ENCLOSURE (B) 6

EFFECT OF REACTION TEMPERATURE ON THE HYDROGENATION OF COAL

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LIST OF TABLES AND ILLUSTRATIONS

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SUMMARY

These experiments were carried out to investigate the effect of temperature on the properties and yields of liquid products prepared by high-pressure hydrogenation, and to determine the most suitable range of temperature for the reaction. FUSHUN coal was used in these experiments and the following results were obtained.

(1) Paste oil, when reacted in the presence of Fe_2O_3 and high-pressure hydrogen, showed a considerable conversion and the extent of this conversion varied according to the reaction temperature.

(2) The most effective temperature range was between 420°C and 480°C. The reaction was incomplete below these temperatures, and large amounts of coke-like substances were formed above these temperatures.

(3) The maximum yield of liquid product when FUSHUN coal (moisture content 5.5%) was used, was 53%, (61% on a moisture and ash-free coal basis) under the above-mentioned conditions.

(4) The maximum yield of liquid product was obtained at a reaction temperature of 420°C. Increase of temperature beyond this resulted in secondary decomposition reactions reflected by decrease in yield.

I. DETAILED DESCRIPTION

A. Tests on Coal and Paste Oil

The apparatus and procedure used were the same as described in report No. 3 on "Studies on the Hydrogenation of Coal." (See Figure 1(B)6). The following materials were charged to the autoclave:

Coal: FUSHUN coal pulverized to below 20 mesh and maintained at constant moisture content.
Paste Oil: Low-temperature tar produced from SHINBARD coal by Davidson retort, from which light oil (30% of total) had been topped.
Ferric Oxide and Hydrogen: Fe_2O_3 was a commercial grade, and hydrogen was of 99% purity.

In the first series of experiments the above materials were used in the following proportions:

Coal	100 grams
Tar	50 grams
Fe_2O_3	5 grams
Hydrogen	100 atm. (Initial pressure at 0°C corresponding to about 19 grams by weight)

Experimental results are summarized in Tables I(B)6 through IV(B)6.

B. Tests on Paste Oil Only

In the next series of experiments, tests were made on the hydrogenation of paste oil only. The charge stocks were mixed in the following

proportions:

Tar	100 grams	
Fe_2O_3	10 grams	
Hydrogen	100 atm.	(Initial pressure at 0°C corresponding about 19 grams by weight)

The experimental results are summarized in Tables V(B)6 through VII(B)6.

II. CONCLUSIONS

A. It was shown that at a reaction temperature of 390°C the appearance of the crude oil product did not differ from the original oil, evolution of gas was very low, and much coal remained unreacted. The coal, however, was changed to benzene soluble state during the reaction.

B. At 420°C, dehydration of the coal was almost completed. No increase or decrease of produced water occurred above this temperature.

C. The oil yield reached a maximum at 420°C, amounting to 61.4% of ash- and moisture-free coal after correcting for the conversion of the paste oil.

D. Above 420°C, the viscosity of the crude oil decreased, while the oil yield decreased and gas production increased.

E. At 500°C, the crude oil product had very low viscosity and organic residues with increasing content of benzene insolubles began to separate, while the yield diminished to 38.2% of moisture-and ash-free coal.

F. The experimental data showed that secondary decomposition of liquid and solids took place at the higher reaction temperatures.

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Table I(B)6
REACTION CONDITIONS AND YIELD OF PRODUCTS

Run Number	34	26	71	63	15	72
Reaction Conditions						
Reaction Temp. (°C)	390	420	440	460	480	500
Initial Press. (atm.)	103	100	102	102	101	100
Pressure Drop (atm.)	9	16	24	25	25	21
Time of Preheating (hr-min)	2-10	2-05	1-50	1-50	1-55	1-40
Time of Reaction (hr-min)	1-0	1-0	1-0	1-0	1-0	1-0
Yield of Products (gm)						
Gas	24.4	29.0	33.2	33.9	41.8	40.6
Water	14.3	17.1	14.7	17.5	11.0	16.5
Oil	76.0	98.6	89.1	84.0	75.1	73.9
Solid Residue	60.0	27.5	25.2	25.0	23.9	20.0
Total	174.7	172.2	162.2	160.4	151.8	151.0

Table II(B)6

GAS ANALYSES (VOLUME %)

Run Number	34	26	71	63	15	72
CO_2	0.6	0.4	0.3	0.2	0.4	0.3
C_nH_{2n}	0.1	0.2	0.1			0.1
CO	0.1	0.6	0.7	0.4	1.1	0.7
H_2	98.5	92.6	86.6	85.1	83.6	82.4
C_nH_{2n+2}	0.7	6.2	12.3	14.3	14.9	16.5
Total	100.0	100.0	100.0	100.0	100.0	100.0
No. of C.		1.3	1.2	1.2	1.5	1.3

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Table III(B)6
YIELD AND DISTILLATION OF CRUDE OIL (gm)

Run Number	34	26	71	63	15	72
Yield of Crude Oil						
Crude Oil "A"	154.0	120.0	116.0	119.0	83.0	93.0
Crude Oil "B"		3.7	13.0	7.5	27.0	17.4
Total	154.0	123.7	129.0	126.5	110.0	110.4
Distillation Test						
-180°C		10.0	11.6	12.8	7.8	14.1
180-230°C		6.5	6.8	5.9	8.5	7.5
230-280°C	76.0	15.4	17.7	17.0	17.3	15.1
280-360°C		25.0	21.5	22.1	19.9	17.7
Pitch		38.0	31.5	26.0	21.0	19.0
Total	76.0	94.9	89.1	83.8	74.5	73.4

Table IV(B)6
YIELD OF SOLID RESIDUE (gm)

Run Number	34	26	71	63	15	72
Soluble in Benzene		3.7		0.2	0.6	0.5
Organic Residue	46.7	14.3	13.5	12.9	12.0	12.1
Ash	13.3	13.2	11.7	12.1	11.9	7.9
Total	60.0	31.2	25.2	25.2	24.5	20.5

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Table V(B)6
REACTION CONDITIONS AND YIELD OF PRODUCTS

Run Number	24	29	23	14	74
Reaction Conditions					
Reaction Temp. (°C)	420	440	460	480	500
Initial Press. (atm.)	102	101	100	100	101
Pressure Drop (atm.)	6	5	7	10	14
Time of Preheating (hr-min)	2-15	2-10	2-15	2-55	2-30
Time of Reaction (hr-min)	1-0	1-0	1-0	1-0	1-0
Yield of Products (gm)					
Gas	21.8	23.4	25.3	38.1	33.0
Water	5.1	2.5	3.2	1.0	2.1
Oil	89.8	90.3	88.2	86.8	80.9
Solid Residue	10.2	6.7	7.5	1.0*	1.5*
Total	126.9	122.9	123.2	121.9	117.5

*Runs Number 14 and 74, 5 grams of Fe_2O_3

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Table VI(B)6
GAS ANALYSES (VOLUME %)

Run Number	24	29	23	14	74
CO_2		0.1		0.6	0.2
C_nH_{2n}		0.1	0.4	1.1	0.1
CO	0.2	0.5	0.4	0.4	0.4
H_2	97.7	97.5	95.4	90.7	88.3
C_nH_{2n+2}	2.1	1.8	3.8	7.2	11.0
Total	100.0	100.0	100.0	100.0	100.0
No. of C.			1.0	1.6	1.0

Table VII(B)6
DISTILLATION TEST OF TOTAL OIL (gm)

Run Number	Raw Crude Tar	24	29	23	14	74
-180°C		2.5	5.6	5.4	11.5	11.8
180-230°C	5.8	3.2	7.5	8.8	10.0	11.7
230-280°C	19.7	24.0	25.0	25.0	24.0	24.5
280-360°C	41.2	35.0	32.2	31.3	27.0	20.8
Pitch	33.3	26.1	20.0	19.7	14.3	2.1
Total	100.0	89.8	90.3	88.2	86.8	80.9