# Thermolysis of Brown Coal from the Baganur Deposit (Mongolia)

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## Abstract

Thermolysis of brown coal from the Baganur deposit (Mongolia) was studied. It was established that this coal is distinguished by the high content of gaseous component (up to 40 %) and low content of the liquid fraction (up to 7 %). The decomposition of the material starts at a temperature of about 200 °C. Within the whole heating temperature range (up to 650 °C) the material does not get agglomerated; the final product of thermolysis is loose graphite-like material.

**Key words:** brown coal, Baganur deposit, thermolysis, gaseous component, thermal analysis, agglomeration

## INTRODUCTION

Fossil coal may be subdivided into anthracite, black and brown coal. Depending on the composition, physical and technological properties, as well as economic, industrial, social and other factors, coal from a definite deposit may be used as energy-bearing or technological raw material. In the first case, coal is used to produce electric energy and heat (in the USSR, 73 % of fossil coal was used for these purposes), in the second case coal is used for metallurgic coke, chemical raw material, semi-products for various branches of industry and other areas [1].

For instance, coal from the Tuva coal basin is coking and thus it is a valuable product for metallurgy, coke chemistry and chemical industry [2, 3]. However, due to unfavourable geological and geographic factors, complicated transport conditions, remote position from industrial regions, coal mined from the Tuva deposit is used only as energy-bearing material, without any preliminary treatment.

In view of the high concentrations of volatile components and the formation of cake, coal is burnt out in furnace units not completely, which causes permanent emission of ecologically dangerous products of coal combustion into the atmosphere. Mountainous landscape and extreme climatic conditions aggravate this unfavourable situation (the formation of stagnant zones, smog *etc.*), which leads to substantial pollution of the environment with carcinogenic substances [4].

For the purpose of eliminating this negative situation and providing efficient use of coal from Tuva, thermal decomposition of coal with the removal of gaseous and low-melting fractions and obtaining dry porous material was studied [5–7]. On the basis of the data thus obtained, coal fuel briquettes without gaseous and low-boiling components − low-temperature coke with the developed porous surface − were prepared in the experimental set-up [8].

A similar situation is observed with respect to coal in Mongolia, in particular, coal from the Baganur and Tavantolgoy deposits. In this

connection, it is reasonable to study thermal decomposition of coal from these deposits situated at adjacent territories. Though coal from the Baganur deposit relates to brown coal group, the data on the dynamics of its thermal decomposition may be necessary for the purpose of obtaining ecologically safe fuel product from this coal.

In the present paper we describe the results of the first stage of investigation of the dynamics of thermal decomposition of coal from the Baganur deposit, with the formation of decomposition products for further studies of their chemical composition and technological characteristics.

## EXPERIMENTAL

Brown coal sample from the Baganur deposit (Mongolia) for investigation was obtained as a snap sample, then as a larger-scale lot (with a mass of about 3 kg), which was a mixture of lump material with the particles of different sizes, and fine loose material.

At the first stage, the dynamics of thermal decomposition was studied by means of differential thermal analysis (DTGA) with the help of a MOM-1000 derivatograph (Paulic–Paulic–Erdey, Hungary). Initial material was preliminarily separated into three fractions: the loose (friable) part, medium-sized fraction (5–20 mm) and lump monolith (particles larger than 20 mm). Then grinding was carried out. The weighted portion was (1±0.2) g. The samples were heated to a temperature of 650–700 °C with a rate of 10 °C/min. To prevent the effect of environment on thermolysis, a quartz crucible with the sample was closed with a cap made of foam-corundum, which provided free diffusion of gaseous components formed during coal heating but excluded contact with air.

Mechanical activation of the samples was carried out in AGO-2, a laboratory planetary mill of periodic action, for 1 min.

Further studies aimed at determination of the quantitative characteristics of the process were carried out in a laboratory set-up with trapping gaseous and low-boiling components of coal. The set-up included a steel reactor in which a crucible with a sample was placed. The reactor was connected through a steel socket to a vessel to collect condensable sublimates. To create the conditions for trapping condensable components, rarefaction was provided by a water-jet air pump. The mass of weighted portion prepared for these experiments was 50–100 g. Experiments were repeated to test reproducibility of the results obtained. The mass loss scattering did not exceed 5 rel. %.

## RESULTS AND DISCUSSION

### DTGA of the snap (preliminary) sample

Results of the investigation of thermal decomposition of different non-averaged samples of coal material are presented in Fig. 1 and in Table 1. One can see that along with the general similarity of the results of experiment series, DTGA curves exhibit some differences in the character of some stages of thermolysis for different samples (see Fig. 1). For example, a small endothermic effect at 80–150 °C is observed for DTA curves of the lump samples (fresh and prepared a day before the experiment, experiments Nos. 1 and 2, respectively). However, this effect is almost absent from the DTA curve related to the sample freshly prepared from loose material – experiment No. 3 (see Fig. 1, *c*) but increases sharply in experiment No. 4 with the sample prepared also from loose material but after different time intervals (Fig. 1, *d*). A clear sequence of endo effects is detected in DTA curves for samples in experiments Nos. 1, 2, 4 within temperature range 380–600 °C, corresponding to step-by-step removal of gaseous and low-boiling decomposition products from the material (see Fig. 1, *a*, *b*, *d*). However, in the case of sample No. 2, instead of a sequence of thermal effects, we observe only one large endo peak within temperature range 400–540 °C (see Fig. 1, *c*). This diversity of curve behaviour in the thermograms of the products under study at the initial stage of heating is likely to be connected with the inhomogeneity of the phase composition of samples of the initial material and with differences in particle sizes. Nevertheless, the sequence of thermal decomposition is the same for all samples: the process starts with explosion-like emission of the gaseous component at relatively low temperatures (exp. No. 1 –
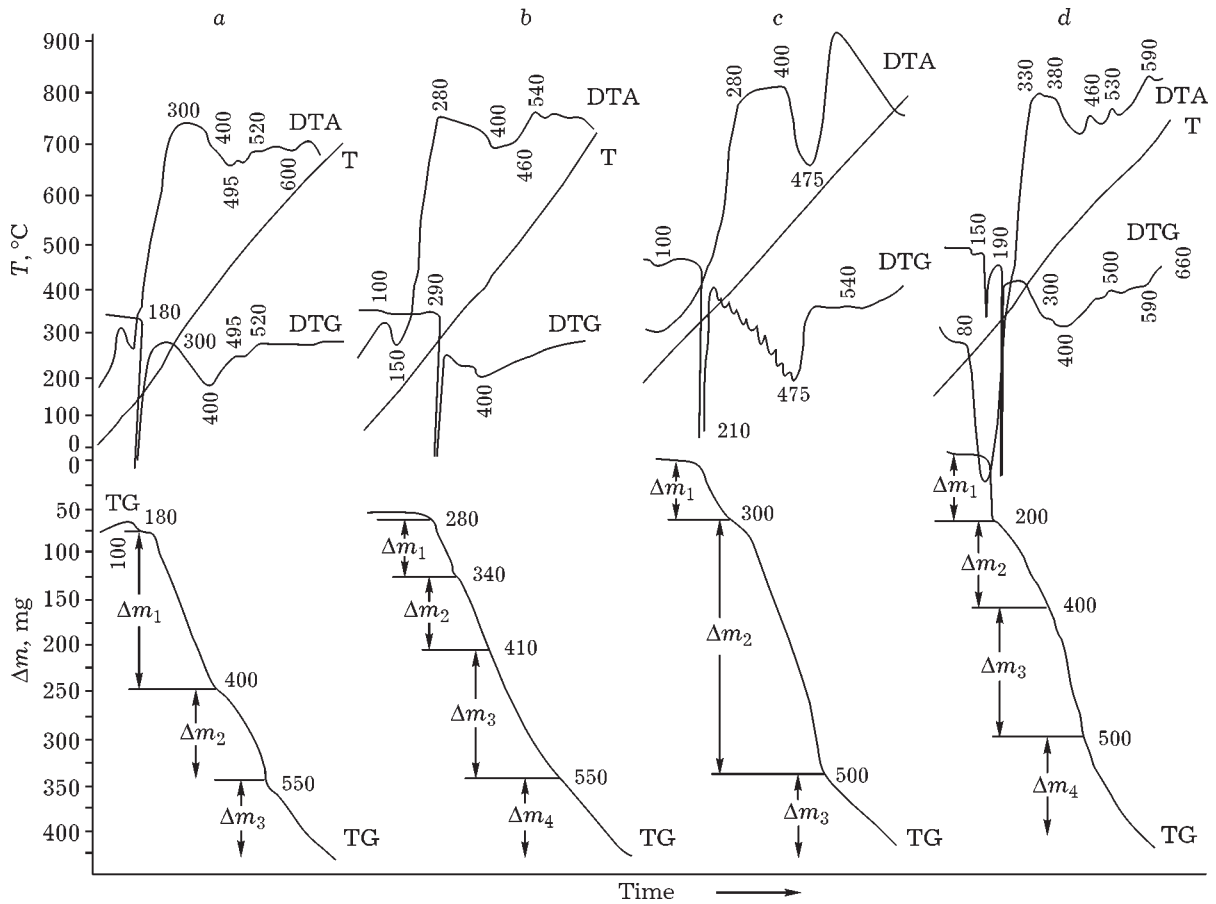
Fig 1. DTGA of the snap (initial) coal sample: $a-d$ − exp. Nos. 1−4, respectively (see Table 1).

within the range 180−210 °C, exp. No. 2 − 280 °C) in the amount of 5.5−7.0 % of the initial weighted portion. This process is not distinguished as a separate stage in experiments Nos. 1 and 4; it is united either with subsequent stage (exp. No. 1) or with the previous stage (exp. No. 4). Total mass loss within the studied temperature range varies within 36.1−42.5 % depending on final temperature of experiment (see Table 1).

The results of thermal analysis of the sample subjected preliminarily to mechanical activation (for 1 min) are presented in Fig. 2 and in Table 1. One can see that thermolysis is also characterized by the low-temperature explosion-type start at 150−200 °C with mass loss within 8−9 %. Then monotonous decrease of the mass is observed, to 38.5 % as a total for heating to 600 °C, and 42.0 % as a total for heating

TABLE 1

Thermal decomposition of brown coal from the Baganur deposit

| Stages | Initial sample | | | | | | | | | Mechanically activated sample | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Exp. No. | | | | | | | | | Exp. No. | | | |
| | 1 | | 2 | | 3 | | 4 | | | 1 | | 2 | |
| | $T$, °C | $\Delta m$, % | $T$, °C | $\Delta m$, % | $T$, °C | $\Delta m$, % | $T$, °C | $\Delta m$, % | | $T$, °C | $\Delta m$, % | $T$, °C | $\Delta m$, % |
| 1 | 80−100 | 0.2 | 100−150 | 0.5 | 100 | − | 80−150 | 5.5 | | 20−200 | 0.5 | 20−50 | 0.5 |
| 2 | 180−300 | | 280−300 | 6.5 | 210−300 | 7.0 | 150−200 | | | 200−280 | 9.5 | 180−200 | 5.1 |
| 3 | 300−400 | 17.1 | 300−400 | 10.5 | 300−400 | 27.5 | 200−400 | 11.2 | | 280−400 | 10.9 | 200−290 | 4.0 |
| | | | | | | | | | | | | 290−410 | 12.4 |
| 4 | 400−550 | 10 | 410−500 | 21.5 | 400−500 | | 400−500 | 12.4 | | 400−600 | 17.6 | 410−660 | 19.9 |
| 5 | 550−700 | 12 | 500−700 | | 500−700 | 8.0 | 500−650 | 7.0 | | | | | |
| Σ | 700 | ~39 | 700 | ~40 | 700 | ~42.5 | 650 | ~36.1 | | 600 | ~38.5 | 660 | ~42.0 |
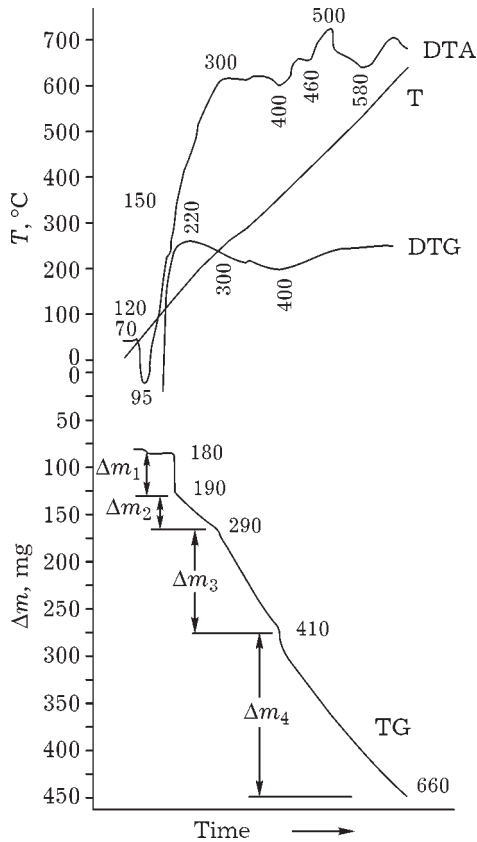
Fig. 2. DTGA of coal sample mechanically activated for 1 min.



Fig. 3. DTGA of larger-scale coal lot.

to 660 °C. So, short-term activation of the initial sample has no substantial effect on the dynamics of sample behaviour during thermolysis but promotes an increase in the sorption activity of the material. Sorption products are removed from the sample at the very beginning of heating, which is confirmed by the endo effect at DTA curve within the range 70–95 °C.

It should be noted that preliminary DTGA data point to the high concentration of volatile and readily boiling components in coal, the final product of thermolysis is not sintered but remains as loose material.

### DTGA of the samples of larger-scale lot

Results of the experiments with the coal samples of larger-scale lot are in good agreement with the results for the snap sample. Some insignificant deviations may be connected with different densities of the material in the crucible, which could affect diffusion of sublimates
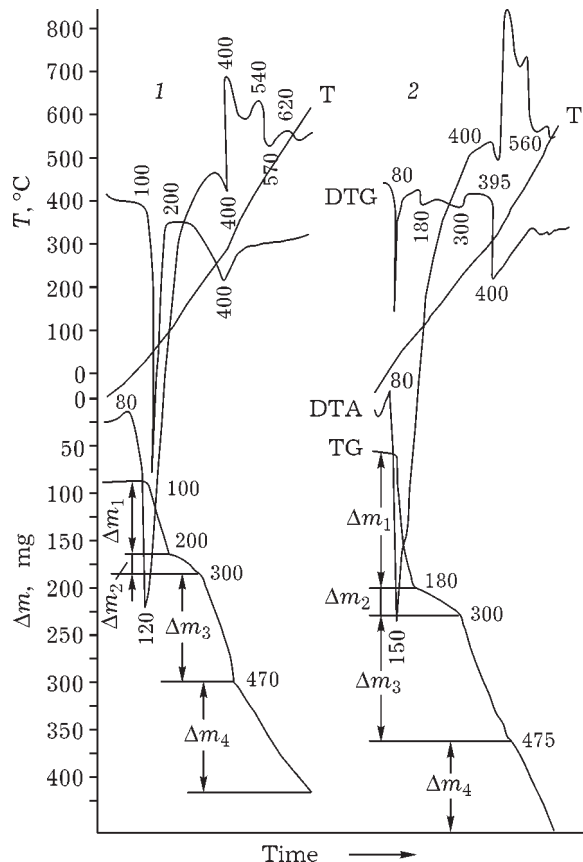
formed during heating. Thermolysis dynamics is shown in Fig. 3.

Thermal decomposition of the material is clearly exhibited in the DTA curve starting from the temperature of 80–100 °C as a strong endo effect with the peak maximum at ~120 °C and its termination at ~300 °C. Then a small but sharp exo effect with the peak maximum at ~400 °C is observed, followed by a sharp exothermal ascent passing into endo effect with the peak at ~500 °C. Subsequent small endo effects of thermal decomposition appear at a temperature of 540–570 and 620 °C.

The position of TG and DTG curves points to stage-by-stage change of sample mass during heating, at 80–200 and 300–470 °C. The first stage within temperature range 80–200 °C ($\Delta m_1$) corresponds to the mass loss of 9.4 %. Passing to the next stage, a small mass loss ($\Delta m_2$) equal to 2.7 % is observed. A decrease in sample mass at the stage within temperature range 300–470 °C ($\Delta m_3$) is 14.7 %. During further heating,

TABLE 2

Results of brown coal thermolysis with heating and exposure at 600−650 °C

| Exp. No. | Material | $T$, °C | Exposure, h | Mass loss, % | Note |
|---|---|---|---|---|---|
| 1 | Fines from packet | 460−525 | 3 | 8.6 | no cake |
| 2 | Medium from packet | 600 | 2 | 47.65 | «  « |
| 3 | Sample from monolith | 650 | 1 | 41.58 | «  « |
| 4 | Fines from packet | 650 | 1 | 38.02 | «  « |
| 5 | Average* | 600−650 | 1−2 | 42.42 | |

* The average of the sum of exp. Nos. 2−4.

the monotonous mass loss ($\Delta m_4$) reaches 14.1 %. So, total mass loss by the sample reaches almost 41 %, and in some experiments even 47 %.

### Determination of the aggregative and quantitative composition of the products of coal decomposition

Experiments on the determination of the composition of products formed in the decomposition of brown coal samples were carried out in different modes: 1) under uniform and continuous heating to the temperature of 600−650 °C, followed by exposure at the final temperature for 1−2 h; 2) under stage-by-stage heating to the maximal temperature (600−650 °C) with exposures for 2 h after each 100 °C.

The results of the experiments are presented in Tables 2, 3.

Under continuous heating of the samples to the temperature of 460−650 °C and exposure at this temperature (see Table 2), the mass loss by the material is 8.6−47.65 % and depends on the final temperature, exposure at the assigned temperature, as well as on the physical and aggregative state of the initial sample (particle size, the presence of the fine fraction, monolith pieces *etc*.). For instance, after heating to 460−525 °C and exposure at this temperature (see Table 2, exp. No. 1), mass loss was only 8.6 % of the initial mass, while heating and exposure at 600 °C are accompanied by mass loss up to 47.65 % (see Table 2, exp. No. 2). Higher mass loss is characteristic of the sample prepared by crushing a whole piece of coal (see Table 2, exp. Nos. 3, 4) after exposure at a temperature of 650 °C (41.58 %) in comparison with the sample prepared from fine middlings taken directly from the packet (38.02 %). This difference is likely due to the partial loss of gaseous components of this kind of coal due to diffusion weathering during storage under the conditions of free contact with the environment.

Therefore, mass loss by coal under thermolysis occurs due to evaporation of both the gas component and the sublimates of readily boiling fractions present in this kind of coal. Visually, the fraction of the condensate of sublimates may be insignificant in the total mass

TABLE 3

Results of thermolysis of brown coal with stage-by-stage temperature exposure (duration of each stage: 2 h)

| $T$, °C | Exp. No. 1 (sample from monolith) | | | Exp. No. 2 (averaged sample from packet) | | |
|---|---|---|---|---|---|---|
| | $\Delta m$, % | Condensate, % | Gaseous fraction, % | $\Delta m$, % | Condensate, % | Gaseous fraction, % |
| 200 | 18.73 | 0.28 | 18.44 | 9.5 | 6 | 3.5 |
| 300 | 9.3 | 1.5 | 7.54 | 10.04 | 4.04 | 6 |
| 400 | 10.2 | 3.65 | 6.6 | 9.74 | 4.14 | 5.6 |
| 500 | 6.58 | 0.64 | 5.94 | 7.7 | 2 | 5.7 |
| 600 | 5.03 | 0.37 | 4.66 | − | − | − |
| 650 | − | − | − | 5.05 | 1.9 | 3.15 |
| Σ | 49.94 | 6.39 | 43.55 | 42.03 | 18.08 | 23.95 |

*Note*. Dash means determination was not carried out.

loss by coal. The condensate is dark coloured with a deep crimson (close to black) hue. A small amount of dark tarry fraction gets layered on its surface during storage.

The results of experiments with stage-by-stage heating and exposure of samples somewhat differ but generally correlate with the data of previous experiments (see Table 3). For example, in the case when the initial sample was freshly prepared lump monolith (exp. No. 1) substantial mass loss (18.73 %) was observed even at 200 °C, mainly in the form of gaseous component. At the same time, in the experiment with averaged sample composed of a mixture of initial fine fragments and small-sized lump material crushed in a mortar (exp. No. 2), exposed at 200 °C, mass loss was equal to 9.5 %. This mass loss is mainly connected with the evaporation of condensable sublimates, and only to a small extent with the removal of gaseous component. Exposures at subsequent temperature stages in both experiments are accompanied by comparable mass losses differing from each other only in the value of the loss of gaseous components. As a consequence, the final results of experiments with stage-by-stage exposures exhibit differences only due to mass losses at the stage of exposure at 200 °C. The total mass loss in exp. No. 1 after the final (600 °C) stage with exposure was 49.94 % and was determined mainly by the removal of the gas components (43.55 %). In exp. No. 2, the total mass loss is smaller by almost 8 % (42.03 %), and it is almost equally determined by the value of condensable sublimates (18.08 %) and the formation of gaseous components (23.95 %); the amount of the latter is almost two times smaller in comparison with similar data for exp. No. 1.

## CONCLUSION

Investigation of thermolysis of brown coal from the Baganur deposit showed that this kind of coal is characterized by the high (>40 %) content of gaseous components that are removed from the material by heating to temperature about 200 °C, insignificant concentration of fluid components (up to 7 %) and the absence of binding components. As a result, the material is not sintered within the whole temperature range of thermolysis up to 650 °C. The matrix solid-phase product of thermolysis is loose graphite-like material which may be briquetted only after the addition of binding compositions.

## REFERENCES

1 Gornaya Entsiklopediya, Sov. Entsiklopediya, Moscow, 1991, vol. 5, pp. 227.
2 Sodnam N. I., Shchipko M. L., Kuznetsov B. N., *Khim. Tv. Topliva*, 6 (1987) 12.
3 Sodnam N. I., Bondarenko O. D., Shklyaev A. A., *Khim. Tv. Topliva*, 1 (1989) 22.
4 Shibanov V. I., Yakovlev I. Yu., *Khim. Tv. Topliva*, 6 (1989) 52.
5 Kaminskiy Yu. D., Kopylov N. I., Kulikova M. P., Nauchnye Trudy TuvIKOPR SO RAN (Treatises), Kyzyl, 2005, issue 8, pp. 123−128.
6 Kopylov N. I., Kaminskiy Yu. D., Kulikova M. P., *Khim. Tekhnol.*, 9, 4 (2008) 168.
7 Kopylov N. I., Kaminskiy Yu. D., *Chem. Sustain. Dev.*, 21, 3 (2013) 319.
   URL: http://www.sibran.ru/en/journals/KhUR
8 Kaminskiy Yu. D., Polurudov A. V., Kulikova M. P., Kopylov N. I., Kara-sal B. K., Kotelnikov V. I., Soyan M. K., Burdin N. V., Moldurushku R. O., Kaminskaya I. A., Svodny Otchet po Konkursnomu Proektu SO RAN No. 28.4.8 (2004−2006), TuvIKOPR SO RAN, Kyzyl, 2006.