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# Study on thermal decomposition and enrichment quality of coal from Mogoin gol deposit in Mongolia

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***Abstract.*** *The main purpose of this study is to investigate the thermal stability and the mechanism of thermal decomposition of Mogoin gol coal, the possibility of liquefaction by pyrolysis and thermolysis, and the possibility of enriching by heavy liquids to reduce the mineral content of coal and improve its quality. Under this purpose, the Mogoin gol coal was characterized by proximate and ultimate analysis, thermogravimetry, and investigated its thermal decomposition (thermolysis and pyrolysis). Thermogravimetric analysis was performed using Japanese HITACHI TG/DTA7300 instrument and pyrolysis investigation was carried out at different heating temperatures 200–700 °C with constant heating rate 20 °C/min for 80 min. On the basis of proximate and elemental analysis results, it has been indicated that the Mogoin gol coal is high-rank coking coal. The pyrolysis of Mogoin gol coal was studied by SNOL furnace at different heating temperatures and obtained from pyrolysis products such as hard residue, tar, pyrolytic water, and gas. From pyrolysis, the yield of pyrolysis tar (6.28 %) was highest at 700 °C. The experiment of thermal decomposition (thermolysis) was carried out in air closed autoclave at 350–450 °C and using hydrogen donor solvent (tetraline) with different mass ratios of coal and solvent (1:1.75; 1:1.5). In the thermolysis experiment, the yield of liquid product is highest with the coal-solvent ratio of 1: 1.5 at 450 °C.*

***Keywords:*** *thermolysis, pyrolysis, enrichment, liquid product, thermogravimetry*

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**ХИМИЧЕСКАЯ ТЕХНОЛОГИЯ**

**Научная статья**
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# Исследование термического разложения и качества обогащения угля месторождения Могоин-гол в Монголии

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***Аннотация.*** *Основной целью настоящего исследования является изучение термической стабильности и механизма термического разложения угля Могоин-гол, возможности ожижения пиролизом и термолизом, а также возможности обогащения тяжелыми жидкостями для снижения минеральности угля и улучшения его свойств. С этой целью могоин-гольский уголь охарактеризован при помощи экспрессивного, предельного анализа и термогравиметрии, исследовано его термическое разложение (термолиз и пиролиз). Термогравиметрический анализ проводили на японском приборе HITACHI TG/DTA7300, изучение пиролиза осуществляли при различных температурах нагрева (200–700 °C) с постоянной скоростью нагрева 20 °C/мин в течение 80 мин. На основании результатов экспресс- и элементного анализа установлено, что могоин-гольский уголь относится к высокосортным коксующимся углям. Пиролиз могоин-гольского угля изучался в печи СНОЛ при различ-*

*ных температурах нагрева и получен из продуктов пиролиза, таких как твердый остаток, гудрон, пиролитическая вода и газ. При пиролизе выход пиролизной смолы (6.28%) был самым высоким при температуре 700 °C. Опыт термического разложения (термолиз) проводили в автоклаве закрытого типа при температуре 350–450 °C с использованием растворителя-донора водорода (тетралина) при различном массовом соотношении угля и растворителя (1:1,75; 1:1,5). В эксперименте по термолизу выход жидкого продукта максимален при соотношении уголь–растворитель 1:1,5 при температуре 450 °C.*

***Ключевые слова:*** *термолиз, пиролиз, обогащение, жидкий продукт, термогравиметрия*

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## INTRODUCTION

Mongolia is among the 10 coal rich countries in the world with the 175 billion ton of geologically estimated coal resources including high quality bituminous coking coals, subbituminous coals and brown coals. More than 1.8 billion ton of this coal resource belongs to the high coking coal. It is important that, to study the mechanism of thermal decomposition of coal, necessary to reduce the amount of minerals in coal [1]. Coal and coal products will play an increasingly important role in fulfilling the energy needs of our society [2–3]. Based on IEA 2008 report, coal accounts for about 26% of the total world primary energy consumption in 2006 [4, 5]. Thermal analysis has been widely used in the recent years for the investigation of combustion and pyrolysis behavior of fossil fuels such as coal, oil shales and tar sands [6–8]. Thermal properties of coal play a significant role industry and impact on coal combustion on environmental pollution. Converting coal into oil and gas allows coal to be utilized as an alternative fuel, which will affect the national security and the economically sustainable development [6–12].

Thermogravimetry is usually used as a means of determining pyrolytic characteristics and kinetic parameters. Thermogravimetry measures the weight change of a tested material as a function of temperature or time in a controlled atmosphere, such as nitrogen or argon atmosphere [13–15]. Measurements are used primarily to determine the composition of materials and to predict their thermal stability at temperatures up to 1000 °C. On the other hand pyrolysis is an efficient form (method) of treatment of organic material at elevated temperatures in the absence of oxygen [16]. It involves the simultaneous and irreversible change of chemical composition and physical phase during thermochemical decomposition of organic material by heat. As a result of pyrolysis, solid (hard residue), condensed liquid (tar and pyrolysis water) and gas (uncondensed) products can be obtained. Solid product is porous material with higher caloric value which can be used as coke, semi-coke, smokeless fuel, adsorbent material and so on. Tar is petroleum like product and can be used as complex raw material for production of chemical substances, gasoline, diesel, oils, bituminous and so on. The gas product can be used as gas fuel after cleaning of nitrogen and sulfur containing pollutants in it [17].

Enrichment plays an important role in increasing the efficiency and effectiveness of coal mining and exploitation. Coal minerals have included some harmful elements in the environment. It is a negative impact on all processes of its use. Reducing the mineral content of coal can significantly reduce its transportation costs, while raw coal used for coking and direct liquefaction of coal is required to be low in minerals [14, 15]. As of 2006, 1/3 of the world's total mined coal was enriched, and there are about 2,500 enrichment plants, of which 2,000 (with a total capacity of 800 million tons/year) are in China and 265 (with a total capacity of 986 million tons/year) in the United States, 53 (total capacity 134 million tons/year) in India, 87 (total capacity 95 million tons/year) in Russia, and 70 million tons/year in Australia [16]. It is possible to reduce the amount of sulfur oxides and volatile toxic elements released from coal combustion by the coal enrichment method [17, 18].

The main purpose of this study is to investigate the thermal stability and the mechanism of thermal decomposition of Mogoin gol coal, the possibility of liquefaction by pyrolysis and thermolysis, and the possibility of enriching by heavy liquids to reduce the mineral content of coal and improve its quality.

## EXPERIMENTAL

The Mogoin gol coal is located in the province Khuvsgul 930 km from Ulaanbaatar, 210 km west of Murun soum of Khuvsgul aimag, and 22 km southeast of Tsetserleg soum, Khuvsgul aimag. The total coal resources are 11 million tons.

The analytical samples of Mogoin gol coal were prepared according to ASTM D 2797. The proximate and ultimate analysis were performed according to MNS 656-79 (moisture content), MNS 652-79 (ash yield), MNS 654-79 (volatile matter yield), MNS 669-87 (gross calorific value) and MNS 895-79 (sulphur content). The content of mineral elements in coal ash and their oxides have been determined by using of X-ray spectrometry.

Thermogravimetric analysis was performed using Japanese HITACHI TG/DTA7300 instrument. The instrument was used to study the heat resistance of analytical coal samples and was tested in an argon gas environment at 25–1150 °C for 120 minutes at a heating rate of 10 °C/min.

Enrichment properties of Mogoin gol coal in laboratory conditions were determined by heavy liquid GOST 4790-93 method. During experimental, coal fraction of 3–1.5 mm was separated in heavy liquid that $ZnCl_2$ solution with different densities such as <1.3; 1.3–1.4; 1.4–1.5; 1.5–1.6; 1.6–1.7 and > 1.7 g/cm$^3$. After that, washing and removing the submerged and floating fractions from the solution residue, determined their yield and ash content. Enrichment curves were constructed on the basis of enrichment results.

The pyrolysis experiments were performed in a laboratory quarts retort (tube) which could contain 1 g analytical coal sample. The retort was placed in a horizontal electric tube furnace with a maximum heating temperature of 950 °C. The pyrolysis experiments have been carried out at different heating temperatures 200–700 °C with constant heating rate 20 °C/min for 80 min.

The experiment of thermal decomposition (thermolysis) was carried out in air closed autoclave at 350–450 °C and using hydrogen donor solvent (tetraline) with different mass ratios of coal-solvent (1:1.75; 1:1.5) for 2 h. After cooling, all uncondensed gas and liquid products were removed from hard residue using chloroform solvent in a Soxhlet apparatus. Then the yield of thermolysis products was calculated by the weight method.

## RESULTS AND DISCUSSIONS

The proximate and ultimate analysis of the Mogoin gol coal was determined and the results are shown in Tab. 1.

From Tab. 1, the volatile matter of the Mogoin gol coal is $V^{daf}$ = 30.7% and the carbon content is $C^{daf}$ = 76.5% and these results show that, it is a coking coal (high rank coal). The infrared spectrum is mainly used to study different kind of functional groups in coal organic mass. Fig. 1 below shows the FTIR spectrum.

It can be seen from Fig. 1 that the Mogoin gol coal has several functional structures. The absorption intensity of hydroxyl at 3500 cm$^{-1}$ and absorption intensity at 1373 to 1604 cm$^{-1}$ totally indicate carboxylic groups. C–O bonds of alcohols, ethers, and esters take to adsorption at 1273 cm$^{-1}$. C–H bond of aromatic ring indicate band at 500–1033 cm$^{-1}$ and aliphatic –CH; –$CH_2$ and –$CH_3$ groups with

**Table 1.** Proximate and ultimate analysis of the Mogoin gol coal

**Таблица 1.** Экспресс- и окончательный анализ угля Могоин-гольского месторождения

| $W^a$, % | $A^d$, % | $V^{daf}$, % | Q, kcal/kg | $C^{daf}$, % | $H^{daf}$, % | $S^{daf}$, % | $(O+N)^{daf}$, % | H/C |
|---|---|---|---|---|---|---|---|---|
| 0.62 | 10.59 | 30.67 | 7531.73 | 76.5 | 5.2 | 1.27 | 17.03 | 0.8 |

**Table 2.** The chemical composition of Mogoin gol coal ash, %

**Таблица 2.** Химический состав золы угля месторождения Могоин-гол, %

| | Mg | Al | Si | P | K | Ca | Ti | Mn | Fe | Zn |
|---|---|---|---|---|---|---|---|---|---|---|
| Elements | 2.2 | 11.2 | 2.6 | 0.2 | 0.4 | 4.7 | 0.2 | 0.3 | 34.2 | 0.1 |
| Oxides | 3.5 | 21.1 | 5.6 | 0.3 | 0.5 | 6.6 | 0.4 | 0.5 | 49.0 | 0.2 |

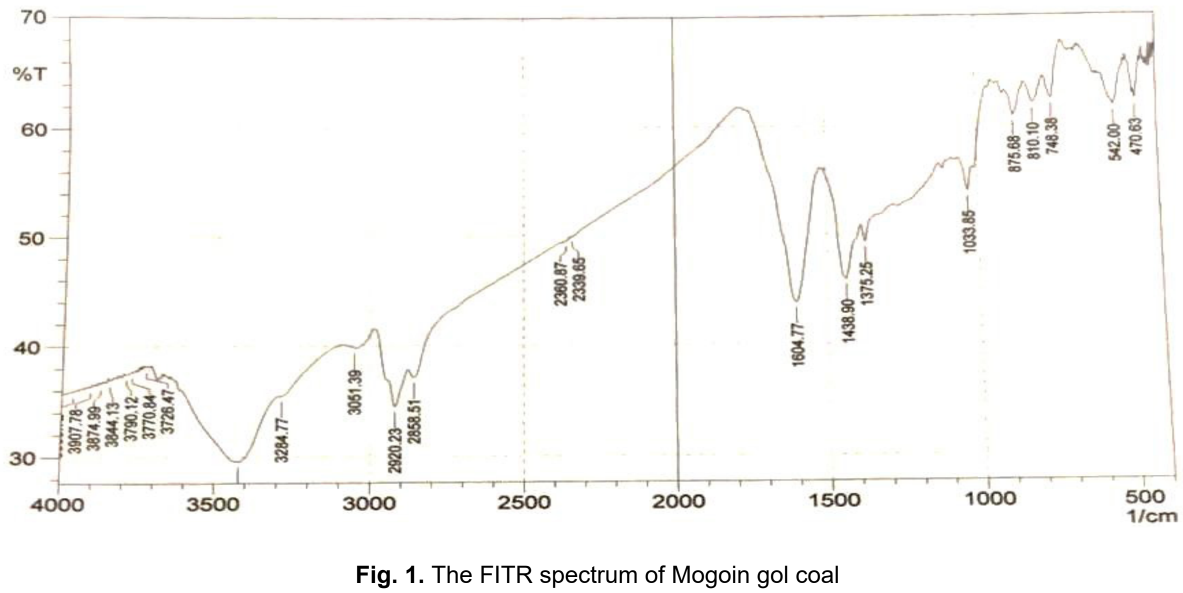


**Fig. 1.** The FITR spectrum of Mogoin gol coal

**Рис. 1.** FITR-спектр угля месторождения Могоин-гол

**Table 3.** The result of enrichment of Mogoin gol coal by heavy liquid method

**Таблица 3.** Результат обогащения Могоин-гольского угля тяжеложидкостным способом

| Density, g/cm³ | Yield of fraction, γ% | Ash of fraction, Aª,% | Total light fraction | | Total heavy fraction | |
|---|---|---|---|---|---|---|
| | | | Yield, γ% | Ash, Aª,% | Yield, γ% | Ash, Aª,% |
| 1.3 | 20.7 | 4.6 | 20.7 | 4.6 | 98.9 | 9.0 |
| 1.4 | 35.0 | 6.0 | 55.7 | 5.4 | 78.2 | 10.2 |
| 1.5 | 39.6 | 11.8 | 95.3 | 8.1 | 43.2 | 13.7 |
| 1.6 | 2.0 | 25.8 | 97.3 | 8.4 | 3.6 | 34.5 |
| 1.7 light | 0.5 | 45.8 | 97.8 | 8.6 | 1.6 | 45.8 |
| 1.7 heavy | 1.1 | 45.8 | 98.9 | 9.0 | 1.1 | 45.8 |

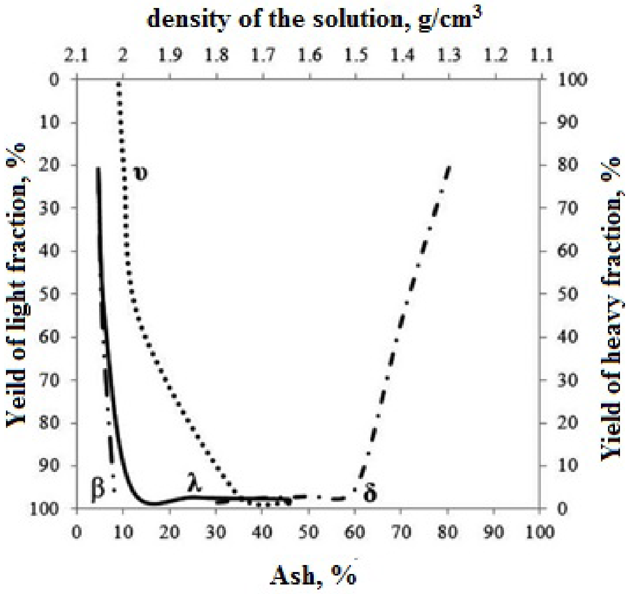


**Fig. 2.** Coal heavy liquid enrichment curve of 1.5–3 mm fraction:
β – light fraction curve; λ – elementary ash curve;
δ – density curve; υ – heavy fraction curve

**Рис. 2.** Кривая обогащения угля фракцией 1,5–3 мм:
β – кривая легкой фракции; λ – элементарная кривая зольности; δ – кривая плотности;
υ – кривая тяжелой фракции

middle intensity at 1376–1456 cm$^{-1}$. The chemical composition of coal ash was studied by X-ray fluorescence spectrum and the results are shown in Tab. 2.

Tab. 2 shows that, the Mogoin gol coal ash has the highest content of iron oxide 49.0% and aluminum oxide 21.1%. The melting temperature of ash varies depending on the chemical composition of the coal ash. For example, as the $SiO_2$ content increases, ash melting temperature is increased.

The fraction of Mogoin gol coal with size 3–1.5 mm was enriched by using of zinc chloride ($ZnCl_2$) as a heavy liquid with a density of 1.3–1.7 g/cm³. The yields of fractions and ash content of faction were determined, and the enrichment curve is constructed and shown as a result of enrichment in Tab. 3, 4 and Fig. 2.

The results in Tab. 3 show that enrichment of Mogoin gol coal in zinc chloride solution with densities 1.3–1.4 g/cm³ show lower ash contents 4.6–6.0%, which is an optimal condition for the enrichment of the Mogoin gol coal.

According to Tab. 4 and Fig. 2, the concentrate's ash in a heavy liquid with a density of δ = 1.3 g/cm³ was low Aa = 4.6% and the yield was γ = 20.7%. As the density of the heavy liquid increases, the ash and yield of the concentrate tend to gradually increase. The tailing ash is Aa = 45.8%, the yield is low γ = 1.1%. The calculation of the yield and ash of the concentrated product using the enrichment quality curve (Tab. 2) shows that the yield of the Middling is γ = 2.5% and the ash is A = 29.4%. Therefore, if the average fractional yield of 1.5–3 mm coal is evaluated, T = 2.5%, which is the property good quality of enrichment. Therefore, sulfur content and calorific value were determined in concentrate and initial coal and shown Tab. 5.

The Tab. 5 above shows that the sulfur content of the concentrate decreased by 18% and the calorific value increased by 8% compared to the initial coal. In order to study the thermal stability and thermal decomposition mechanism of the organic mass of the Mogoin gol coal, the thermogravimetric analysis was performed in an argon gas environment and the TG and DTG curves were shown Fig. 3. The thermal stability indices T5%, T15%, T25% are calculated from the TG curve and are shown in Tab. 6.

From termogravimetric study, Mogoin gol coal ended 72.5% weight loss at 980 °C.

The TG curve of Mogoin gol consists of different temperature intervals (steps) such as 100–360; 360–470; 470–590; 590–980 °C. In the first step (until 360 °C) the weight loss is due to the release of some absorbed gas and moisture in coal.

In the second step at 360–470 °C, (TG – 85.3%, DTG – 84.8*10$^{-3}$ mg/min) intensive thermal decomposition of the organic matter of the coal samples start forming liquid (tar and pyrolysis water) and gas products. In the third step at 550–850 °C, the weight loss strongly decreases, which is an indication for ending the thermal decomposition and starting carbonization and polymerization of the coal sample (TG – 79%, DTG – 24.4*10$^{-3}$ mg/min). In the fourth step at 850–980 °C, the weight loss slowly

**Table 4.** Coal enrichment products yield, and its ash content

**Таблица 4.** Выход продуктов обогащения угля и его зольность

| Products of heavy liquid | Yield, γ, % | Ash, $A^a$, % |
|---|---|---|
| Concentrate | 95.3 | 8.1 |
| Middling | 2.5 | 29.4 |
| Tailing | 1.1 | 45.8 |

**Table 5.** The result of sulfur content and caloric value of concentrate and initial coal

**Таблица 5.** Сернистость и калорийность концентрата и исходного угля

| Samples | Sulfur, $S_t^d$, % | Caloric value, $Q^{daf}$, kcal/kg |
|---|---|---|
| Concentrate | 1.09 | 7971.15 |
| Initial coal | 1.27 | 7531.73 |

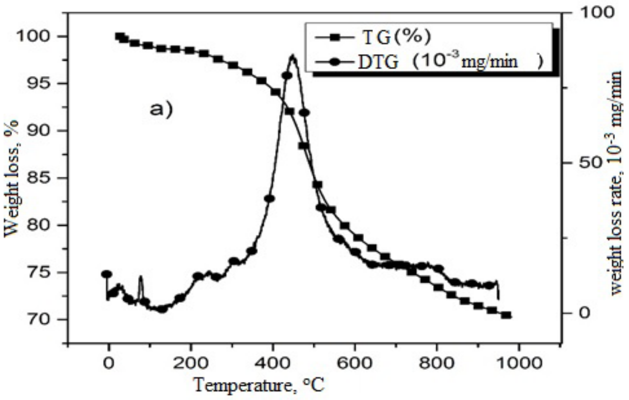


**Fig. 3.** The TG and DTG curves of Mogoin gol coal

**Рис. 3.** Термогравиметрические и дифференциальные термогравиметрические кривые угля месторождения Могоин-гол

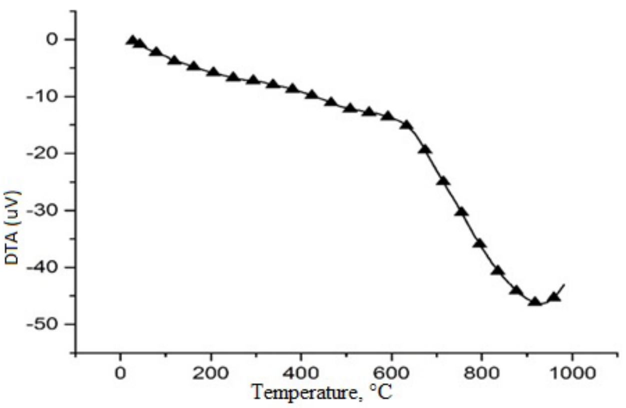


**Fig. 4.** DTA curve of Mogoin gol coal

**Рис. 4.** Кривая дифференциального термического анализа угля месторождения Могоин-гол

increases, which is related with the release of gas, e.g. $CO_2$, $H_2$, CO from the mineral matter of coal sample (TG – 70.3%, DTG – $10.2*10^{-3}$ mg/min).

The calculated value of thermal stability indices: T5% = 382.2 °C, T15% = 501.5 °C, T25% = 740.0 °C from the TG curve (Tab. 6) show that Mogoin gol coal characterizes with much higher thermal stability than for example brown coals of lignite type.

To explain the mechanism of thermal decomposition of coal, let us consider the DTA curve that takes place during the thermal decomposition of coal obtained by thermogravimetric analysis (Fig. 4).

Shown at the DTA curve of the Mogoin gol coal, thermal decomposition is an endothermic process that Heat absorption took place at 400–980 and at 900 °C. The thermal stability of different types of solid fossil fuels, such as lignite and hard coal, varies, and the yield of semi-coking and pyrolysis products, such as thermal decomposition, is used to determine whether the coal is suitable for pyrolysis, liquefaction and gasification [19–20]. The yield and properties of the resulting product vary depending on the pyrolysis temperature and heating rate. In this part of the study, the results of pyrolysis experiment of Mogoin gol coal (200–700 °C) are shown in the following Tab. 6.

Experimental results show that when the heating temperature is low at 200–300 °C, the yield of liquid products, pyrolysis water, and gaseous products is low and when an increase in heating temperature, decreased in the solid residue yield and increase the yield of liquid and gaseous product. Maximum yield of liquid product is 6.28% at 700 °C. The results of the technical analysis of hard residue have been shown in Tab. 7.

According to the table above, the yield of volatile matter in pyrolysis hard residue is 16 times lower than the initial coal, and pyrolysis hard residue's yield is (71%) high and ash content down, so the coal is more suitable to produce high-quality smokeless fuel.

Hydrogen donor solvent tetraline was used in the thermolysis experiment. Coal and solvent (coal:solvent = 1: 1.5; 1: 1.75) were taken by mass ratio and tested in a standard laboratory autoclave made of stainless steel at 350, 400, 450 °C for 2 h and the results are shown in the Tab. 8.

According to the experiment, the conditions for the highest yield of liquid products were the carbon-solvent ratio of 1:1.5 at 450 °C. According to the thermolysis process, the yield of liquid products increases as the temperature increases, and when coal decomposition increases, the yield of solid residues decreased. This is due to the fact that, under the influence of high temperatures, the organic mass of coal is decomposed as much as possible and secondly, temperature is a key factor in generating free radicals, so free radicals from the decomposition of coal macromolecules react with donor solvents to increase liquid yield. Fig. 5 and 6 also shows the FTIR spectrum of the pyrolysis and thermolysis process liquid products.

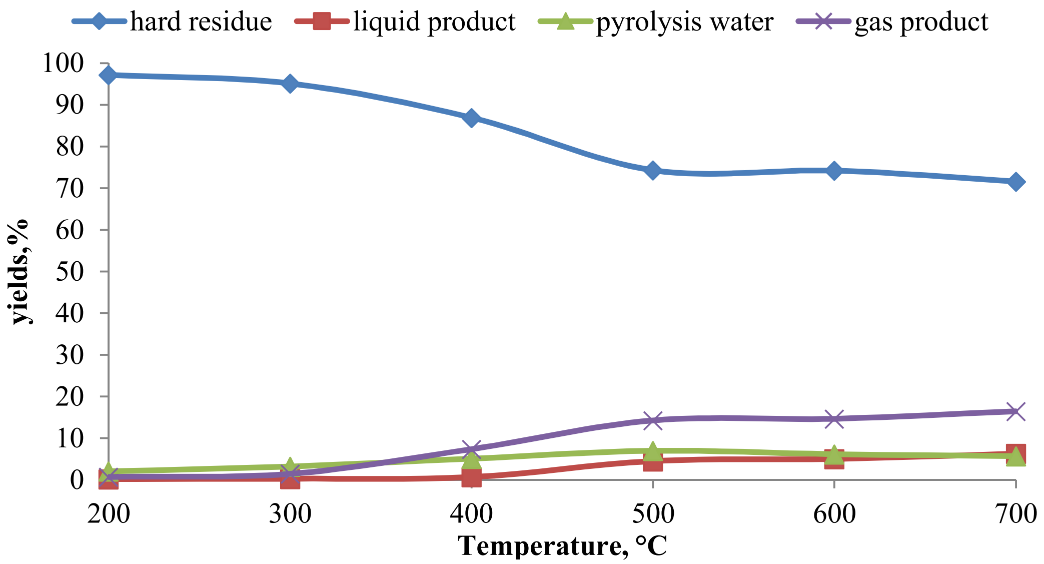


**Fig. 5.** The results of pyrolysis of Mogoin gol coal
**Рис. 5.** Результаты пиролиза угля месторождения Могоин-гол

**Table 6.** The thermal stability index of the Mogoin gol coal

**Таблица 6.** Показатель термической стабильности угля месторождения Могоин-гол

| Heating Rate, °C/min | Thermal stability indices, °C | | |
|---|---|---|---|
| | $T_{5\%}$ | $T_{15\%}$ | $T_{25\%}$ |
| 10 °C/min | 382.2 | 501.5 | 740.0 |

It can be seen from Fig. 6 A and B that the liquid products have similar functional structures but have different band absorption intensities. The absorption intensity of hydroxyl at 3404 $cm^{-1}$ significantly decreased in thermolysis liquid product. Ideally, hydrogen-bonded hydroxyl groups tend to be destroyed or broken during thermal treating processes [13]. The absorption intensity at 1707 $cm^{-1}$ totally came out in thermolysis and pyrolysis liquid products, indicating carboxylic groups. According to the literature, C–O bonds of alcohols, ethers, and esters start to decompose over 200 °C, thus the bands at 1273 $cm^{-1}$ are almost completely condensated after pyrolysis and thermolysis. The intensity of bands related to aliphatic hydrocarbon, including the vibration of aliphatic methylene and methyl group content significantly. This intensity coincides with the fracture of aliphatic side chains to form methane or other light hydrocarbon gases. Therefore pyrolysis liquid product of Mogoin gol coal consists of mainly aliphatic, aromatic and aromatic-aliphatic compounds with above mentioned functional groups in their molecules.

**Table 7.** The results of the technical analysis of pyrolysis hard residue of Mogoin gol coal

**Таблица 7.** Результаты технического анализа твердого остатка пиролиза угля месторождения Могоин-гол

| $W^a$, % | $A^a$, % | $V^a$, % | $V^{daf}$, % | $Q^{daf}$, % |
|---|---|---|---|---|
| 0.64 | 11.01 | 1.63 | 1.84 | 7882.20 |

**Table 8.** The thermolysis result of Mogoin gol coal

**Таблица 8.** Результат термолиза угля месторождения Могоин-гол

| Ratio | Temperature, °C | Hard residue | Liquid product, % | Gaseous product, % |
|---|---|---|---|---|
| 1:1.5 | 350 | 87.31 | 12.69 | - |
| | 400 | 34.53 | 59.39 | 3.92 |
| | 450 | 32.21 | 63.87 | 6.08 |
| 1:1.75 | 350 | 90.61 | 9.39 | - |
| | 400 | 47.59 | 48.84 | 3.57 |
| | 450 | 40.73 | 53.91 | 5.36 |

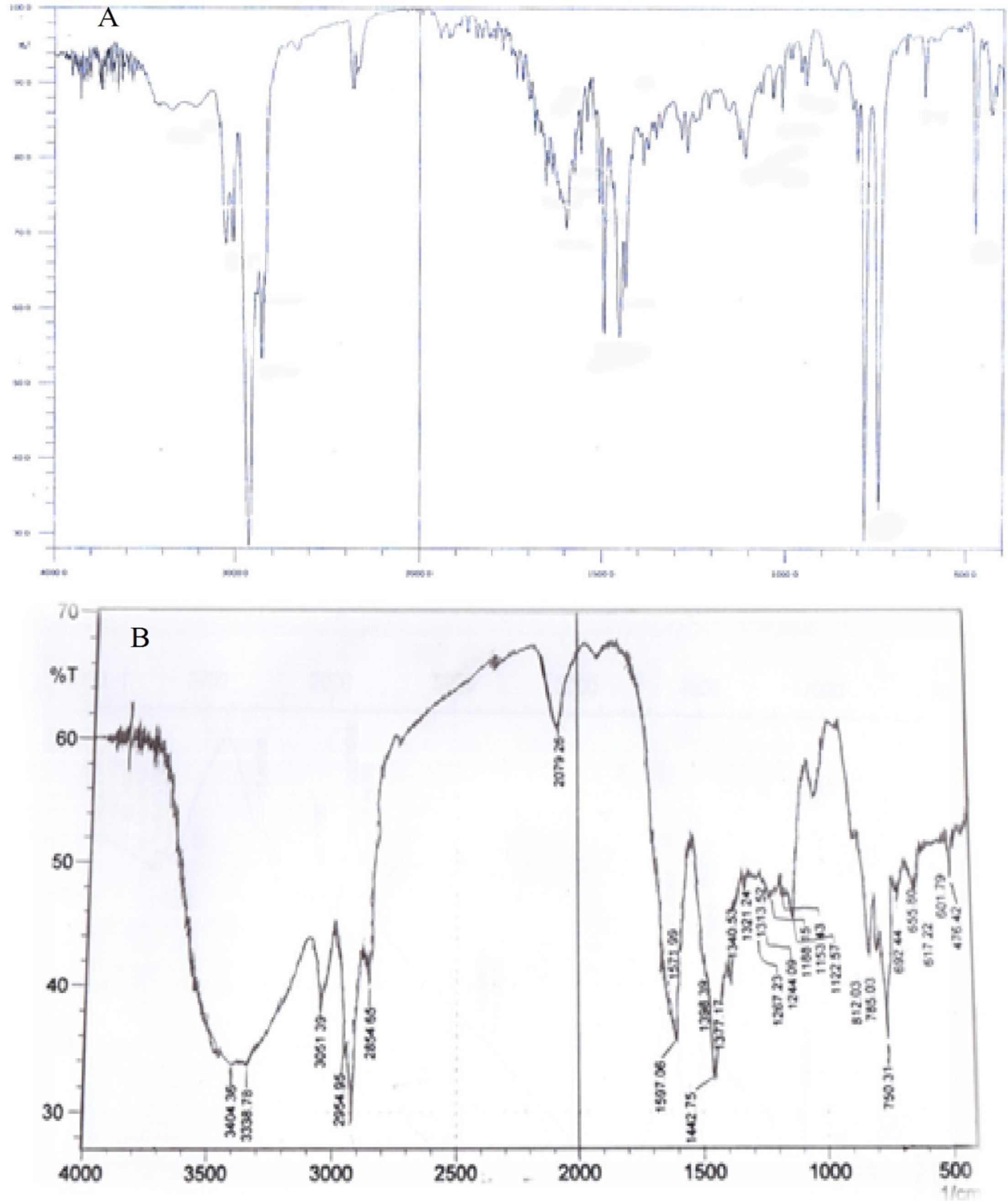


**Fig. 6.** FTIR spectrum: A – thermolysis liquid product; B – pyrolysis liquid product

**Рис. 6.** ИК-Фурье спектр: A – жидкий продукт термолиза; B – жидкий продукт пиролиза

## CONCLUSION

On the basis of proximate and ultimate analysis results, it has been confirmed that the coal of Mogoin gol deposit is a high rank coking coal. The ash content of concentrate of Mogoin gol coal is 1.5 times less than initial coal and is the result of good quality of enrichment.

Selecting the heating temperature regions and explaining the process taking place in each region, the thermal effects were recorded as endothermic at 25–100 °C, exothermic at 360–590 °C, and endothermic at 595–980 °C. The temperature value occurs is T = 470–590 °C. The calculated value of thermal stability indices: $T_{5\%}$ = 382.2 °C, $T_{15\%}$ = 501.5 °C, $T_{25\%}$ = 740.0 °C from the TG curve show that the Mogoin gol characterizes higher thermal stability than for example brown coals.

The pyrolysis process has a yield of 71% hard residue at 700 °C, low ash content and volatile matter yield, and high calorific value, which makes it possible to produce high quality smokeless fuel from Mogoin gol coal. As a result of the thermolysis experiment, coal (coal:solvent = 1:1.5) processed at 450 °C in a donor solvent tetraline with a high yield of 63.87%, the liquid product includes benzene and its derivatives and aliphatic-alkylated aromatic hydrocarbon compounds.

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