

Artificial graphite from Shubarkol coal obtained by sublimation of carbon atoms into the gas phase followed by desublimation into high-purity graphite

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ABSTRACT

This article discusses a plasma-chemical method for producing high-purity graphite from an air suspension of low-ash coal particles from the Shubarkol deposit in Kazakhstan. The technological process is based on the ability of carbon to transform from a solid to a gaseous state, bypassing the liquid state. This means it sublimates at high temperatures and desublimates as the temperature of the gaseous medium in the reactor zone decreases. The use of a graphite catalyst allows for controlled formation of the graphitized material. Atomic carbon graphitization occurs over a wide temperature range. It was established that graphite obtained in high-temperature reactor zones is purer than graphite obtained in reactor zones close to 500°C. This feature of the graphitization process enables product classification by quality. The design of a reactor based on sublimation and desublimation processes for graphite production is discussed. The use of a high-frequency electromagnetic zone in the plasma-chemical reactor design allows for controlled graphitization of atomic carbon, intensifying desublimation processes over a graphite powder catalyst. The plasma-chemical apparatus design includes a dust collection system and carbon monoxide neutralization, which can occur due to variations in the component proportions in the feedstock, which includes carbon powder, graphite powder catalyst, and carbon dioxide. The developed apparatus can be used to produce a sorbent – thermochemically expanded graphite – from graphite by varying the operating mode. The aim of this research is to develop a plasma-chemical technology for producing graphite from coal based on sublimation and desublimation processes in a single reactor, with the separation of impurities during the graphitization of carbon atoms over a graphite catalyst.

Keywords: plasma, sublimation, desublimation, graphitization, carbon, graphite, separation, catalyst.

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Introduction

In the next decade, graphite will be the most sought-after material for nuclear power, mechanical engineering, chemical engineering, and metallurgy. Graphite is used to manufacture many specialized products, such as battery electrodes, protective screens, sorbents, and much more. But the most important aspect of graphite today is the growing demand for high-purity, synthetic graphite. Obtaining high-purity graphite from readily available raw materials, which are abundant in Kazakhstan, such as the Shubarkol coal deposit, is a pressing issue. Developing technologies for processing coal into high-purity graphite is a promising solution. A promising solution is to utilize carbon's ability to sublime in a non-oxidizing atmosphere and desublime upon cooling, creating conditions conducive to graphitization.

An analysis of modern achievements in the science of obtaining artificial graphite [[1], [2], [3], [4], [5], [6], [7], [8], [9], [10]] allows to use some of the subtleties of artificial graphite production in technology to improve the quality of the main product.

Graphite is a mineral composed of carbon atoms arranged in layers. High bond strength between atoms within layers and low bond strength between atoms between layers account for graphite's low mechanical strength. Graphite's good electrical and thermal conductivity is explained by the high atomic density within layers and the free interlayer space, which ensures electron mobility and high electrical conductivity in an electric field. Another important property of graphite, important for this study, is that when heated, graphite does not melt; it burns only in a stream of oxygen, and evaporates in an electric arc under non-oxidizing conditions, i.e., it sublimates.

Graphite is used to manufacture a variety of specialized products, including refractory materials, solid lubricants, electrical machine contacts, paints, pencil leads, battery electrodes, and much more. But the most important aspect of graphite today is its growing demand and widespread use in nuclear power, the chemical industry, the production of conductive rubber, and metallurgy (as a material for electrodes, crucibles, and refractory screens).

Natural graphite contains various chemical impurities: SiO_2 , Al_2O_3 , P_2O_5 , MgO , CuO , FeO , CaO . This limits its use and requires refining to remove these impurities. This is associated with enormous costs, comparable to those of producing synthetic

graphite. Therefore, a significant portion of industrial graphite is synthetic.

Coke and pitch (petroleum and wood tar, coal tar) are used as raw materials in the production of artificial graphite. Given the demand for graphite, various technologies were developed for producing artificial graphite, which differs from natural graphite in chemical purity and applications. Common types of artificial graphite include: Acheson graphite, produced by heating a mixture of pitch and coke to 2800°C in electric furnaces; Pyrolytic (retort) graphite, designed for the electrical industry and synthesized from a gaseous hydrocarbon; Blast furnace graphite, formed as a by-product during the cooling of cast iron; and Carbide graphite, produced by the thermal decomposition of carbides.

The most cost-effective technology for producing graphite is coke. The primary raw material is coal coke. This intermediate product is produced by heating coal and peat to $1000\text{--}1100^\circ\text{C}$ in the absence of oxygen. Coke is used as a high-energy fuel, a reducing agent in metallurgy, and a raw material for graphite production.

Technological features of artificial graphite production [[11], [12], [13], [14]]: Coke powder is fired under special conditions, ensuring carbon graphitization. To produce artificial graphite with desired properties, the graphite semi-finished product is impregnated with pitch, formaldehyde, or other substances. Often, to achieve the desired characteristics, graphite is subjected to heat treatment and impregnation several times. Each subsequent technological stage is carried out according to a specialized scheme. Graphite obtained by various methods differs and has a number of unique properties. These include increased strength, oxidation resistance, acid resistance, and high electrical conductivity. Artificial graphite of high chemical purity is produced in the form of a powder of various fractions. To produce parts by molding, the powder is pressed and sintered using specialized technologies [[15], [16]].

Graphite manufacturers, for example JSC "Donkarb Graphite", produced all types and grades of artificial graphite for industry. This is a colloidal powder of all possible fractions (fine, medium, coarse-grained), pressed, with special properties (grades ATM, OSCh MG, GMZ, PPG and others).

Global demand for graphite products is growing exponentially [[17], [18]]. Increasing temperature limits and thermomechanical loads in new

technologies place increased demands on graphite quality. The carbon industry's development strategy is focused on creating new, durable graphite materials with unique physical and chemical properties. Leaders in this area of industrial development include China, the United States, Japan, and the Netherlands.

The share of imported graphite in Kazakhstan's graphite consumption is expected to increase. Therefore, the development of graphite production technologies based on high-purity graphite is relevant and aligns with the priority areas of scientific and technological development.

The aim of this research work is to create a plasma-chemical technology for producing graphite from coal based on sublimation and desublimation processes in a single reactor with the separation of impurities during the graphitization of carbon atoms on a graphite catalyst.

Methods

This publication presents some of the results of work conducted at the Research Laboratory of High-Temperature Synthesis of Composite Materials, M. Auezov South Kazakhstan University. The technological features of the graphitization process were determined, graphite synthesis tests were conducted from coal using a simple laboratory setup, and a carbon graphitization reactor based on these results was designed using coal from Kazakhstan. An application for a patent of the Republic of Kazakhstan "Method for producing graphite from an air suspension of coal particles and a device for implementing it" was filed. Registration No. 2025/1516.2 dated November 15, 2025.

Based on the achievements of scientists in the field of synthesis of artificial graphite [[2], [3], [4], [6], [8]] and experience of the article authors [9,10], a process flow chart and equipment for obtaining artificial graphite from Shubarkol coal in Kazakhstan were developed. The technology utilizes the properties of carbon and its modifications. One of them is the ability of carbon atoms to sublime when heated in a non-oxidizing environment, for example, in carbon dioxide at a temperature above 2000°C. (Sublimation is the transformation of a solid into a gaseous substance without going through the liquid state). The reverse process, desublimation, in this case is graphitization – the formation of graphite from atomic carbon. The graphitization process is

widely known in scientific practice. It is important to note that, depending on the conditions, this process can occur over a wide temperature range, for example, from 500°C to 2000°C [[13], [14], [16], [18], [19], [20], [21], [22], [23], [24], [25], [26]]. For this reason, the work utilizes a graphitization catalyst, high-purity graphite, which allows to expand the temperature range of the graphitization process. Graphitization of atomic carbon on the surface of the catalyst – the centers of graphitization nucleation, at high temperatures, when all impurities are still in a gaseous state, allows the separation of graphite from impurity atoms. The plasma reactor (Figure 1) is made of graphite. A powdered catalyst, in the form of graphite powder, is fed into the graphitization zone as a seed for artificial graphite produced by desublimation of atomic carbon. Artificial graphite particles not captured in the separator are fed with the gas flow to a hydrofilter, where the graphite dust is captured with a special aqueous solution. Purified carbon dioxide is returned to the process. The resulting graphite from all graphitization zones and from the water filter is periodically extracted, analyzed, and sorted.

Reference: "Shubarkolkomir (coal)" / Shubarkol – the black pearl of Sary-ArkaShubarkol. The industrial coal reserves of the Shubarkol deposit amounted to over 1.5 billion tons. Coal from the Shubarkol deposit is classified as grade D (long-flame) hard coal and contains very little ash. The ash content of Shubarkol coal is only up to 12 percent, and the ash content of individual coal seam layers was only 3-6%. The coal has a low sulfur content (up to 0.5%) and a high calorific value (from 5200 to 5700 kcal/kg), and produces a lot of heat when burned [[1], [2], [17]]. Based on these properties, Shubarkol coal was chosen as the main raw material for producing artificial graphite using the plasma-chemical method.

The chemical composition of Shubarkol coal was studied by mass spectral analysis at the Regional Testing Laboratory of Engineering Profile "Structural and Biochemical Materials" (M. Auezov South Kazakhstan University), the results are presented in Table 1.

Given the complexity of the plasma-chemical graphite production process, the coal is sorted by ash content, and samples with the lowest ash content are selected (Table 1). A sample of Shubarkol coal after milling is shown in Figure 2.

Table 1 - Chemical composition of Shubarkol coal samples: Carbon 97-97.5%

| No. | Chemical element | Content of impurities, % - Content in coal | Ash, impurity content | Ash content, % - content in ash | Note |
|-----|------------------|--|--------------------------------|---------------------------------|--|
| 1 | Si | 1.33-1.44 | SiO ₂ | 56-57 | When burned in oxygen, some of the silicon evaporates in the form of SiO |
| 2 | Al | 0.88-0.93 | Al ₂ O ₃ | 22-22.6 | |
| 3 | Fe | 0.15-0.20 | Fe ₂ O ₃ | 7.11 | |
| 4 | Ca | 0.08-0.11 | CaO | 2.6-2.8 | |
| 5 | Mg | 0.02- 0.03 | MgO | 1.8-2.0 | |
| 6 | S | 0.04-0.06 | SO ₃ | 3.4-3.43 | |
| 7 | K | 0.04-0.06 | K ₂ O | 1.28-1.3 | |
| 8 | Na | 0.06-0.07 | Na ₂ O | 1.65-1.72 | |

To ensure the graphitization of atomic carbon produced in the electric arc plasma torch, a catalyst for the physicochemical process is fed into the graphitization zone. In this study, a graphitization catalyst made of high-purity graphite powder was used. This allowed for an expanded temperature range for the graphitization process. The plasma reactor is made of graphite; in the graphitization zones, a powdered graphite catalyst and a graphite plasma reactor ensure the quality of the artificial graphite produced by desublimation of atomic carbon. A particle separation system for the artificial graphite separates the resulting products by quality, traps heavy impurities from the gas stream, and separates graphite dust with a special aqueous solution. Purified carbon dioxide is then returned to the beginning of the process – to the plasma-chemical reactor.

The resulting graphite from all graphitization zones and from the water filter is periodically extracted, analyzed, and sorted.

This method of producing graphite from coal is promising and important for the creation of products from artificial graphite: electrodes in electrochemistry, in the production of electric batteries, protective screens in nuclear energy, sorbents in the form of thermally expanded graphite in the petrochemical industry.

Features of the plasma-chemical synthesis process for graphite from Shubarkol coal: To efficiently produce graphite by desublimation, a powdered catalyst – “OSCh 7-3” grade graphite – is fed into the reactor’s graphitization zone (position 5, Figure 1). On the surface of the catalyst particles, for which high-purity graphite powder was selected, the graphitization process occurs over a wide temperature range, significantly increasing the reactor’s graphitization zone and, consequently, the

duration of the graphitization cycle, which is crucial for the complete conversion of coal to graphite.

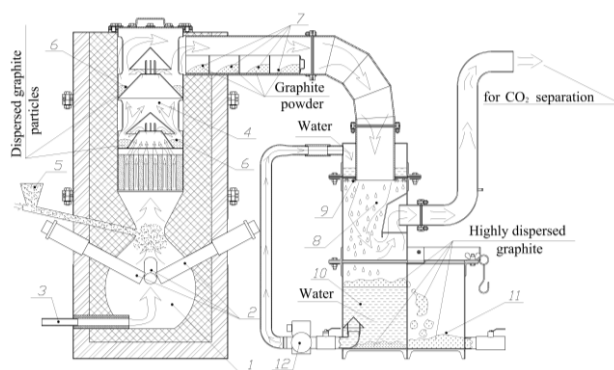


Figure 1 - Schematic diagram of the plasma-chemical apparatus for obtaining graphite from coal of the Shubarkol deposit in the Republic of Kazakhstan:

1 – the plasma reactor chamber, 2 – the graphite electrodes of the plasma torch, 3 – the tuyere for feeding reagents into the reactor, 4 – the dome – a dust collector of gas flows in the graphitization zone, 5 – the feeder for feeding the graphitization catalyst into the reactor, 6 – the cassettes for collecting the obtained graphite, 7 – the separator: storage tanks for synthesized graphite powder, 8 – the water shower chamber, 9 – the water spray nozzles, 10 – the water sump, 11 – the graphite storage tank, 12 – the water pump with a filter

The catalyst was prepared from “OSCh 7-3” grade graphite bars by crushing and grinding, followed by fractional separation. To use the powdered catalyst in a plasma-chemical graphite synthesis reactor, it is necessary to select a graphite particle size that, upon entering the high-temperature reactor zone, does not completely sublime but remains solid and acts as artificial nuclei for the desublimation of atomic carbon obtained from the carbon particles in the sublimation zone. These conditions reduce the energy barrier for the phase transition of atomic carbon from the gas phase to graphite. For this

purpose, the yield of artificial graphite was determined using catalyst of different fractions. It was found that, for the conditions of this plasma-chemical apparatus, a fraction of 0.3-0.5 mm in cross-section is optimal.

Research Results and Discussion

The raw material – Shubarkol coal powder, specially processed to a fraction smaller than 0.3 mm – is fed into the plasma zone of a carbon dioxide reactor. The high heating rate in the reactor's plasma flow ensures the sublimation of carbon atoms. This results in an ionized gas environment consisting of carbon ions and oxygen ions. A catalyst – “OSCh 7-3” grade graphite powder – is fed into this ionized environment. The catalyst acts as an activator for the graphitization of carbon ions on its surface. The atomic structure of the graphitized product corresponds to that of the graphite catalyst. Samples of the resulting graphite are shown in Figure 2 (d).

Using a graphite catalyst with the required structure, pyrolytic graphite is produced by depositing carbon from the gas phase at high temperatures. It is characterized by high purity and anisotropic properties. (Thermally (chemically) expanded graphite – TEG and TCEG).

The chemical composition of plasma-chemically synthesized graphite is shown in Table 2.

Table 2 - Carbon and impurity content in synthesized graphite (average value of 7 samples from each graphitization zone of the plasma-chemical reactor)

| No. of graphitization zone | Content of carbon, % | Content of impurities, % | Note |
|----------------------------|----------------------|--------------------------|--|
| 1 | 99.82 | 0.18 | Samples were taken from zones 6, 7, and 11: The purest graphite, sample 2, was from zone 7 (Figure 1), sample 1 was taken from zone 6, and samples 4 and 5 were from zone 11 (Figure 1). This is due to the different condensation temperatures of impurities from the reactor's gas environment, which requires further study and analysis of the graphitization process in such a reactor. |
| 2 | 99.94 | 0.06 | |
| 3 | 99.77 | 0.23 | |
| 4 | 98.68 | 1.32 | |
| 5 | 98.65 | 1.35 | |

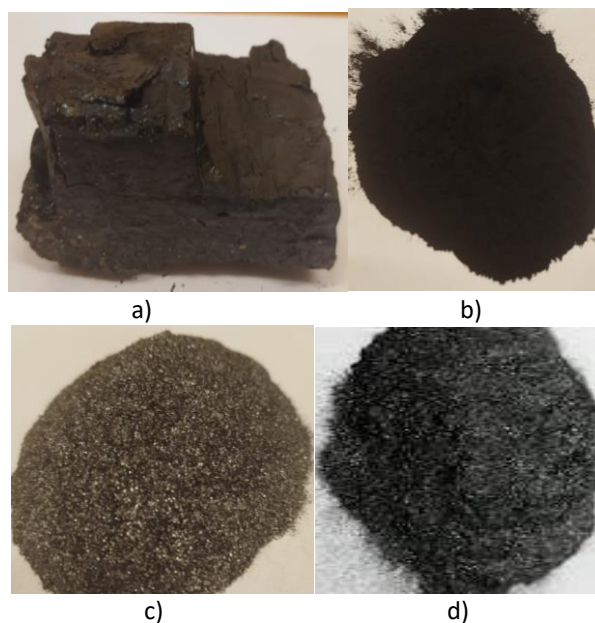


Figure 2 - Photos: a) Shubarkol coal, b) Shubarkol coal powder, c) Graphite – Catalyst, d) Graphite – graphitized by desublimation of carbon atoms on the catalyst.

The method and apparatus design proposed in this article will enable the production of high-purity graphite with a catalyst-like structure. Such processes require study using a newly developed plasma-chemical facility capable of controlling graphitization conditions and separating graphitization products in various temperature ranges for desublimation of atomic carbon.

Conclusions

A high-purity graphite catalyst was used in the study to expand the temperature range of the graphitization process. A graphite plasma reactor ensures the quality of the synthetic graphite produced by desublimation of atomic carbon.

The artificial graphite particle separation system allows for the separation of the resulting products by quality, the capture of heavy impurities from the gas flow, the separation of graphite dust with a special aqueous solution, and the return of purified carbon dioxide back into the process.

The resulting graphite from all graphitization zones and the water filter is sorted and analyzed. It was established that the graphite obtained in the plasma-chemical reactor zone is purer of impurities, indicating the important role of process temperature. Furthermore, as the graphite moves away from the plasma torch, it appears to adsorb impurities.

This method of producing graphite from coal is promising and important for the creation of artificial

graphite products: electrodes in electrochemistry, in the production of electric batteries, protective shields in the nuclear power industry, and expanded graphite sorbents in the petrochemical industry. The graphitization process requires continued research to ensure high product quality and address process control issues.

Conflict of Interest. On behalf of all authors, the corresponding author declares that there is no conflict of interest.

CRedit author statement: Zh. Aimenov: Supervision and overall guidance of the research; A. Protopopov: Conceptualization, Methodology, Software; A. Saipov: Data Curation, Writing – Original Draft Preparation; E. Suleimenov: Visualization, Investigation; M. Protopopov: Software, Validation of results; A. Merekeyeva: Review and Editing of the English version of the text.

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Көміртек атомдарын газ фазасына сублимациялап, содан кейін жоғары таза графитке десублимациялау арқылы Шұбаркөл көмірінен алынған жасанды графит

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Коммерциялық емес акционерлік қоғамы М.Әуезов атындағы Оңтүстік Қазақстан университеті, Шымкент, Қазақстан

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ТҮЙІНДЕМЕ

Мақалада Қазақстанның Шұбаркөл кен орнының күлі аз көмір бөлшектерінің ауа суспензиясынан тазалығы жоғары графитті алудың плазмохимиялық әдісі қарастырылады. Технологиялық процес сұйық күйді айналып өтіп, қатты күйден газға айналатын көміртектің ерекшеліктерін пайдалануға негізделген, яғни жоғары температурада сублимацияланады, ал реактор аймағындағы газ ортасының температурасы төмендеген кезде десублимацияланады. Графит катализаторын пайдалану графиттелетін материалдың түзілуін басқаруға мүмкіндік береді. Атомдық көміртектең графитациясы температураның кең диапазонында жүреді. Жоғары температуралы реактор аймақтарында алынған графит 500°C-қа жақын реактор аймақтарында алынған графитке қарағанда таза екендігі анықталды. Графитация процесінің бұл ерекшелігі өнімді сапасы бойынша жіктеуге мүмкіндік береді. Графит алу үшін сублимация және десублимация процестеріне негізделген реактордың дизайны қарастырылады. Жоғары жиілікте электромагниттік әсер ететін аймақтың плазмохимиялық реакторының дизайнында пайдалану графит ұнтағы катализаторындағы десублимация процестерін күшейту арқылы атомдық көміртектең графитациясын басқаруға мүмкіндік береді. Плазмохимиялық аппараттың конструкциясында көміртегі ұнтағы, графит ұнтақ катализаторы және көмір қышқыл газы бар бастапқы шикізаттағы компоненттер пропорцияларының ауытқуы кезінде мүмкін болатын шаң жинау және көміртегі оксидін бейтараптандыру жүйесі қарастырылған. Жасалған аппаратта технологиялық жұмыс режимін өзгерту арқылы графиттен сорбент - термохимиялық кеңейтілген графит алуға болады. Зерттеудің мақсаты - графит катализаторында көміртек атомдарын графиттеу кезінде қоспаларды бөліп, бір реакторда сублимация және десублимация процестеріне негізделген көмірден графит алудың плазмалық-химиялық технологиясын жасау.

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| | Түйін сөздер: плазма, сублимация, десублимация, графиттеу, көмір, графит, бөлу, катализатор. |
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Искусственный графит из Шубаркольского угля, полученный сублимацией атомов углерода в газовую фазу с последующей десублимацией в графит высокой чистоты

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| Поступила: 17 октября 2025 Рецензирование: 11 ноября 2025 Принята в печать: 18 ноября 2025 | АННОТАЦИЯ В статье рассматривается плазмохимический метод получения высокочистого графита из воздушной взвеси частиц малозольного каменного угля Шубаркольского месторождения Казахстана. Основа технологического процесса заключена в использовании особенности углерода превращаться из твердого состояния в газообразное, минуя жидкое состояние, то есть сублимировать при высокой температуре, а при снижении температуры газовой среды в зоне реактора десублимировать. Использование катализатора из графита позволяет управлять формированием графитизируемого материала. Графитация атомарного углерода происходит в широком диапазоне температур. Установлено, что графит, полученный в зонах реактора с высокой температурой более чистый, чем графит, получающийся в зонах реактора близких к 500°C. Эта особенность процесса графитации предоставляет возможность классифицировать продукт по качеству. Рассмотрена конструкция реактора, на основе процессов сублимации и десублимации, для получения графита. Использование в конструкции плазмохимического реактора зоны с высокочастотным электромагнитным воздействием позволяет управлять графитацией атомарного углерода, интенсифицируя процессы десублимации на графитовом порошковом катализаторе. В конструкции плазмохимического аппарата предусмотрена система пылеулавливания и нейтрализации угарного газа, возможного при отклонениях пропорций компонентов в исходном сырье, в котором присутствует угольный порошок, графит порошковый катализатор и углекислый газ. В созданном аппарате возможно получать из графита сорбент - термохимически расширенный графит, изменяя технологический режим работы. Целью исследования является создание плазмохимической технологии получения графита из угля на основе процессов сублимации и десублимации в одном реакторе с отделением примесей при графитации атомов углерода на катализаторе из графита. |
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| | Ключевые слова: плазма, сублимация, десублимация, графитация, уголь, графит, сепарация, катализатор. |
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