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Development of a Technology for Coal Conversion in the Presence of Coal Tar

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Abstract—A new process for the hydrogenation of coal in the presence of wide-cut coal tar was proposed; it involves cavitation treatment, mixing with catalytic additives, and heating the resulting mixture at an elevated pressure in an atmosphere of hydrogen. The yields of hydrocarbon fractions to 300°C and gas condensate were evaluated.

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Large reserves of low-energy brown coal are concentrated in the territory of the People's Republic of China (PRC). However, up to now, they are not widely used as a source material for large-scale chemical conversion into liquid hydrocarbon products to be used as fuel and for other purposes [1]. In this context, the main goal of the coal processing complex consists in an increase in the effectiveness of coal technologies and in the production of liquid hydrocarbons. Researchers who studied hydrocarbons proposed various processes and technologies for increasing the yield of light oil products [2–6].

Prospects for the development of new and the intensification of well-known processes for converting fossil fuel materials are related to the efficient preactivation of source materials, which will make it possible to decrease the expenditure of energy or to obtain more valuable production [7].

Cavitation treatment accelerates the degradation of the organic matter of hydrocarbon raw materials and intensifies the destruction of these materials. Cavitation disrupts a continuous chain by destroying bonds between molecular moieties and influences changes in the intrinsic viscosity, that is, the temporary rupture of van der Waals bonds. In the course of the pulse cavitation treatment of oil and petroleum products, energy released during the collapse of cavitation bubbles is used for the rupture of chemical bonds between atoms in the bulky molecules of hydrocarbon compounds [8]. The subsequent hydrofining with the use of catalytically active additives wide-cut coal tar makes it possible to obtain a larger yield of light and medium fractions, as compared with that upon classical hydrogenation processing [9].

Here, we report data on a study of the joint application of a preliminary cavitation treatment and the H-donor properties of wide-cut coal tar in the hydrogenation conversion of the organic matter of coal.

EXPERIMENTAL

Brown coal obtained at the northern foot of the Tien Shan Mountain in Xinjiang (PRC) served as a test material. Tables 1 and 2 summarize the proximate and

Table 1. Proximate and ultimate analyses and mineral composition of brown coal

Proximate analysis				Ultimate analysis, % on a *daf* basis		Mineral composition of coal, wt %			
S_t^d, %	0.31	FC_d, %	65.27	C	82.68	SiO_2	26.76	MgO	2.16
M_{ad}, %	3.08	CRC	3.00	H	4.54	Fe_2O_3	19.88	SO_3	7.08
A^d, %	4.02	Q_{grad}, MJ/kg	31.66	O	12.02	Al_2O_3	25.35	K_2O	0.88
V^{daf}, %	34.01	H/C	0.67	N	0.76	TiO_2	1.78	Na_2O	1.68
						CaO	8.18	SiO_2	6.25

Table 2. Petrographic constituents of Chinese brown coal

Vitrinite, %	Exinite, %	Inertinite
18.5	0.0	81.5

Table 3. Chemical composition of the catalyst

Mineral	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	MnO	FeO
ω, %	5.00	0.50	65.00	1.00	0.7	0.1	26

ultimate analyses and petrographic constituents of this brown coal.

The hydrogenation experiments were carried out in an autoclave; the volume of a chemical reactor was 0.5 l. A powder-like metallurgical waste of size 0.1 mm (according to screen analysis) was used as a catalyst; Table 3 summarizes the composition of this catalyst.

Coal was subjected to catalytic hydrogenation, and the catalyst and wide-cut coal tar (20%) as a hydrogen donor were added. The elemental composition of the coal tar was the following (%): C, 91.1; H, 7.5; S, 0.2; N, 0.5; and O, 0.7. The coal tar density was $d_4^{20} = 0.9341$ g/cm^3. The fractional composition of the coal tar was published elsewhere [10]; the parent tar did not contain tetralin. The coal tar was not subjected to preliminary dehydration, and its moisture content was 10.04%. The purity of tetralin (commercial) was 99.9%.

The cavitation pretreatment of the coal tar was performed using rotor-pulsation cavitation [10] under the following conditions: $t = 40$°C, $\tau_{tr} = 5$ min, and $\nu_{rot} = 300\,000$ rpm.

Table 4 summarizes treatment process conditions in more detail.

RESULTS AND DISCUSSION

The experiments performed confirm the effectiveness of the use of metallurgical wastes for the liquefaction of Chinese coals in a hydrogen-donor solvent. They are comparable to a commercial alumina–cobalt–molybdenum catalyst, which is used in oil processing, in terms of activity.

In this case, the process of coal hydrogenation in a hydrogen-donor solvent occurs by an indirect catalysis mechanism: the liquefaction of coal is predominantly accomplished by hydrogen atom transfer from organic solvent molecules (in this case, wide-cut coal tar) rather than by molecular hydrogen. In this case, the catalytic additives simultaneously restore the proton-donor properties of the tar. It is likely that hydrogen donor carriers prevent the occurrence of the condensation reaction of coal component associates as the temperature in the reaction system is increased.

The results of the experiments on the cavitation–hydrogenation processing of coal (Table 5) showed that, with the use of a solvent (tetralin or wide-cut coal tar) (see no. 2), the yield of liquid products with boiling points of <300°Cwas 35.7%, the total volume of released gas was 36.25%, and the solid fraction was 28.1%. However, an increase of the amount of sulfur as a constituent of the catalyst leads to an increase in the yield of light fractions; it is likely that sulfur increases

Table 4. Experimental conditions (reactor volume, 0.5 l)

Experiment no.	τ, min	T_{set}, °C	P, MPa	H_2 : CO	Coal		Catalyst, g	S*, g	Total		Solvent or paste-forming agent			
					m, g	m^{daf}, g			%**	% on a *daf* basis	coal tar, g	tetralin, g	S : C	S : (C^{daf})
1	60	430	6.0	1 : 0	18.00	15.41	0	0	0.0	0.0	0	0	0 : 1	0 : 1
2	60	430	6.0	1 : 0	18.20	15.57	0.72	0.18	4.9	5.8	0	0	0 : 1	0 : 1
3	60	430	6.0	1 : 0	18.05	15.45	0.86	0.22	6.0	7.0	0	18.00	1.0 : 1	1.2 : 1
4	60	430	6.0	1 : 0	18.00	15.40	1.00	0.25	6.9	8.1	26.8	0	1.5 : 1	1.7 : 1
5	60	430	6.0	1 : 0	18.00	15.40	1.15	0.29	8.0	9.3	27.85	0	1.5 : 1	1.8 : 1
6	60	430	6.0	1 : 0	18.02	15.42	1.44	0.36	10.0	11.7	27.00	0	1.5 : 1	1.8 : 1
7	60	430	6.0	1 : 0	18.01	15.41	0.42	0.11	3.0	3.4	26.68	0	1.5 : 1	1.7 : 1
8	60	430	6.0	1 : 0	18.00	15.40	0.29	0.07	2.0	2.3	27.00	0	1.5 : 1	1.8 : 1
9	60	430	6.0	1 : 0	18.00	15.40	0.29	0.07	2.0	2.3	28.00	0	1.5 : 1	1.8 : 1

Notes: * Elemental sulfur additive.

** Ratio between the total weights of the catalyst and sulfur on an organic matter of coal basis.

*** Ratio between the total weights of the catalyst and sulfur on a dry coal basis.

Table 5. Experimental results (reactor volume, 0.5 l)

Experiment no.	Fraction of <300°C	Gas			Solid residue	Total	Degree of conversion
		in the course of reaction	in the course of distillation	total			
1	–5.9	22.5	16.8	39.3		100.0	33.40
2	–6.0	26.3	21.4	47.7	33.40	100.0	41.70
3	35.7	13.9	22.3	36.2	41.70	100.0	71.90
4	13.7	34.2	10.5	44.7	71.90	100.9	58.40
5	20.4	28.2	15.6	43.7	58.40	100.4	64.10
6	25.6	18.0	15.6	33.6	64.10	99.0	59.20
7	18.4	29.3	13.6	42.9	59.20	100.2	61.30
8	14.6	36.2	12.9	49.1	61.30	100.6	63.70
9	22.5	20.5	12.7	33.2	63.70	99.3	55.70

the donor properties of the catalyst with respect to hydrogen.

The low yield of fractions to <300°C (see nos. 1 and 2) was likely due to the absence of a paste-forming agent during the refining; in this case, the yield of gas condensate was 39.3-47.7%. The high yield of gas condensate (no. 8) was reached at lower amounts of sulfur in the catalytic mixture.

CONCLUSIONS

Thus, energy put into a system by means of cavitation treatment and wide-cut coal tar with H-donor properties facilitates the deeper chemical modification and destruction of the organic matter of hydrocarbon raw materials and leads to a considerably higher yield of light-oil products. Because of this, the development and application of nontraditional processing methods and alternative donors of hydrogen make it possible to increase the efficiency of well-known technologies in this area.

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