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INVESTIGATION AND PILOT-SCALE PROCESSING OF RUSSIAN COALS

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Russian coals of various grades were processed in a pilot-scale retort. Semicokes obtained from this process were characterized by high specific heat of combustion (26-31 MJ/kg), low sulfur (0.5-0.9 %) and low volatile content (1.4-1.9 %). It was shown that long-flame coals and slightly-caking coals are desirable raw materials for a waste-free process with the aim of producing high-quality low-sulfur liquid and solid fuels.

Test experiments of Russian coals of several grades were carried out during 1996 and 1997 in an experimental retort designed by the Oil Shale Research Institute and erected by AS *Kiviter* [1]. Lean, anthracite, slightly-caking and long-flame coals were investigated. The experiments were carried out in order to evaluate the technical feasibility of processing the coals for production of high-quality oil and semicoke. These products could be usable as liquid and solid fuels. Preliminary investigations were carried out under laboratory conditions.

Characteristic data for the Russian coals is given in Table 1. One can see that the moisture content of the tested coals is quite low. This could be explained by fact that the coals were kept in an industrial warm-room for several days before testing. The coals are considered low-sulfur. The Fischer retort analysis (of 50 g samples) of the slightly-caking and long-flame coals may provide an indication of the coal's ability to produce liquid fuel. Silicone oxides prevail in the ash composition of tested coals, except long-flame coals. Semicokes obtained in the Fischer retort have a high specific heat of combustion - 28-33 MJ/kg and, naturally, a low sulfur content (Table 2). This gives evidence that the semicokes from the tested coals are of interest as high-quality solid fuels. Gases produced during semicoke production, except when using anthracite, have a high calorific value and do not contain much sulfur, i.e. the hydrogen sulfide concentration is low (Table 3).

Table 1. **Properties of Russian Coals**

Indices	Lean coal	Anthracite coal	Slightly-caking coal	Long-flame coal
Investigated coal sample number	1	2	3	4
Moisture of coal tested in experimental retort, %	5.7	6.0	4.3	7.0
Content (dry basis), %:				
Carbon dioxide $(CO_2)^d{}_M$	0.61	0.52	1.04	1.15
Ash A^d	8.20	4.20	10.90	5.30
Organic matter*	91.19	95.28	88.06	93.55
Total sulphur $S^d{}_t$	0.24	0.71	0.26	0.25
Volatile matter (dry basis), %	7.6	2.1	27.2	40.7
Fischer assay product yield (standard retort), %:				
Oil	0.84	—	7.78	12.74
Pyrogenetic water	1.50	4.80	4.10	8.14
Semicoke	97.58	95.10	83.50	71.27
Gas and losses (by difference)	0.08	0.10	4.62	7.85
Heating value (bomb calorimeter):				
MJ/kg	31.74	30.98	29.27	29.52
kcal/kg	7580	7400	6990	7050
Ash composition, %:				
SiO_2	56.31	30.09	52.37	13.44
Fe_2O_3	7.41	16.19	8.47	12.98
Al_2O_3	23.93	22.68	24.50	8.58
CaO	4.16	9.89	4.55	26.64
MgO	1.27	6.57	2.06	16.09
SO_3	2.04	10.27	3.77	15.86
Total	95.12	95.69	95.72	93.59

* Here and later on the organic matter content is equal to: 100 - $(CO_2)^d{}_M$ - A^d.

Table 2. **Characteristics of Semicoke Obtained from Coals in the Fischer Retort**

Indices	Investigated coal sample number (see Table 1)			
	1	2	3	4
Content (dry basis), %:				
Carbon dioxide $(CO_2)^d{}_M$	0.5	0.5	0.6	0.9
Ash A^d	5.7	3.9	5.2	4.5
Organic matter*	93.8	95.6	94.2	94.6
Total sulphur $S^d{}_t$	0.29	0.40	0.30	0.12
Volatile matter (dry basis), %	7.5	1.6	11.4	14.7
Heating value (bomb calorimeter):				
MJ/kg	28.39	32.95	32.53	29.64
kcal/kg	6780	7870	7770	7080

In order to maintain a high quality of semicoke, the coal tests were carried out using conditions without semicoke gasification, that is, reverse gas flow was not induced in the heat transfer zone (in the bottom section of retort). In order to minimize the volatile content of the semicoke, the experiments were carried out under severe thermal conditions.

Table 3. **Yield and Characteristics of Gas* Obtained from Coals in the Fischer Retort**

Indices	Investigated coal sample number (see Table 1)			
	1	2	3	4
Specific gas yield (dry basis), m^3/t	17.7	9.0	32.5	62.4
Content of components, vol. %:				
CO_2	34.8	69.3	20.6	23.2
H_2S	0.4	2.1	0.6	0.4
H_2	10.4	—	6.2	11.6
CO	9.6	4.9	7.5	14.1
C_nH_{2n+2}	41.2	22.6	60.1	44.5
Including:				
CH_4	29.8	19.0	44.8	27.4
C_2H_6	7.8	2.5	11.0	11.6
C_3H_8	2.5	0.8	2.8	3.7
C_4H_{10}:				
n-Butane	0.6	0.2	0.7	0.9
iso-Butane	0.1	—	0.2	0.1
C_5H_{12} - *n*-Pentane	0.4	0.1	0.5	0.6
C_6H_{14} - *n*-Hexane	—	—	0.1	0.2
C_nH_m	3.6	1.1	5.0	6.2
Including:				
C_2H_4	1.2	0.3	1.8	2.2
C_3H_6	1.5	0.5	1.9	2.6
C_4H_8:				
Butene-1	0.5	0.2	0.7	0.8
trans-Butene-2	0.1	—	0.2	0.1
cis-Butene-2	0.1	—	0.2	0.1
C_5H_{10}:				
Pentene-1	0.1	0.1	0.1	0.2
trans-Pentene-2	0.1	—	0.1	0.1
C_6H_{12} - hexene-1	—	—	—	0.1
Total	100.0	100.0	100.0	100.0
Calculated heating value, $\frac{MJ/m^3}{kcal/m^3}$:				
high	$\frac{25.66}{6130}$	$\frac{11.89}{2840}$	$\frac{34.96}{8350}$	$\frac{32.41}{7740}$
low	$\frac{23.45}{5600}$	$\frac{10.89}{2600}$	$\frac{31.94}{7630}$	$\frac{29.73}{7110}$
Density, kg/m^3	1.202	1.572	1.102	1.164

* Here and later on all characteristics of gas are given at 20 °C and 760 mm Hg.

The capacity of the pilot-scale experimental retort was estimated to be between 365 and 557 kg per day. The retorting temperatures for the various coals were more or less similar (Table 4).

As expected, semicokes obtained from the coals in the experimental retort are characterized by high calorific value and low sulfur content (Table 5). The volatile content in the aforementioned semicokes is very low - 1.4-1.9 %. These properties make these semicokes usable for special applications in metallurgy, in addition to their use as solid fuels.

Table 4. **Operating Conditions of Processing Coals in the Experimental Retort**

Indices	Investigated coal sample number (see Table 1)			
	1	2	3	4
Feed coal throughput rate, kg/day	500	365	520	557
Temperature, °C:				
Oil vapours from the retorting zone	284	312	276	265
Heat carrier into retorting zone	805	777	822	873
Underpressure at gas outlets from retorting zone, Pa	820	600	470	760
Specific combustion air, m^3/t	308	—	600	552
Specific consumption of recycle gas for heat carrier preparation, m^3/t	960	1316	922	862

Table 5. **Characteristics of Semicoke Obtained from Coals in the Experimental Retort**

Indices	Investigated coal sample number (see Table 1)			
	1	2	3	4
Moisture content, %	5.6	3.0	8.9	4.4
Content (dry basis), %:				
Carbon dioxide $(CO_2)^d{}_M$	1.3	1.3	1.3	3.7
Ash A^d	10.2	4.8	11.9	15.4
Organic matter	88.5	93.9	86.8	80.9
Total sulphur $S^d{}_t$	0.51	0.54	0.81	0.86
Volatile matter (dry basis), %	1.47	1.42	1.90	1.92
Heating value (bomb calorimeter):				
MJ/kg	28.97	30.73	28.47	26.50
kcal/kg	6920	7340	6800	6330

Oil yields obtained from the slightly-caking and long-flame coals were very low - only 8-11 % of the Fischer assay oil. That is explained by the severe thermal conditions used for the coal processing. The produced oils, without exception, are paraffinic and solidify at room temperature. The sulfur content of these oils is low and they have a high heat of combustion (Table 6). Water removal from the coal-derived oils is very difficult even when heated up to 80 °C.

The pH value of the tar water of coal retorting is high - 8-9. (In comparison, the pH of kukersite shale retorting tar water is 5-6.) The water is contaminated with phenols, including volatile ones, and chlorides, sulfur, and volatile bases and acids (Table 7). Water-soluble phenols were separated from the tar water with ethyl ether and were analyzed using a *Carlo Erba* gas chromatograph. This instrument has a flame-ionization detector and a quarz capillary column 0-1(25 m × 0.32 mm)*. The phenols were determined as the silyl ether derivatives.

Table 6. **Yield and Characteristics of Oils Obtained from Coals in the Experimental Retort**

Indices	Investigated coal sample number (see Table 1)	
	3	4
Yield of oil, %:		
Plant yield:		
Raw coal basis	0.60	1.30
Dry coal basis	0.63	1.40
Yield of Fischer assay oil	8.1	11.0
Density at 20 °C, kg/m^3	1063	1005
Water, %	10.9	4.8
Entrained fines, %	2.6	4.8
Viscosity at 100 °C, $1 \cdot 10^{-6}$, m^2/s	23.0	4.0
Flash point, °C	138	110
Pour point, °C	+23	+11
Coking value, %	22.5	8.3
Phenolic compounds, %	3.5	1.6
Heating value (bomb calorimeter):		
MJ/kg	40.19	40.50
kcal/kg	9600	9690
Initial boiling point, °C	202	207
Distillation, vol. %, at:		
220 °C	2.5	24
240 °C	—	38
260 °C	—	52
280 °C	14.5	62
300 °C	—	70
320 °C	37.0	74
340 °C	—	80
360 °C	50.0	86
Elemental composition (dry basis), %:		
C	85.0	85.2
H	9.7	9.2
S	0.7	0.2
N + O (by difference)	4.6	5.4

* Analyses were carried out by researcher E.V. Aitsen, Oil Shale Research Institute.

Table 7. **Yield and Characteristics of Tar Water Obtained from Coals in the Experimental Retort**

Indices	Investigated coal sample number (see Table 1)			
	1	2	3	4
Specific tar water yield (raw coal basis), l/t	27	115	114	52
pH	7.9	3.7	8.6	9.1
Total phenols, mg/l	1340	786	5180	11600
Volatile phenols, mg/l	325	210	1848	8720
Dry residue, g/l	18.10	16.50	25.87	13.40
Chloride (Cl^-), mg/l	1740	1950	6270	3480
Total sulphur, mg/l	7700	5200	9650	3500
Volatile ammonia, mg/l	7200	2560	8640	5940
Volatile acetic acid, mg/l	5700	1140	13500	2400
COD, gO_2/l	8.00	7.63	18.75	58.75

As seen in Table 8, the water-soluble phenols of coal retorting have elevated concentrations of phenol, *m*- and *p*-cresols, pyrocatechol and methylpyrocatechol.

Table 8. **Characteristics of Water-Soluble Phenols Obtained from Coals in the Experimental Retort, % by weight**

Indices	Investigated coal sample number (see Table 1)			
	1	2	3	4
Monohydric phenols	47.7	47.6	63.5	53.2
Including:				
Phenol	21.6	20.4	34.9	26.7
o-Cresol	6.7	7.5	9.6	6.4
m-Cresol	9.9	9.9	9.2	7.6
p-Cresol	9.5	9.8	9.8	12.5
Dihydric phenols	10.7	10.7	13.8	25.1
Including:				
Resorcinol	1.6	1.1	2.5	1.0
Pyrocatechol	3.6	4.6	6.5	10.8
Methylpyrocatechol	5.5	5.0	4.8	13.3
Not identified phenols	41.6	41.7	22.7	21.7
Total	100.0	100.0	100.0	100.0

Conclusions

Pilot-scale processing of Russian coals was carried out in an experimental retort. The produced semicokes are characterized by high heat of combustion 26-31 MJ/kg (6300-7300 kcal/kg) and low sulfur (0.5-0.9 %) and low volatile matter content (1.4-1.9 % which was 30-40 % of the source coals). This proves that such semicokes can be used as solid fuels and for special applications in metallurgy.

It has been established that vertical retorts operating at relatively severe thermal regimes drive off practically all the volatile products from coal. Low oil yields are obtained as compared with those of Fischer assay. These low levels, 8 % for slightly-caking coals and 11 % for long-flame coals, are caused by the severe conditions of retorting. Coals treated under mild pyrolysis conditions lead to yields close to those achieved in the laboratory with a Fischer assay. Oil obtained from the coals during retorting is paraffinic and solidifies at room temperature. Water removal from coal-derived oils is very difficult, even when heated up to 80 °C. Oils derived from slightly-caking coals and long-flame coals have a good solubility in kukersite-based oils.

Tar water obtained at retorting of coals is characterized by high pH (8 to 9) and when compared with kukersite-based water, the former is strongly contaminated by phenols, sulfur, chlorides, and volatile bases and acids. It is characteristic for the water-soluble phenols to contain oxybenzene, *m*- and *p*-cresols of pyrocatechol and methyl pyrocatechol. These phenols are considered a good raw material for the chemical industry.

Slightly-caking coals and long-flame coals could be used as a basis for a waste-free process to yield high-quality low-sulfur liquid and solid fuels.

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