



STUDY OF EBULLATED-BED FLUID DYNAMICS FOR H-COAL. QUARTERLY PROGRESS REPORT NO. 3, JANUARY 1, 1981-MARCH 31, 1981

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STUDY OF EBULLATED-BED FLUID DYNAMICS FOR H-COAL

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انها وروبه الی . ۹- ده او او چه در سر ارد آراف برده یا . ۱۹ و از فروه مودن از دورکوه دارد از مانه ۲۰۰۰ . ۱۹ و از فروه مودن از دورکوه دارد از مانه ۲۰۰۰ .

QUARTERLY PROGRESS REPORT NO. 3 JANUARY 1, 1981-MARCH 31, 1981

R. J. SCHAEFER, D. N. RUNDELL

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FOREWORD

The H-Coal process, developed by Hydrocarbon Research, Incorporated (HRI), involves the direct hydroliquefaction of coal to low-sulfur boiler fuel or synthetic crude oil. The 200-600 ton-per-day H-Coal pilot plant is being operated next to the Ashland Oil, Incorporated, refinery at Catlettsburg, Kentucky, under DOE contract to Ashland Synthetic Fuels, Incorporated. The H-Coal ebullated bed reactor contains at least four discrete components: gas, liquid, catalyst, and unconverted coal and ash. Because of the complexity created by these four components, it is desirable to understand the fluid dynamics of the system. One objective of this program is to apply the results of prior cold flow model experiments (1) to the operating H-Coal PDU reactor in Trenton, New Jersey. Studies are also planned to examine the coalescence behavior of gas bubbles in three-phase ebullated beds.

The work to be performed is divided into four parts: fluid dynamics measurements on the PDU reactor, gas bubble coalescence studies at Northwestern University, cold flow and mixing tests at Amoco's Naperville Research Center, and model implementation. The objective of this quarterly progress report is to outline progress in the first two areas.

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SUMMARY

Characterizations of liquid samples from PDU Run No. 10 were performed. Densities, solids content, and gamma-ray mass absorption coefficients were measured for 13 slurry mix tank samples. Densities and mass absorption coefficients were obtained for an equal number of clean oil tank samples. Battelle Institute completed viscosity measurements on 14 reactor liquid product samples; however, recent modifications to the viscometer will require that some measurements be repeated. Spent Amocat I-A catalyst was obtained for future cold flow tests at Amoco.

Cold flow experiments were started at Northwestern University. Some problems with gas foam formation were noted. Two different probes were fabricated.

INTRODUCTION

The fluid dynamics of the H-Coal reactor have been previously studied in a cold flow unit. Reference 1 provides details of the construction of the unit and results of tests with a variety of gases, liquids, and catalyst sizes. A semi-theoretical model was developed to predict the volume fractions occupied by the gas, liquid, and catalyst phases. The aims of this new contract are fourfold:

- 1) The model developed using cold flow unit test results will be extended to apply to the operating H-Coal PDU reactor.
- Because gas bubble dynamics are crucial in determining the nature of the flow, studies of bubble flow will be performed at Northwestern University using optically clear beds.
- 3) Liquid mixing tests will determine the residence time distribution of liquid in the reactor. Under the previous contract, it was determined that the coal char fines (simulating the unreacted coal and ash) were uniformly distributed throughout the bed. Hence, the measurement of liquid data is essential for modeling the residence time and kinetic parameters associated with the unreacted coal.
- 4) The model will be implemented into a readily usable format.

DATA COLLECTION

HRI PDU Fluid Dynamics Study

<u>Viscosity of PDU Reactor Liguid</u>.--Of the 16 PDU reactor liquid product samples taken during PDU-10, Samples 1, 2, 3, 7, and 8 were tested by Battelle in January. Excessive data scatter was noted in these experiments, and Battelle

made several modifications to improve their traveling bob viscometer. After the modifications were completed, viscosity and density measurements of ten PDU liquid product samples (1, 3, 4, 5, 6, 11, 12, 14, 15, 16) were obtained. Bomb No. 13 was empty. Battelle's third through sixth monthly progress reports are reproduced in Appendix A.

<u>Properties of Clean Oil Tank and Slurry Mix Tank Samples</u>.--Densities, coal and ash concentrations, and mass absorption coefficients for clean oil tank and slurry mix tank samples were obtained. Specific gravities were measured at 20°C or 60°F using a pycnometer in ASTM Method D-1480. Attempts to measure densities at higher temperatures were hampered by gas evolution from the samples above 100°C. Slurry mix tank samples were filtered through a millipore filter to remove suspended coal and ash. The liquid recovered was designated "oil." Solids remaining on the filter were washed with THF to remove any soluble material. Remaining solids were combusted at 775°C to measure ash content.

Mass absorption coefficients were measured for all clean oil tank and slurry mix tank samples. The experimental apparatus is sketched in Figure 1. The gamma-ray absorption of the sample combined with comparable measurements through the empty can or a can filled with water or kerosene allows calculation of mass absorption coefficients.

<u>Amocat I-A Catalyst</u>.--Fifty pounds of Co-60 tagged Amocat I-A catalyst withdrawn from PDU-10 was obtained from Sandia Laboratories.

Northwestern University Bubble Dynamics Study

Experiments were initiated to study bubble performance in a mixture of Dowanol DPM and diphenyl ether. This combination was selected to match the density and viscosity of H-Coal liquids under operating conditions, as well as the optical refractive index of the glass catalyst cylinders. In this mixture, the gas formed a foam of extremely small bubbles, behavior contrary to that observed in other fluids having similar properties. A search was made for other suitable fluids; a mixture of 75% dipentene and 25% p-cymene was ordered.

A laser probe was constructed to measure the bubble size and velocity distributions in the flow regime where the mean free path between bubbles is on the order of a few millimeters. The electronics for this probe, as well as the conventional impedance probe, are now under construction. Appendix B contains the Northwest in progress reports for this period.

DATA ANALYSIS

HRI PDU Fluid Dynamics Study

<u>Properties of PDU Reactor Liquids</u>.--Only data obtained after Battelle modified the viscometer are reported here. Each slurry may be modeled as a Bingham fluid whose stress-shear relationship is: $T = T_0 + \eta_{pl} \dot{r}$

Values of ${\rm T}_{\rm O}$ and ${\rm n}_{\rm pl}$ at various temperatures and pressures were taken from Appendix A.

The plastic viscosity was correlated against temperature using an Eyring activation energy model:

 $\eta_{pl} = a \exp(-b/T)$

As may be seen in Figures 2-5, this form fits much of the data quite well. Between hydrogen pressures of 2000 and 3000 psia, no significant effect of pressure on viscosity was observed. Correlation of yield stresses (T_0) is less satisfying, as shown in Figures 6-8. Yield stress measurements were often difficult to reproduce, as shown by the dashed line in Figure 6. This line shows a repeat measurement on the No. 3 sample after high-temperature (842^{OF}) viscosity measurements. At this elevated temperature, the sample seemed to be undergoing a decomposition reaction, as evidenced by a declining yield stress. Other samples such as Amoco-1 exhibited a high-temperature yield stress that increased with time, suggesting polymerization or selective loss of light material. Appendix A contains Battelle's data observing this phenomenon.

The density of the slurry was correlated by the conventional expression:

 $\rho = c + dT$

The fit, illustrated by Figures 9-13, is reasonably good over the temperature range of $625^{\circ}-725^{\circ}K$ ($666^{\circ}-846^{\circ}F$).

Values of density, yield stress, and plastic viscosity at $719^{\circ}K$ (835°F) were estimated by graphical interpolation. The results are shown in Table I and are contrasted with ORNL's three characterizations at the same temperature (2). The density agreement is good. Samples Amoco-5 and ORNL-1, taken 9/4/80, show similar stresses at 150 sec⁻¹ shear rate. Agreement between Amoco-6 and ORNL-2, both taken one week later, is not as good, largely due to the higher yield stress T_{\odot} calculated by ORNL's analysis.

<u>Properties of Clean Oil Tank and Slurry Mix Tank Samples</u>.--Data for the slurry mix tank and clean oil tank are summarized in Tables II and III, respectively. Within experimental error, the mass absorption coefficients for each type of liquid are constant, with the exception of the material from 8/5/80 and 9/4/80. Measurements from Battelle suggested that the PDU reactor liquid from 8/5/80 is less dense and less viscous than other samples. These four samples will also be retested.

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PLANS FOR NEXT PERIOD

- 1) Repeat measurement of mass absorption coefficient of clean oil tank and slurry mix tank samples from 8/5/80 and 9/4/80.
- 2) Complete viscosity and density measurements at Battelle.
- 3) Using Battelle characterizations, continue analysis of fluid dynamics data taken during PDU-10.
- 4) Begin cold flow experiments at Naperville with HDS-2A catalyst and coal char/kerosene slurries whose viscosities match those of the PDU liquid products. The original plan proposed use of Amocat I-A; however, the production of this catalyst for Catlettsburg has been deferred.
- 5) Begin literature search and planning for liquid tracer tests.
- 6) Continue studies at Northwestern University.

5

REFERENCES

- I. A. Vasalos, et al., Final Progress Report, "Study of Ebullated Bed Fluid Dynamics for H-Coal," Contract DE-AC05-77ET-10149, February, 1980.
- J. R. Hightower, ORNL Fossil Energy Quarterly Progress Report, October-December, 1980, Section 2, "Coal Conversion Development."

NOMENCLATURE

a	Regression coefficient	N•S/m ²
b	Regression coefficient	oĸ
С	Regression coefficient	g/cc
đ	Regression coefficient	g/cc ^o K
T	Temperature	oĸ
8	Shear rate	Sec ⁻¹
n _{pl}	Plastic viscosity	N•S/m ²
ρ	Slurry density	g/cc
Т	Shear stress in viscometer	N/m ²

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TABLE I

PDU REACTOR LIQUID VISCOSITIES AND DENSITIES

Battelle	т.	(2)					0.33	0.462							-			
ORNL	L*	(2)					0.378	0.761										×
	d	(9)					0.908	0.985			0.882							
ORNL	ταμ	<u>(5)</u>					1.683	3.027			1.730							
	To	(4)					0.1260	0.3067			0.2324							
	d	(3)	0.765		0.880	0.885	0.903	0.942					0.942	0.876	}	0.869	0.929	0.960
Battelle	1 _c u	(2)	1.358		2.295	1.792	2.223	2.449					2.293	1.607	I I	0.770	2.227	2.189
	°.	(1)	0.123		0.145	0.102	0.060	0.095					0.245	0.062	1	0	0.072	0.045
		Date	7/30/80	7/30/80	8/05/80	8/28/60	9/04/80	09/TT/6	ME 08/61/6	9/19/80 PM	08/19/80 FM	9/20/80 AM	9/20/80 PM	9/21/80 AM	9/21/80 PM	9/22/80 AM	9/22/80 PM	9/23/80 AM
	Period Sample	<u>Was Taken</u>	130-93-04A	130-93-04A	130-93-10A	130-93-20A	130-93-27A (B)	13C-93-34A	130-93-41B	130-93-42A	130-93-42A	130-93-42B	130-93-43A	130-93=43B	130-93-44A	130-93-44B	130-93-45A	130-93-45B
		Sample ID	moco-1	-2	τ Ι	14	-5 ORNL-1	-5 ORNL-2	L-	8-	-9 ORNL-3	-10	-11 OFNL-4	-12	-13 (9)	-14	-15	-16

Notes

 N/m^2 interpolated to 719^oK from figures 6-8. 3

mPa.s (cp), interpolated to 7190K from Figures 2-5. (2)

g/cc, interpolated to 719^oK from Figures 9 and 10. N/m^2 from Reference 2. (E)

mPa.s from Reference 2.

g/cc from Reference 2. N/m² at 150 sec⁻¹ shear rate. Amoco-5 taken 9/4/80 at 1830 hours (Period 27B). Sample bomb contained only gas. (3)

DNR/ml 4/15/81

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2

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TABLE II

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SLURRY MIX TANK SAMPLE CHARACTERIZATION

				<u>sol id</u>		:
	Specific	Liquid		Ash Oxide		Mass Absorption
Sample	Gravity,	8 2	сњ	Wt& of	8 THF	Coefficient,
Date	g/cc	011 (1)	Coal (2)	THF Insolubles	Soluble	£t ² /lbm
8/05/80	1.2244	40	48	13.1	12	0.0432
9/04/80	1.2094	EE	, , ,	14.4	1	0.0435
0%/11/80	1.2374	38	ŢŸ	15.2	18	1050.0
MA 21/9	1.174	56	32	12.7	10	0.0405
M4 61/6	1.1781	57	34	13.6	6	0.0397
9/20 AM	1.1943	60	32	14.9	7	0.0392
9/20 PM	1.1800	53	40	10.3	2	0.0383
9/21 AM	1991.1	50	40	12.9	8	0.0396
9/21 PM	1.1925	54	3 6	13.6	4	0.0387
9/22 AM	1.2427	51	40	16.1	ß	0.0400
9/22 PM	1.2460	43	43	15.1	8	0.0401
9/23 AM	1.2504	. 25	47	10.1	19	0.0414
Wd £2/6	1. 2853	48	44	20.6	4	£6E0.0
Water	1.00				.0	0.0432
Note: 1) 2)	% oil determined Defined to be THF	from millipor insoluble.	e filtratio	л.		

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DNR/ml: 2/19/81

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TABLE III

CLEAN OIL TANK CHARACTERIZATION

Date ()	1980)	Specific Gravity (60/60°F)	Mass Absorption Coefficient (Ft2/Thm)
The Case of States			
8/05		0.9540	0.0458
9/04		0.9681	0.0463
9/11		0.9633	0.0425
9/19 A	M	0.9715	0.0421
9/19 P	M	0.9742	0.0424
9/20 A	м	0.9778	0.0417
9/20 F	M	0.9769	0.0414
9/21 A	М	0.9669	0.0416
9/21 P	M	0.9739	0.0410
9/22 A	М	0.9707	0.0416
9/22 P	М	0.9766	0.0425
9/23 A	M	0.9765	0.0411
9/23 F	M	0.9802	0.0411

DNR/ml 3/5/81



To Detector Electronics

















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OWZOHF> - UNOO

-24

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Figure 11



DWZOHH> - UNOO

. -26



0WZ0HF> V\UU



BATTELLE INSTITUTE MONTHLY PROGRESS REPORTS

Columbus Laboratories S King Avenue Columbus, Ohio 43201 Telephone (614) 424-6424 Telex 24-5454

April 3, 1981

Dr. Douglas Rundell AMOCO Research Center Post Office Box 400 Naperville, Illinois 60540

Dear Doug:

This is the sixth monthly report on your program on the viscosity of H-COAL samples. It covers the month of March. The purpose of the program is to determine the rheological properties of H-COAL samples over the temperature range 350 to 450 C. The effect of pressure over the range 2000 to 3000 psi is being examined with only a few of the samples.

During the past month we have made measurements on five of your samples. The results are shown in Table 1 and the plastic viscosities are shown in Figures 1 through 3.

We were unable to obtain sample from Bulb 13. Since gas could be passed downward through the bulb we assume it is empty.

We first transfer enough sample to fill a 300-cm³ autoclave, then, after mixing well, we transfer about 80 g to the viscometer. In the case of AMOCO 14 we did not find enough sample to fill the mixing autoclave, but a normal viscometer charge was obtained. The unused portion was returned to the sample bulb. The sample was easily pourable at room temperature. We made an error in setting the instruments for this determination. The result was that much of the data was obtained at high shear rates. The results are not considered very reliable. We hope to be able to repeat the determination, especially because of the unusually low plastic viscosities apparently observed.

The AMOCO 15 vessel contained more than enough sample to fill the mixing autoclave. The sample poured easily at room temperature. Measurements were made without any unusual difficulties.

We had difficulty transferring AMOCO 16, presumably due to the formation of a plug in one of the valves. The viscometer had a smaller than normal sample, but the measurements were made as usual without difficulty.

Dr. Douglas Rundell

In attempting to transfer AMOCO 3 for a rerun on this sample the viscometer was filled without difficulty but we were unable to return unused sample to the sample vessel. A plug had formed in the transfer line and could not be removed. Furthermore, the viscometer could not be drained. We therefore opened the viscometer and aspirated the sample out the top, later returning it to the sample bulb by the same method. In this case, therefore, the small amount of sample now in the sample bulb has been heated to 450 C in the viscometer and has subsequently been exposed to air. The measurements proceeded without difficulty. Figure 2 shows average values at each temperature. Table 1 shows that the effect of pressure was less than the scatter in the data. Some repeated measurements on this sample (see Table 1) seem to indicate a lowering of plastic viscosity with time.

AMOCO 1 was rerun without unusual difficulties. There was less than 300 cm³ of sample, but enough to provide a standard viscometer sample. At room temperature the sample seemed to be unusually thin.

Results are also shown in Table 1 and Figure 3 for the original measurements on this sample, made in November. We encountered difficulty during the first run because of excessive sample transferred to the viscometer, due, no doubt, to its unusually low viscosity. Some sample got inside the bob. We are not sure that we have adequately accounted for this in treating the data, but the agreement between the two determinations seems satisfactory, given the difficulties encountered early in the program. We show these older results especially because of the apparent thickening taking place at the highest temperature. This could be due to polymerization or to loss of solvent, although we are reasonably confident that very little solvent escapes the apparatus. The apparent effect of time seems to be opposite that observed with AMOCO 16 and AMOCO 3.

We see no reason to question our density results except at 410 K where the system is insensitive for balancing the bob immersed in the liquid.

In most cases the yield stress seems to decline continuously. This generalization includes the results for AMOCO 3, in which case measurements were made at lower temperatures after completing the work at the highest temperature. Viscosities seemed to drop somewhat after heating to 725 K, but the yield stress values dropped dramatically.

There seem to be several indications of changing results, evidently brought about by holding the sample at elevated temperatures. It is difficult to investigate this phenomenon by present procedures, since each measurement is made after holding the sample for varying periods of time at one or several other temperatures. We could introduce a cold sample into a hot viscometer and make measurements at a constant temperature as a function of time. To do this at more than one temperature, however, would require a lot of sample and a lot of time.

Dr. Douglas Rundell

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April 3, 1981

We have been committing the instrument about half time to the program. We should be able to work about one more full month. We expect to spend that time rerunning the samples which have not been reported to you in the last 2 months, as well as AMOCO 14, if possible.

We expect to make some further checks on the effect of time, but do not believe it would be useful to spend any more effort on the effect of pressure.

If you have any questions, requests or suggestions, I would urge you to call me soon at (614) 424-4933.

Very truly yours,

John W. Droege Synfuels Technology Development Section

JWD:drr

Temper	ature	Press	ure	Viscosity, n _{pl}	Yield Stress, To	Density,
K	F	MPa	psia	mPa•s (cP)	N/m ²	kg/m ³
			,	AMOCO 14		
411.2 624.0 647.8 673.4 698.0 724.3	280.4 663.4 706.3 752.4 796.6 844.0	17.24 17.24 17.24 17.24 17.24 17.24	2500 2500 2500 2500 2500 2500	24.7 1.77 1.47 1.18 .89 .74	1.41 .08 .08 .05 .01 .00	1096 978 949 928 897 864
				AMOCO 15		
410.0 626.1 650.0 674.3 697.6 723.3	278.2 667.2 710.2 754.0 795.9 842.2	17.24 17.24 17.24 17.24 17.24 17.24	2500 2500 2500 2500 2500 2500	96.3 6.74 5.24 3.53 2.88 2.13	.76 .38 .26 .21 .18 .06	1190 1031 1002 975 947 928
•				AMOCO 16		·
408.7 623.7 648.2 673.5 698.5 724.0	275.9 662.9 707.0 752.5 797.5 843.4	17.24 17.24 17.24 17.24 1.'.24 1.'.24	2500 2500 2500 2500 2500 2500	59.9 4.92 4.14 3.49 2.54 2.11	.88 .11 .11 .11 .05 .04	1220 1028 1012 994 966 956
				AMOCO 3	·	
409.0 627.0 627.2 627.4 622.7 649.5 650.2 650.0 673.4 673.2 672.8 699.0	276.4 668.8 669.4 669.2 669.6 661.1 709.3 710.6 710.2 752.4 752.0 751.3 798.4	17.24 13.79 17.24 20.69 17.24 13.79 13.79 17.24 20.69 20.69 17.24 13.79 13.79	2500 2000 2500 2500 2000 2000 2500 3000 2500 2000 20	80.2 5.81 5.53 5.60 5.23 5.22 5.00 4.97 4.94 3.58 3.52 3.70 2.95	2.0 .42 .42 .35 .37 .39 .38 .35 .32 .29 .28 .22	995 996 1000 985 993 979 977 973 944 941 945 911

TABLE 1. VISCOUS PROPERTIES OF FIVE SAMPLES

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	Temper K	rature °F	Press MPa	ure psia	Vis	cosity, n _{pl} Pa•s (cP)	Yield Stress, To N/m ²	Density, kg/m ³
		842	17.	24 AMO	CO_3 (Cont'd)		
(a .	699.0	798.4	20.69	- 3000		9.77	19	0.00
	723.2	1798.4	20.69	2500		2.77	.10	900
	699.1	798.6	17.24	2500		2.11		075
	409.6	277.5	17.24	2500		46.3	200	1093
					AMOC	01		
	408-2	275.0	17 24	2500		10 /	20	
	625.5	666.1	79.70	2000		10.4 3 07	.20	1065
	624.9	665.1	17.24	2500		2.07	.05	909
	625.2	665.6	20.69	3000		2.13	•04	907
	648.5	707.5	13.79	2000		2.03	.03	901
	649.2	708.8	17.24	2500		1 07	.00	188
	648.9	708.3	20.69	2000		1 0/	.05	884
	673.2	752.0	20.69	3000		1 77	.04	882
	673.5	752.5	17.24	2500		1 77	.08	828
	673.2	752.0	13.79	2000		1.71	.08	000
	699.2	798.8	17.24	2500		1 73	•0a	002
	724.0	843.4	17.24	2500		1 33	109	020
	673.2	752.0	17.24	2500		1.84	.06	832
				AMOCO	1_(Nov	ember Data)		
	413.4	284.4	17.24	2500		<i>a</i> o	* 0	
	623.5	662.5	13.79	2000		0.0	.13	1033
	624.3	664.0	17.24	2500		1 70	.00	900
	624.8	664.9	20.69	3000		1 70	.02	899
	645.3	701.8	13,70	2000		1 65	.01	897
	645.7	702.5	17.24	2500		1.62	• 1 2	869
	646.2	703.4	20.69	3000		1 / 6	<u>د</u> ا.	,870
	669.2	744.8	13.79	2000		1 91	- <u>-</u>	870
	698.2	797.0	17.24	2500	*	1 55	.12	820
	723.2	842.0	17.24	2500	4	1.28	.00	010 760
	722.7	841.1	17.24	2500	18	1 68	•±0 2/	/02 755
	723.1	841.8	17.24	2500	26	1.87	• 44	700
	723.2	842.0	17.24	2500	34	2.03	.33	750

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TABLE 1. (Continued)

*Time (min) at this temperature.

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FIGURE 1. PLASTIC VISCOSITY OF TWO SAMPLES



FIGURE 2. PLASTIC VISCOSITY OF TWO SAMPLES



FIGURE 3. PLASTIC VISCOSITY OF AMOCO 1



March 12, 1981

Dr. Douglas Rundell AMOCO Research Center Post Office Box 400 Naperville, Illinois 60540

Dear Doug:

This is the fifth monthly report on your program on the viscosity of H-COAL samples. It covers the month of February. The purpose of the program is to determine the rheological properties of H-COAL samples over the temperature range 350 to 450 C. The effect of pressure over the range 2000 to 3000 psi will be examined with only a few of the samples.

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During the past month we have made measurements on four of your samples. The results are shown in Table 1 and the plastic viscosities are shown in Figures 1 and 2.

Samples 5, 11, and 12 were similar in room temperature appearance to most other samples. AMOCO 6 appeared to be definitely thicker, just barely liquid at room temperature. Measurements on AMOCO 5 were made at 13.1 MPa (1900 psi) because of our difficulty in getting delivery of high pressure hydrogen. The other three samples were run at 17.2 MPa (2500 psi). One point for AMOCO 12 (698.5 K) is known to be inaccurate. Our measurement requires that the apparatus be subjected to a constant low amplitude vibration to prevent stick-slip friction from disturbing the results. At this temperature the vibration was insufficient, a condition noted and corrected at the next higher temperature.

Several aspects of the data seem to us to be curious, leading to speculations which we hope to resolve, at least in part, with further work. Some of these are related to what appear to be changes experienced by the samples with time. Our procedures are based on the assumption that such changes will not occur. We therefore have little opportunity, within the scope of the program, to study this point extensively. We will attempt to make such observations as we can in an effort to shed some light on the effect of time at temperature.

The first curiosity is that in several of the measurements, both of plastic viscosity and of density, the point at 620 K (350 C) seems a little low, compared with other points. Since this is the first measurement after raising

March 12, 1981

the temperature well above ambient, it may be an indication of an initial change. We will attempt to check on this point during the coming month.

For several samples the yield stress at the highest temperature seems to show a decided drop over the values observed at lower temperatures. This, again, could be a time-related phenomenon. We would like to repeat some measurements at lower temperatures after completing the series. We will attempt to find occasion to do this.

The densities as estimated at high temperatures do not usually line up well with the value estimated from the observations at 408 K. This is probably due to the difficulties in making the necessary observation when the viscosity and possibly the yield stress are high. It seems strange that the densities should show so much variation from sample to sample. We are not sure what to make of this.

There seems to be a considerable variation from sample to sample in the observed yield stress, as well as rather a lot of variation with temperature for a single sample. Because of the lack of an independent density measurement the yield stress values at the lowest temperature are probably not very reliable. They suffer from the same uncertainties that effect the density estimates at this temperature. For the measurements at higher temperatures we are disposed to believe that the reported values are approximately correct.

One expects the curve of plastic viscosity with temperature, as shown in Figures 1 and 2, to be slightly concave upward. For Samples 5 and 12 this is about what is observed. The failure of Samples 6 and 11 to follow this pattern may be due to time-dependent phenomena, as already suggested, but we cannot be sure.

During the coming month we will complete the measurements on the remaining specimens and, as time permits, repeat some of the older measurements. If there are any questions, do not hesitate to call.

Very truly yours,

John W. Droege Synfuels Technology Development Section

JWD:drr

Attachments

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Dr. Douglas Rundell

2

Tempera	ature	Viscosity, npl	Yield Stress, τ_0	Density, p
K	F	mPa·s (cP)	N/m ²	kg/m ³
		AMOCO 5, 13.	<u>1 MPa H2</u>	
408.2	275.0	73.0		
623.6	662.9	3.49	.10	981
647.4	705.6	2.66	.11	971
673.2	752.0	2.62	.07	946
698.7	797.9	2.38	.06	924
722.7	841.1	2.19	.06	897
		AMOCO 6, 17.	2 MPa H2	
408.5	275.5	197		
623.7	662.9	5.26	.42	1032
647.2	705.2	4.93	40	1021
670.5	747.1	4.46	.32	995
697.4	795.6	3.42	.26	966
723.2	842.0	2.30	.08	938
		AMOCO 11, 17	7.2 MPa H2	
409.7	277.7	31.0	.23	1244
620.2	656.6	4.92	.48	1028
649.4	709.2	4.43	•47	1011
672.7	751.1	3.88	.46	990
697.9	796.9	2.95	.41	965
721.4	838.8	2.23	.23	938
		AMOCO 12, 1	7.2 MPa H2	
408.2	275.0	18.4	.49	1067
~723. 7	662.9	2.81	•08	950
647.7	706.1	2.25	.10	931
672.7	751.1	1.94		(906)
698.5	797.5	(2.05)	.11	890
723.5	842.5	1.58	-06	873

TABLE 1. VISCOUS PROPERTIES OF FOUR SAMPLES

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Temperature, F

FIGURE 1. PLASTIC VISCOSITY OF TWO SAMPLES



Temperature, F

FIGURE 2. PLASTIC VISCOSITY OF TWO SAMPLES

Chiumbus Laboratories 505 King Avenue Columbus, Ohio 43201 Telephone (614) 424-6424 Telex 24-5654

February 6, 1981

Dr. Douglas Rundell AMOCO Research Center Post Office Box 400 Naperville, Illinois 60540

Dear Doug:

This is the fourth monthly report on your program on the viscosity of H-COAL samples. It covers the month of January. The purpose of the program is to determine the rheological properties of H-COAL samples over the temperature range 350 to 450 C. The effect of pressure over the range 2000 to 3000 psi will be examined with only a few of the samples.

We have remarked several times that the scatter in our data at low viscosities has been so high as to obscure the effect of pressure and the differences among the AMOCO samples. We have made a major effort during January to resolve these difficulties and I believe we have had good success.

For our bob to move up and down within the autoclave without friction requires good alignment. We have been relying on a flexible joint which allowed the bob to center itself in the autoclave even if the magnet assembly is not perfectly aligned. During January, we made the following changes:

- 1. A tool was devised which enabled us to align the axis of the magnet housing accurately with the axis of the autoclave.
- 2. One of the guides which kept the magnet assembly running true was removed. This eliminated one possible source of Friction.
- 3. The flexible joint was replaced with a fixed connector.
- 4. The system was calibrated over the range 0.3 to 33 mPA·s (cP).

The calibrations, which are based on the known viscosities of five liquids, are in satisfactory agreement. The scatter in the measurements on coal liquids has been greatly reduced.

Mr. Douglas Rundell

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Since making these modifications, we have measured the viscosity of one more sample. The results are given in Table 1 and shown in Figure 1. We have had trouble obtaining high-pressure hydrogen and so have made these measurements at 2000 psi only. Density is estimated from the same data. We do not expect the viscosities to follow a straight line; therefore the dashed line should not be taken as an indication of viscosity at intermediate temperatures.

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During the coming month, we plan to proceed with measurements on several additional samples.

Please call me at (614) 424-4933 if you wish to discuss this report or our program.

Very truly yours,

John W. Droege Synfuels Technology Development Section

JWD:jc

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Tei	nperature	Plastic Viscosity	Yield Stress,	Density
K	- F	mra's (Cr)	N/Ш~	Kg/ш
407.2	273.2	31.8		·
626.2	667.6	3.65	.18	958
649.2	708.8	3.51	.18	⁻ 942
673.6	. 752.7	2.78	.20	922
697.4	795.7	2.12	.20	896
722.4	840.7	1.74	.09	887

TABLE 1.VISCOUS PROPERTIES OF AMOCO 4AT 13.9 MPa (2000 psi) HYDROGEN

3



FIGURE 1. PLASTIC VISCOSITY OF AMOCO 4 AT 13.9 MPa (2000 psi) HYDROGEN PRESSURE

APPENDIX A



Telex 24-5454

Telephone (614) 424-6424

January 12, 1981

Dr. Douglas Rundell AMOCO Research Center Post Office Box 400 Naperville, Illinois 60540

Dear Dr. Rundell:

This is the third monthly report on your program on the viscosity of H-COAL samples. It covers the month of December. The purpose of the program is to determine the rheological properties of H-COAL samples over the temperature range 350 to 450 C. The effect of pressure over the range 2000 to 3000 psi will be examined with only a few of the samples.

We have now completed measurements on AMOCO Samples 1, 2, 3, 7, 8, and 9. As reported last month, we are experiencing excessive scatter in the data. We have also been having some trouble with obtaining reproducible calibration data. These difficulties are of a different kind but may be related. The calibrating oils give satisfactory data and good straight-line plots with little scatter, but calibrations performed on separate occasions do not yield the same results. We are now attempting to resolve this problem, which is related to the low viscosities at which we are working in this program.

Because of the uncertainty in calibration, we have not yet reduced all the data to produce results in units of viscosity. However, we have done so for sample AMOCO 2 in order to illustrate the data we have been getting. Figure 1 shows the data obtained at 669 F and 2000 psi. The ordinate gives indication of the upward force applied to the bob, expressed in arbitrary units. The abscissa shows the output signal which is proportional to the rate of movement of the bob. The force required to suspend the bob in the sample is related to the mid-point of the two intercepts at zero on the abscissa. This mid-point, which cannot always be precisely located from the data, depends on the density of the liquid. Therefore, an independent determination of the density aids in interpreting the data. Lacking such a determination, an approximate value of the density can be found from these results. The plot approximately covers the range $\pm 150 \text{ s}^{-1}$ in shear rate.

-47 . January 12, 1981

Dr. Douglas Rundell

The data for each determination were treated in this way. Then, assuming a value for the calibration constant, the slopes of the lines for ascending bob were converted to plastic viscosity and the intercepts were used to determine the yield stress. Results are shown in Table 1. Calculated densities, based on the mid-point between the two intercepts, is also shown.

The first point, at about 400 K, has been determined as a way of comparing the samples which may be more sensitive to sample differences than the determinations at higher temperatures. At higher temperatures a general trend toward lower plastic viscosities and toward higher yield stress is evident. The scatter is such that one hesitates to come to any firm conclusion. The pressure effect is evidently within the scatter. Based on previous experience one expects the pressure effect in plastic viscosity to be of about the size and direction corresponding to the 2500 and 3000 psi results at 647 and 648 K. The densities seem reasonable at lower temperatures and appear to be a little low at higher temperatures, but we have little data for comparison.

We have had no further difficulties with the sample transfer procedures. The sample transferred to the mixing autoclave but not used for viscosity measurements has been returned to the shipping bulb. The sample used for our measurements is discarded.

We will be making further efforts during January to resolve our difficulties in calibration and to find ways to reduce the scatter in the measurements. We will proceed with measurements on several more AMOCO samples.

Please call me at (614) 424-4933 if you wish to discuss this report or our program.

Very truly yours,

John W. Droege Fuels and Combustion Section

JWD:drr

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TABLE 1. VISCOUS PROPERTIES OF AMOCO 2

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Tenper	ature	Prese	aure	Plastic Viscosity,	Yield Stress,	Densițy,
K	54	MPa	þei	mPa g (cP)	N/m ²	kg/m ³
405.2	269,6	17.2	2500	13.5	. 29	1102
627.0	668.9	13.8	2000	2.2	•06	922
627.2	669.2	17.2	2500	2.1	.03	928
626.6	668.3	. 20.7	3000	2.0	.07	922
646.0	703.2	13.8	2000	1.6	60 *	868
646.6	704.3	17.2	2500	1.9	.11	895
648.2	707.0	20.7	3000	2.0	.07	896
672.7	751.1	17.2	2500	1.4	.15	862
696.2	793.4	17.2	2500	1.6	.16	832
719.6	835.7	17.2	2500	1.6	.21	795



Power Control Setting, 2

APPENDIX B

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NORTHWESTERN UNIVERSITY MONTHLY PROGRESS REPORTS

4

PROGRESS REPORT ON AMDCO DOE CONTRACT "ON H-COAL FLUID DYNAMICS" December and January, 1981.

1) Experiment

In mid-December our laboratory ventilation system was finally installed, and this enabled us to add the proper amount of diphenyl either to the Dowanol DFM. Upon testing the system with this binary fluid mixture, we immediately found a great difference in the type of bubbly flow we were getting compared to what we saw with only DPM in the system. The average bubble size is now much smaller, so small in fact that even at very low gas flow rates it is impossible to see more than a half centimeter or so through the fluid.

Since the changes in bulk fluid parameters normally associated with determining average bubble size (viscosity and surface tension) changed by only minimal amounts in going from DFM to the binary mixture, some other major parameter had to be involved. A search of the literature on foams, bubble coalescence and stability of thin films was begun with two purposes in mind:

- Understanding of the mechanisms involved in bubble coalescence and break-up
- Determine what could be done to our fluid to allow use of the optical measuring methods

Briefly, what we have found so far is that both coalescence and break-up of bubbles depends strongly on the surface properties of whatever fluid is under consideration. For coalescence this involves the rate of thinning of the fluid film between two bubbles in contact, and for break-up it seems as though it should involve the dynamic rather than the static surface tension. There has been very little work done in this area, and the above statement is

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based partially on observations by people in the medical field working with surfactants taken from the lungs of mammals. In their work with expanding and contracting surfaces, measurements of zero dynamic surface tension during surface contractions are regularly made. The physical reality of a strong correlation between turbulence intensity, rate of contraction of a surface necessary to produce zero surface tension, and average bubble size would seem a certainty.

Assuming the above ideas concerning the importance of dynamic surface tension are true, and going one step further by hypothesizing that fluids with "structured" gas-liquid interfaces would be those that would exhibit a decreased surface tension during contraction, we tested the ideas on one fluid (DPM plus diphenyl ether). Since DPM is a polar, non-polar solvent, it will assume a preferved orientation at a gas-liquid interface. Assuming it is this orientation which produces a structured interface we added a low surface tension solvent which would not have any preferred orientation (hexane) to a test sample of our fluid. The average bubble size resulting from vigorous shaking increased, as had been expected assuming our line of thought was good. This method could not be used in our overall system, however, since the solvents with low enough surface tensions have vapor pressures which are too high to be used.

This left us in a position of either obtaining different fluids which would result in larger bubbles, or else reconsidering our measurement methods. Some new test samples of some chemicals have been ordered, and also a preliminary idea on a method to determine size distributions of very small bubbles is under consideration.

2) Impedance Probe

The impedance probes depend on the principle that the impedance of the gas and liquid phase are different. Therefore, the resistance of the fluid plays an important role in the probe measurement.

The resistance of the liquid mixture (66% DFM, 33% Diphenyl ether) was found to be about $10^{9} \Omega/cm$. In other words, if 15 V is applied to the probe system, there is only 15 x 10^{-6} A current passing through the liquid when the tip of the probe is in the liquid phase. The circuit set up in November was not sensitive enough to detect such a small current. Hence, a new circuit was designed and built. Figure 1 is sketch of this new probe circuit. As shown in that figure, parts (B) and (C) are used to eliminate the noise from power supply, 15 V AC. In Part (A), LF 356 H is used as an amplifier. The gain of the first LF 356 H (from Probe) is 1 V/µA, i.e. there is 1 V output if the current detected is 1 µA. The 20 KΩ variable resistor (on the top of the 2nd LF 356 H) is used to adjust the gain of the second LF 356 H. With 20 KΩ in that variable resistor, we can have the maximum gain, 21, while with no resistance, the gain is 1. Therefore, the maximum gain of the two LF 356 H combined is 21 V/µA.

Single threshold method is adopted to compensate for hydrodynamic and surface tension effects. The threshold can be set by the 20 KO Variable Resistor as shown in the zero adjust circuit. The top curve in Fig. 2 shows an output signal from probe (as in checking point). If the signal is passed through LM 311, in which a threshold level is set, the resultant signal shown in the bottom curve os Fig. 2, is a square wave, which can be used to find the average void fraction.

In order to reduce the hydrodynamic effects introduced by the presence of the probe, a new probe is designed. There are no significant changes in the design in comparison to the old one, only the size of the probe is reduced as shown in Figure 3.





Fig. 2

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PROGRESS REPORT ON AMOCO DUE CONTRACT

"ON H-COAL FLUID DYNAMICS"

February 1981

1) Laser Diagnostic

Work is proceeding along two paths in our efforts to determine bubble size and bubble velocity distribution. Our initial plan was to determine a fluid we could use which would result in bubble sizes which could be measured using holography. A variety of new chemicals were obtained and our first choice among them, dipentene, appears as though it will serve our needs. The fluid ordered from Aldrich Chemicals, is actually a mixture consisting of 75% dipentene and 25% p-cymene. It has the following basic properties:

> $n_d = 1.474$ $\rho = 0.84 \text{ gm/cm}^3$ $\mu = 1 \text{ to } 1.54 \text{ Cp}$ $\sigma = 27.5 \text{ dynes/cm}^2 @ 20^\circ \text{C}$ Vapor Pressure : 1 mm @ 14.0°C Flash Point : about 110°F

The dipentene can be refractive index matched to our pyrex by tempemature adjustment.

We were not able to find much toxicity information in dipentene, other than its effects are thought to be similar to turpentine.

Our second approach to bubble size and bubble velocity measurement is the construction of a probe which should be able to determine bubble size distributions in flows where the mean free path between bubbles is as low as a few

-59 - 2 -

willimeters. The basic idea of the probe is that a stationary, hypothetical line segment of a given length, situated in a bubbly fluid, will randomly intersect bubbles. The length of time that any given bubble "breaks" the line is related to the bubble size, velocity, and the closest approach distance of the bubble's center to the line.

The "line" we will be using will be a laser beam focused down to a diameter of about .080 mm. The distance it passes through the fluid will be chosen such that the odds of two bubbles breaking the beam at once is minimal. By measuring both the transition time that it takes a bubble to break the finite diameter beam, and the length of time that the beam remains broken, an indication of the bubbles size and speed normal to the line is obtained. By recording a sufficient number of such data pairs on the FDP11, we should be able to calculate the bubble size distribution and average speed (normal to the beam) of the bubbles. The mathematics of these calculations are fairly straightforward.

The major assumption which must be made is that the bubbles are spherical. For the small bubbles, which we are currently seeing in our experimental set-up is not a bad assumption. Also, of course, the analysis only applies to bubbles with a diameter less than the beam distance.

Our progress towards implementing this technique is moving along well. The mechanical construction of the probe (actually there are two probes which enter the fluid from opposite sides of the column) is nearly complete and we are now working on the electronics design.

2) Impedance Probe

Real time data recording is pursued using the impedance probe in a bubbly air water column. A tape recorder is used for the real time recording. The recorded data is then analyzed by PDP1-11/36.

PROGRESS REPORT ON AMOCO DOE CONTRACT "ON H-COAL FLUID DYNAMICS"

March, 1981

1) Laser Technique

Work on the probe to determine bubble size distributions is progressing well. The probe has been set up in a test stand and connected to the breadboarded electronic circuit. Using an oscilloscope as the output device we have obtained the expected signal shapes.

The probe is now being set up in the column and the details of the optics necessary for alignment are being ironed out.

For data collection and analysis the department's PDP-11/34 will be used (at least initially). Work has begun on developing the necessary programs and a computer terminal for remote use has been ordered.

2) Impedance Probe

The signal from impedance probe must be processed to compensate for hydrodynamic and surface tension effects. The most common technique is by external thresholding. However, the fluid dynamics of the system we are studying is so complicated that a more sophisticated method of setting the threshold has to be used. As shown schematically in Fig. 1, the threshold is accomplished by signal differentiation. This method has been applied successfully by T. Kobori (1).

The original signal is recorded by a HP 3960 instrumentation tape recorder and interpreted with the aid of a PDP 11/34 computer. At this

moment, the program constructed can just be used to convert analog signal into digital values and to calculate the average void fraction.

Ref. (1) Kobori, T., et al "Application of the Needle Contact Probe to Blowdown Tests" Proceedings of the CSNI Specialists Meeting of Transient Two-Phase Flow, Paris, 1978.

__upper clip level lower clip level (a) Probe signo vp. Clipped signal (c) Differentiated signal (d) Shaped signal

FIGURE 1

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