	—	2.569	0.845	—	245.	172	984
0.	20	65	4.	—	2.569	0.845	—	245.	173	320
0.	20	65	6.	—	2.569	0.845	—	245.	394	689
0.	10	65	2.32	2.416	2.70	0.845	—	245.	101	394
0.	10	65	2.32	6.950	2.70	0.845	—	245.	173	541
0.	10	65	3.17	3.859	2.70	0.845	—	245.	136	541
0.	10	65	3.17	6.360	2.70	0.845	—	245.	246	763
2.337	10	65	4.	—	2.569	0.845	1.293	245.	135	541
7.011	10	65	4.	—	2.569	0.845	1.293	245.	173	615
11.685	10	65	4.	—	2.569	0.845	1.293	245.	246	689
21.033	10	65	4.	—	2.569	0.845	1.293	245.	246	763

Table 1: (cont.) Gas/Liquid/Solid Fluidization Results for 60° Axial Impeller

Q_g (std cc/mm)	H_b (mm)	H_t (mm)	d_p (mm)	L_p (mm)	ρ_p (g/cc)	ρ_t (g/cc)	$\rho_g \times 10^3$ (g/std.cc)	μ_t (cp)	S_1 (rpm)	S_2 (rpm)
2.337	10	65	6.	—	2.569	0.845	1.293	245.	320	984
11.685	10	65	6.	—	2.569	0.845	1.293	245.	394	1131
21.033	10	65	6.	—	2.569	0.845	1.293	245.	394	1353
2.788	10	65	4.	—	2.569	0.845	0.1786	245.	173	541
8.364	10	65	4.	—	2.569	0.845	0.1786	245.	246	615
13.940	10	65	4.	—	2.569	0.845	0.1786	245.	246	615
25.092	10	65	4.	—	2.569	0.845	0.1786	245.	320	689
2.788	10	65	6.	—	2.569	0.845	0.1786	245.	320	984
13.940	10	65	6.	—	2.569	0.845	0.1786	245.	320	1058
25.092	10	65	6.	—	2.569	0.845	0.1786	245.	394	1131
0.	10	50.	4.	—	2.569	0.867	—	120.	615	1795
0.	10	55.	4.	—	2.569	0.867	—	120.	468	1352
0.	10	65.	4.	—	2.569	0.867	—	120.	320	910
0.	10	75.	4.	—	2.569	0.867	—	120.	283	910
0.	10	85.	4.	—	2.569	0.867	—	120.	246	836
0.	10	50.	6.	—	2.569	0.867	—	120.	1205	—
0.	10	60.	6.	—	2.569	0.867	—	120.	763	—
0.	10	65.	6.	—	2.569	0.867	—	120.	615	1500
0.	10	70.	6.	—	2.569	0.867	—	120.	468	1500
0.	10	65.	3.1	—	8.424	0.845	—	245.	394	—
0.	10	75.	3.1	—	8.424	0.845	—	245.	394	1795
0.	10	65.	4.67	—	8.254	0.845	—	245.	763	—
0.	10	85.	4.67	—	8.254	0.845	—	245.	763	—

Other groups might possibly arise due to surface tension and gas phase viscosity. However, previous results for the radial flow impeller indicated that surface tension was not a significant variable (7). Also, the viscosities of helium and air are almost identical at the conditions of these experiments. Further experimentation is therefore required to test for this effect. As described in the previous report, nonspherical particles are treated in terms of an equivalent spherical diameter and a particle shape factor.

Using the data of Table 1, correlations of the type:

$$Re = fn \left\{ Ga, Fr, \frac{\rho_p - \rho_\ell}{\rho_\ell}, \frac{\rho_g}{\rho_\ell}, \frac{d_p}{d_i}, \frac{H_b}{d_i}, \frac{H_\ell}{d_i} \right\}$$

Were sought for both stirrer speeds S_1 and S_2 . Nonlinear regression on these data provided the following correlation for the stirrer speed at incipient fluidization:

$$Re_1 = 1.823 Ga^{0.7639} \left(\frac{\rho_p - \rho_\ell}{\rho_\ell} \right)^{-0.1244} \left(\frac{d_p}{d_i} \right)^{-0.9881} \left(\frac{H_\ell}{d_i} \right)^{-2.566} \\ \times [1 + 10.58 Fr^{0.3440}]$$

Experimentally it was observed that neither gas density nor rest bed weight had a significant effect on S_1 . A similar analysis on the S_2 results provided:

$$Re_2 = 0.6148 Ga^{0.7410} \left(\frac{\rho_p - \rho_\ell}{\rho_\ell} \right)^{0.2127} \left(\frac{d_p}{d_i} \right)^{-1.383} \left(\frac{H_b}{d_i} \right)^{-0.6415} \left(\frac{H_\ell}{d_i} \right)^{-1.889} \\ \times [1 + 10.82 Fr^{0.3038}]$$

Again, gas density does not enter into the correlation, but rest bed height is significant.

Parity plots for these two correlations are presented in Figures 4 and 5. It is apparent from an inspection of these plots that the predictions for S_2 are less precise than are the S_1 predictions. Experimental uncertainties are likely responsible for the relative scatter about the two correlations. Since, it was experimentally much more difficult to locate S_2 as compared to S_1 .

Some S_2 entries in Table 1 are omitted. In these cases particle carryover was never achieved. The bed expanded with increasing stirrer speed up to a point, and thereafter no further expansion of the bed was observed. A figure demonstrating this behavior was presented in the last report (7). At about the point corresponding to cessation of the bed expansion, cavitation was observed in the impeller. Experimentally it was not difficult to observe the onset of cavitation, and so a correlation was developed for the critical stirrer speed, S_3 , at which cavitation is first observed. A preliminary analysis has provided:

$$Re_3 = 0.3071 Ga^{0.5137} \left(\frac{\rho_p - \rho_l}{\rho_l} \right)^{-0.5013} \left(\frac{d_p}{d_i} \right)^{-1.543} \left(\frac{H_l}{d_i} \right)^{1.801} \\ \times [1 - 27.03 Fr^{0.3742}]$$

Note the strong positive dependence of S_3 (or Re_3) on the liquid depth, H_l . Without sufficient liquid depth above the impeller, gas is sucked into the inlet and circulation is limited. This behavior is, however, quite unlike that observed with the radial flow impeller where circulation ceases when the blades come into contact with gas. A parity plot for the S_3 correlation is presented in Figure 6.

The correlations for S_2 and S_3 can be used to predict whether or not particle carryover can be achieved. When the calculated value of S_3 is less than S_2 cavitation commences before the bed is expanded com-

Figure 4. Parity plot for S_1 correlation. 60° axial impeller.

Figure 5. Parity plot for S_2 correlation. 60° axial impeller.

Figure 6. Parity plot for S_3 correlation. 60° axial impeller.

pletely and particle carryover is not observed. In every case where this is indicated in Table 1, i.e., where the (-) appears under the S_2 heading, the calculated value of S_2 exceeds S_3 . This calculation is relevant because the data which form the basis of the S_3 correlation are separate from the Table 1 data.

Preparation of a Coal Paste (T. Zhang):

The feed to our reactor will consist of three components: 1) a coal paste, 2) a "make-up solvent" and 3) hydrogen gas. These will be combined in the feed line prior to entering the reactor. The make-up solvent is a liquid at room temperature and is fed to the reactor by means of a liquid chromatography pump. It may be a hydrogen deficient material like raw creosote oil. This would have the desirable effect of placing a considerable demand on the catalyst making it relatively easy to observe changes in catalyst activity. On the other hand, should it prove necessary to moderate the heat effect, it may be desirable to use a partially hydrogenated material like tetralin as the make-up solvent.

The coal paste will be extruded into the reactor feed line by means of a high pressure syringe pump. Our target was to prepare a paste having roughly the consistency of toothpaste at ambient temperature. This paste was to consist of 1 part of dry coal and 1 part of coal derived distillate + residue obtained from the Wilsonville pilot plant. By combining with an additional 1 part of make-up solvent a 2/1 solvent/coal ratio is achieved.

The coal chosen for this study is from the Arco Black Thunder mine in Wyoming and the distillate and residue materials were derived from this same coal. In particular the residual material is deashed resid from the KM unit (S/N 99672) and the distillate is 650°F⁺ distillate

solvent (S/N 99673). Both samples were collected on September 25, 1989.

A coal paste having good extrusion properties was prepared as follows: Both the coal and resid are ground and screened to $< 210 \mu\text{m}$. The coal is dried at 104°C for approximately $1\frac{1}{2}$ hours to constant weight. Five parts of distillate oil are first mixed with one part of resid and the resulting material is combined with six parts of pulverized dry coal (by weight). The coal paste prepared in this manner does not appear to settle over time and is easily extruded. The composition by weight of the coal paste is:

Dry Coal	50.0%
Distillate	41.7
Residue	<u>8.3</u>
	100.0

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