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Dynamics of Thermal Decomposition of Black Coal from the Tavantolgoy Deposit in Mongolia

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Abstract

Integrated investigation of the thermolysis of black coal from the Tavantolgoy deposit (Mongolia) was carried out. It was discovered that coal decomposes during thermolysis within temperature range 300–500 °C with the evolution of gas, low-melting and low-boiling phases, intensively increases in volume and gets agglomerated. As a result, rather strong porous cake is formed. Its volume is 1.5–2 times larger than the volume of the initial sample. It was shown that coal from the Tavantolgoy deposit (without any binding additives) can be used for briquetting.

Key words: Tavantolgoy deposit, black coal, thermolysis, gas components, low-melting, volatile fractions, baking, porosity, briquetting

INTRODUCTION

Coal from the Tavantolgoy deposit (Mongolia), similarly to coal from the Tuva deposit (Russia), relate to black coal – one of the major kinds of fossil coal [1]. Many researchers at different times considered the Tuva coal basin as a potential source of caking coal promising from the viewpoint of broadening of the raw material basis for coking [2–5]. However, in practice, the efficiency of develop0ment of its explored resources is limited by the high prime cost of mined coal, complicated transportation scheme of its realization and, as a consequence, the absence of export demand [10]. In this connection, it is reasonable to use the black coal of the Tuva deposit and the products of its processing as energy carriers.

At the same time, there are no technologies of preliminary treatment of this kind of coal. In view of the high content of volatile and low-melting components (non-condensable gases, coal-tar resin) coal gets caked during combustion in steam generating units, which is accompanied by substantial chemical underburning. Inhabited localities including the city of Kyzyl are situated in intermountain valleys, where stagnant zones are formed in the atmosphere, especially in winter, with the formation of a kind of inversion roof hindering air mixing and air purification. The absence of circulation in the surface air layer leads to the pollution of air by the products of the incomplete combustion of coal. The situation becomes even worse because of the sorption of many pollutants including carcinogenic polyaromatic hydrocarbons on the surface of soot particles. The concentration of polyaromatic hydrocarbons reaches several grams per kilogram [7]. The use of ecologically safer coal fuel, in particular dry coal briquettes, will allow a decrease in the amount of hazardous emissions into the atmosphere and the damage to environment from coal power engineering. Another problem significant for the region is connected with the necessity to introduce the technologies of integrated energy-chemical coal processing for the purpose of increasing the economic efficiency of the use of coal raw material for obtaining mineral oil, motor fuel *etc*.

All these problems are urgent also for the coal-mining branch of Mongolia, so the works on this subject were initiated by the Mongolian side and are carried out with the coal from the Baganur and Tavantolgoy deposits in Mongolia.

Thermal decomposition of black coal of the Tavantolgoy deposit was studied in [8] by means of DTGA. Samples from two coal beds were studied. One sample (coal bed VIII) was averaged finely ground initial product, while the second sample (coal bed IV) was lump coal. It was established that thermolysis at relatively low temperature (300–400 °C) causes destruction of the organic mass of coal, volatile products are evolved, while solid hydrocarbons undergo subsequent melting and sublimation. The depth and quantitative characteristics of the thermal decomposition of coal during heating are to a high extent determined by its initial aggregate state: the size of particles, preliminary treatment, heating mode, and final temperature. According to the data of thermal analysis, the contents of the gas component in the fine ground and lump initial samples differ almost by a factor of two. This is due to the high diffusion of the gas component from the ground material during storage.

In the present work we carried out integrated investigation of the thermal decomposition of a large-scale lot of black coal from the Tavantolgoy deposit and quantitative determination of the gas, low-melting and condensed components.

EXPERIMENTAL

The studies were carried out with the samples of a large-scale lot (mass about 3 kg) of black coal from the Tavantolgoy deposit. The sample was a mixture of fine and lump coal with different particle sizes.

Thermal analysis (DTGA) was carried out with a MOM-1000 derivatograph (Paulik-Paulik-Erdey, Hungary). The upper temperature limit was 650–700 °C. Heating rate was maintained at a level of 10 °C/min. The weighted portion was (1.0±0.2) g. Experiments were carried out in a quartz crucible with a cap made of foamed corundum, which protects the samples from the effect of environment and does not prevent diffusion of gas components evolved from coal during heating.

Determination of the quantitative and aggregate changes during heating and trapping of the sublimates of coal fractions were carried out using a laboratory set-up composed of a steel reactor in which the crucible with the weighted portion (50–100 g) of the material

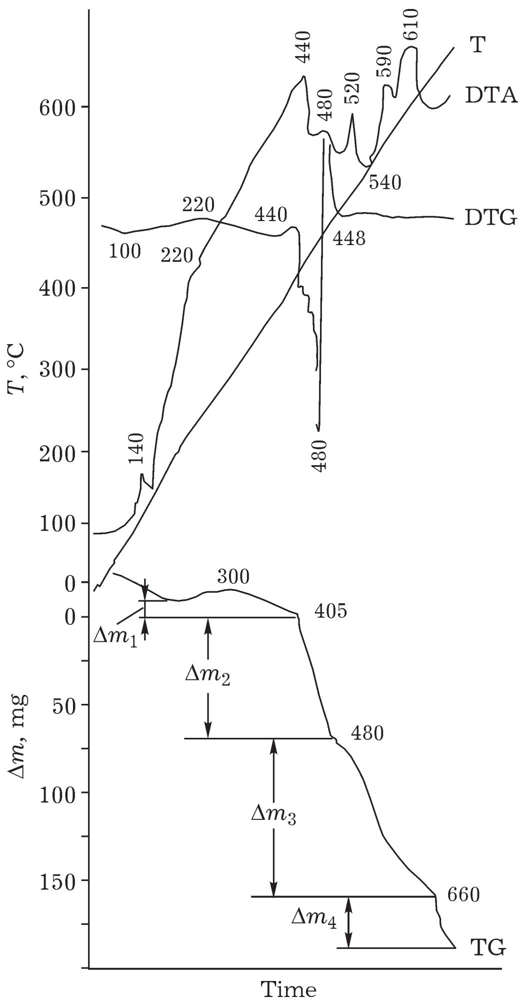

Fig. 1. DTGA of the sample of larger-scale lot of black coal from the Tavantolgoy deposit.

TABLE 1

Results of experiments with the samples of black coal from the Tavantolgoy deposit with heating and exposure (1 h) at 600 °C

Exp. No.	Material	$\Sigma\Delta m$, %	Condensate, %	Gas fraction, %	Notes
1	Average sample	24.6	7.9	16.70	Swelling, sintering
2	The same	25.45	9.3	16.15	The same
3	The same	20.7	8.73	12.00	The same
4	Fine particles from the packet	10.45	8.90	1.60	No swelling; sintering
5	Sample from monolith	20.35	10.09	10.26	Porous cake
6	Averaged*	22.78	9.00	13.78	Swelling, sintering

* Average total over experiments Nos. 1–3, 5.

was placed, and a collector of condensable sublimates, connected with the reactor through a steel tube. The conditions for trapping condensable fractions of sublimates were provided by adjusting rarefaction in the system using a water-jet pump.

RESULTS AND DISCUSSION

Investigation of the dynamics of thermal decomposition of the material by means of thermal analysis (DTGA)

The thermogram of the average sample of the material under investigation characterizing its thermal decomposition is presented in Fig. 1. One can see that the phase and chemical changes taking place in the material have complicated dynamics. Even in the regions of DTA, DTG and TG curves corresponding to a temperature of 100–140 °C, a small effect is observed, accompanied by insignificant mass loss. Then the effect at approximately 300 °C with a weak increase in sample mass (0.5 %) is observed; after that, the mass again decreases insignificantly (by about 1.8 %). Within this temperature range the total mass loss by the sample was 2.4 %.

Subsequent explosion endo effect at 405–440 °C appears on DTG and DTA curves and is characterized by a sharp mass loss on the TG curve (Δm_2 ~ 8.2 %). At this moment, the DTG curve sharply goes down and then also sharply up, after which is reaches its plateau. Subsequent phase transformations appearing at the DTA curve at a temperature of 500, 540, 590 and 650 °C are accompanied by monotonous mass decrease. Within temperature range 480–610 °C, total mass loss reaches 11.0 %, while within the range 610–680 °C it is 3.5 %.

Total mass loss is about 2.5 % of the initial mass. Intense swelling, the formation of porous cake in the upper part of the sample and dense sintering in the bottom part were observed in all the samples.

TABLE 2

Results of experiments with stage-by-stage temperature exposure (2 h) for samples of the Tavantolgoy black coal

T, °C	Exp. No. 1, average sample			Exp. No. 2, monolith sample			Exp. No. 3, average sample		
	$\Sigma\Delta m$, %	Condensate, %	Gas phase, %	$\Sigma\Delta m$, %	Condensate, %	Gas phase, %	$\Sigma\Delta m$, %	Condensate, %	Gas phase, %
200	–	–	–	3.2	0.9	2.3	0.5	~0.5	–
300	2.54	1.1	1.44	1.0	–	1.0	2.3	0.76	1.54
400	7.30	1.9	5.40	10.82	2.94	7.88	8.7	2.0	6.7
450	5.9	1.5	4.4	–	–	–	–	–	–
500	3.4	1.5	1.9	4.8	2.7	2.1	5.8	1.0	4.8
550	1.8	1.3	0.5	–	–	–	–	–	–
600	1.9	1.6	0.25	3.5	2.5	1.0	4.4	2.82	1.58
Σ600	22.84	8.9	13.85	23.32	9.04	14.28	21.74	7.12	14.62

TABLE 3

Integrated data of the experiments on thermolysis of the Tavantolgoy black coal, % of initial mass of the weighted portion

Exp. No.	Sample	T, °C	Time, h	$\Sigma\Delta m$, %	Condensate, %	Gas phase, %	Notes
1	Snap sample	680	–	24.7	–	–	DTGA experiment, swelling
2	Average over mass	600	1.0	24.6	7.9	16.7	Increase in volume by a factor of 1.5–2, porous cake
3	The same	600	1.0	25.45	9.3	16.15	The same, porous bottom cake
4	The same	600	1.0	20.7	8.73	11.27	Emission of gas from reactor at 450–500 °C
5	Average of the loose lot	600	1.0	10.45	8.9	1.55	Increase in volume, sintering
6	From monolith	600	1.5	20.35	10.09	10.26	Increase in volume, porous cake
7*	Average over mass	600	2.0	22.84	8.9	13.94	No increase in volume; dense cake in the bottom part, loose upper layer
8*	From monolith	600	2.0	23.32	9.04	14.28	The same; gas emissions from the reactor at 400–500 °C
9*	Average over mass	600	2.0	21.74	7.12	14.62	Increase in volume, porous cake
10	Averaged	600	–	22.96	8.73	14.23	Excluding exp. No. 5

* Experiments with exposure for 2 h at each temperature interval (see Table 2).

Larger-scale experiments on thermal decomposition of black coal

Results of experiments are presented in Tables 1–3. It follows from the data shown in Table 1 that the mass loss under heating to 600 °C and exposure at this temperature (exp. Nos. 1–3, 5) varies within the range 20.35–25.45 % (with the average value equal to 22.78 %). In exp. No. 4, mass loss is insignificant (10.45 %) and does not correlate with the data of other experiments. This is likely due to the features of the aggregate state of initial material, low content of the gas component in coal. Long-term existence of coal in the form of fine loose grains in contact with the air promotes intense diffusion of the gas component into the environment, as well as chemical and physical weathering of readily oxidized components and the substances removed into the environment.

The amount of the condensate of sublimates of low-boiling fractions was nearly 9 % of the initial product as average (within the range 7.9–10.9). The maximal amount of the condensate (10.09 %) was obtained in exp. No. 5 where the initial sample was prepared from the ground material of lump monolith. At the same time, the data on the mass of condensate for exp. No. 4 (8.9 %) where the initial product was composed of the small particles from the packet of enlarged lot are in good agreement with the data of other experiments.

Results on heating within the general temperature range up to 600 °C with exposure for 2 h at the intervals of 100 °C are presented in Table 2. One can see that thermal decomposition for averaged samples prepared by grinding from the lump material with different particle size and for the sample obtained by grinding of entire monolith proceeds *via* the common mechanism.

Thermal decomposition is intense within temperature range 400–500 °C with the maximal distillation of gaseous and readily volatile coal components: up to 15 % of the initial sample mass, among which the removal of gas component accounts for 10–12 %, that is, 70–80 % of the total mass loss by the sample.

Mass loss by the material at a temperature up to 300 °C reaches almost 3 % for averaged samples and 4 % for the lump sample. The condensate is evolved almost uniformly within all temperature stages: starting from insignificant concentrations (0.5 %) at ≤200 °C and 1.5–3.0 % at each next temperature stage. The total mass loss in all experiments is comparable and equal to 21.74–23.32 % of the initial sample mass. This value is a sum of the amount of condensate from sublimates (within the range 7.12–9.04 %) and gas fraction (within the range 13.85–14.62 %). This series of experiments is characterized by the absence of swelling and the absence of an increase in sample volume. After each stage, starting from 300 °C, sintering was observed in the bottom part of the crucible with the conservation of the upper loose layer.

Results of the experiments on thermal decomposition of black coal lot under study are brought together in Table 3. One can see that the results of all experiments (with stage-by-stage exposure of the material and without exposure) correlate with each other. Exclusion is the data of the experiment with the loose part of the sample (No. 5), where the amount of condensate is comparable with the average values (8.9 %) while the amount of gas fraction is an order of magnitude less (1.6 %). As a result, total mass loss for this sample during its thermal decomposition was 10.45 %, which is smaller than the half of the mass loss for averaged samples and the monolith sample.

A characteristic feature of the behaviour of this type of coal during its thermal decomposition is an increase in sample volume by a factor of 1.5–2.0 during heating in the continuous mode to temperatures ≥600 °C. At the same time, this is not observed during stage-by-stage exposures but sintering of the material in the bottom part of the sample is conserved, likely due to the drainage of liquid fractions during their formation and subsequent heating.

CONCLUSIONS

Investigations showed that thermal decomposition of black coal from the Tavantolgoy deposit is determined by the initial aggregate state of samples, that is, particle size of the initial material, and the final temperature of thermolysis.

Total mass loss during thermal decomposition of the samples averaged by the size of initial material during continuous heating to 600 °C and exposure for 2 h varies within the range 20.35–25.45 %. This value is a sum of the amount of condensate from sublimates (7.90–10.09 %) and the gas component (10.26–16.70 %).

The data on thermal decomposition of the sample prepared from fine, loose part of the material of larger-scale lot sharply differ from those for averaged samples. Though they are close to average values with respect to the amount of condensates of sublimates removed from the samples (8.9 %), with respect to the gas component they are smaller by about an order of magnitude (1.6 %) than the data for coal of this lot. This agrees with the data obtained in experiments with averaged ground sample of coal from bed VIII [8].

The total mass loss by the samples in experiments with stage-by-stage temperature exposure is equal to 21.74–23.32 % and comparable with the data obtained during continuous heating. This value is a sum of the amount of condensate of sublimates (7.12–9.04 %) and the gas component (13.85–14.62 %). Thermal decomposition of coal starts already at a temperature ≤250 °C (0.5 %) and develops intensively at 400–500 °C with mass loss up to 15 % of the initial weighted portion, including almost 10 % related to the gas component.

During continuous heating, sintering and intense swelling of the material occur, its volume increases by a factor of approximately 1.5–2.0. The material is spongy porous product. Its bottom part is denser than the upper one. During stage-by-stage temperature exposures, starting from 300 °C and at all subsequent stages, the material gets sintered in the bottom part while loose structure is conserved in its upper part. Sample volume does not increase.

It follows from these results that black coal from the Tavantolgoy deposit may be used for briquetting (without adding binders) and for obtaining valuable coal-chemical products.

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