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Study of refractory raw materials of the Republic of Kazakhstan

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ABSTRACT

Important aspects of increasing the competitiveness of domestic metallurgy and mechanical engineering are increasing the durability of thermal units and involving non-traditional, cheaper energy resources in production. One of these resources is carbon waste from the aluminum industry - electrode scrap and waste from the carbon lining of electrolyzers. The problem with their use as substitutes for solid fuels (coke coal) in the metallurgical, engineering, and energy industries is fluorine- and alkali-containing salts that impregnate them, destroying the traditional lining of thermal units. The development of effective refractories resistant to fluorine- and alkali-containing corrodents (melts and gases) makes it possible to increase the competitiveness and efficiency of thermal units both through the use of cheaper energy carriers and by increasing the duration of their campaign. An important aspect is the simultaneous disposal of hazardous industrial waste and the reduction of the environmental burden on the ecosystem of the Republic of Kazakhstan. The basis for the development of new refractory materials resistant to fluorine- and alkali-containing corrodents is the analysis of the existing raw material base of the Republic of Kazakhstan and the choice of materials that make it possible to obtain aluminosilicate refractories with increased chemical resistance. In the work, the phase and chemical composition of refractory clays and kaolins of the Republic of Kazakhstan were studied, and their rheological and thermophysical properties were investigated. Selected raw materials for the development of technology for the production of dense aluminosilicate refractory products.

Keywords: aluminosilicate refractories, refractory clays, kaolins, chamotte, plasticity, sintering, open porosity, water absorption.

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Introduction

The most important task of the metallurgical and energy complexes of the Republic of Kazakhstan is to increase the competitiveness of products and industries, with the maximum possible combination of interests of the Republic, its regions, and specific joint-stock companies.

Among the main manufacturing industries of the Republic of Kazakhstan, an important place is occupied by metallurgy (43.7% of GDP) and

mechanical engineering (12.3% of GDP) [[1], [2]]., while the metallurgical and machine-building complex remains energy- and resource-intensive, where at least 15% of primary fuel, 35% of electricity, 40% of fossil raw materials are used. Reducing costs through the involvement of secondary resources in the production, and waste disposal is the most urgent task for the development of the industry of the Republic of Kazakhstan.

Currently, the cost of coke fluctuates around \$550-600/t, and the cost of anthracite is at the level of \$412-418/t at the shipping station [[3], [4]]. Replacing coke with cheaper fuel will improve the technical and economic performance of thermal units.

It should also be considered that the expansion of the use of secondary raw materials, including carbon-containing ones, is an important factor in the efficiency of the entire industry of the Republic of Kazakhstan [[4], [5]]. On the other hand, JSC "Kazakhstan Electrolysis Plant" (KEP), which is part of ENRC (Eurasian Natural Resources Corporation) (Pavlodar), annually generates up to 1,500 tons of waste carbon refractory lining and up to 25,000 tons of cinders of baked anodes [[6], [7]]. with a minimum carbon content of 85 wt. % contaminated with harmful substances from cryolite-alumina melt. The presence of alkaline cations (Na +, K +) and fluorine in the waste do not allow storing them in sludge fields to avoid a negative impact on the environment. The simplest and most efficient way to dispose of such wastes is to use them as fuel instead of expensive and scarce coke in the metallurgical, engineering, or energy industries, where fluorine and alkaline salts will also help to liquefy the slag, saving more on use fluorite slag modifiers [[8], [9]].

At the same time, the presence of alkali cations and fluorine anions in the composition of the melting charge significantly complicates the service conditions of the aluminosilicate lining of thermal units.

The solution to this problem can be the development of the densest (non-porous) aluminosilicate refractories, which requires researching the raw material base of aluminosilicate refractories of the Republic of Kazakhstan, choosing the most promising raw materials, and developing technology for dense refractory products with a maximum mullite content with high chemical resistance.

The production of aluminosilicate should include the production of a dense, durable mullite-containing aggregate (chamotte) with water absorption of no more than 1%, the preparation of a binder of plastic clay, and the production of actually dense products with an open porosity of no more than 5%.

Raw materials for the production of mullite-containing refractories can be [[10], [11]]: clays and kaolin, natural high-alumina raw materials (bauxites, aluminosilicates), and synthetic materials

(commercial alumina, tabular alumina, fused corundum, reactive alumina).

At present, deposits of refractory clays, kaolin, and bauxites are known in the Republic of Kazakhstan, and it also has the production of technical alumina [[12], [13]].

The objects of study in this work are three types of refractory clay raw materials: clays of the Arkalyk (AG) and Berinsky (BG) deposits and kaolin of the Alekseevsky (AK) deposits, selected on the basis of an analysis of the physicochemical and technological properties of refractory raw materials the Republic of Kazakhstan.

The Arkalyk deposit of refractory clays is located in the Arkalyk district of the Torgai region, in the suburbs of the city of Arkalyk. The characteristic features of the deposit are the unsustainable thickness of the layers, a sharp transition from one variety to another, as well as a predominantly dry type of clay products. Refers to highly basic raw materials with Al_2O_3 content up to 48 wt. % per calcined substance and an average content of coloring oxides ($Fe_2O_3 + TiO_2$) of 4.6 wt. %. According to the mineral composition, clay is a polymineral raw material with a predominant content of kaolinite and gibbsite. The coarsely dispersed part contains quartz and hematite. Part of the iron is associated with clayey rocks (mainly with gibbsite). The deposit is actively developed and used to produce lumpy refractory chamotte.

The Berlin deposit of refractory clays is located on the border of the Komsomolsky district of the Kostanay region and the Troitsky district of the Chelyabinsk region, 22 km west of the Buskol railway station. A characteristic feature of the deposit is a horizontal reservoir deposit of kaolinite-hydromicaceous clays of variable thickness from 0 to 9 m. It belongs to the main raw material with an Al_2O_3 content of up to 32 wt. % and an average content of coloring oxides ($Fe_2O_3 + TiO_2$) of 2.9 wt. %. According to the mineral composition, clay belongs to the monomineralic kaolinite-hydromicaceous raw material. Quartz is presented as an impurity. The birthplace is actively developed and used to produce both fired fireclay and plastic binder clays [[14], [15]].

The Alekseevskoye kaolin deposit is located in the Kokshetau district of the North Kazakhstan region, 33 km north of the city of Kokshetau, southeast of the village of Alekseyevka. A characteristic feature of the deposit is the variable power of the lenses lying in at various levels, a smooth transition of color from white to grayish-

yellowish, as well as a predominantly harsh type of rocks. Along with crackers, there are also loose varieties. Refers to a highly basic raw material with an Al_2O_3 content of up to 44 wt. % and an average content of coloring oxides ($\text{Fe}_2\text{O}_3 + \text{TiO}_2$) 1.1 wt. %. According to the mineral composition, the raw material is represented by kaolin, quartz, and feldspar impurities. The deposit is being developed and used to obtain enriched kaolin, quartz-kaolinite mixtures and quartz-feldspar concentrate [[16], [17]].

The experimental part

The results of assessing the granulometric composition of clay raw materials by the sedimentation method according to GOST 21216-2014 are given in Table 1 and in (figure 1), allow us to classify Berlin clay as a highly dispersed raw material with a particle content <0.001 mm of more than 99%, and Arkalyk clay and Alekseevsky kaolin as a medium-disperse raw material with a particle content <0.001 mm of less than 60%.

According to the Okhotin diagram, clay from the Berlin deposit belongs to the group of plastic raw materials, clay from the Arkalyk deposit and kaolin from the Alekseevsky deposit belong to the group of low-plastic clay raw materials.

Table 1 - Granulometric composition of the studied raw materials

Raw material	Content of particles, wt. %, size, mm					
	1.00-0.25	0.25-0.06	0.06-0.01	0.01-0.005	0.005-0.001	< 0.001
Arkalyk clay	7.0	1.8	23.2	9.6	17.7	31.6
Berlinsk clay	0	0	12.0	5.2	12.4	70.3
Alekseevskiy kaolin	5.5	14.5	31.6	7.5	24.4	16.5

The mineralogical composition of the clay raw material was evaluated by X-ray phase analysis performed on a Miniflex 600 diffractometer (CuK α radiation, $\lambda=1.541862$ Å, survey interval – 3.00–60.00°, scanning step – 0.02°). The samples were taken without heating in an air atmosphere at a counter speed of 1 deg/min.

Analysis of the obtained data shows that the phase composition of the Arkalyk clay is represented mainly by kaolinite, with an admixture of gibbsite, and quartz and hematite are present as

non-plastic impurities; Berlin clay - kaolinite; quartz, hydromica, and montmorillonite; Alekseevsky kaolin - kaolinite; quartz, hydromica and calcium feldspar.

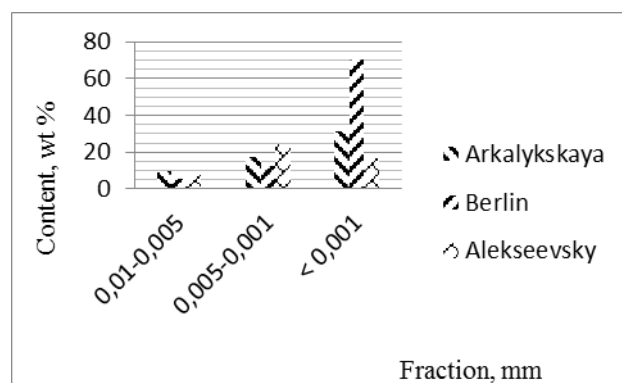


Figure 1 – Histogram of distribution of particles by fractions and raw materials under study

The main technological properties of aluminosilicate raw materials to produce refractories include plasticity, sintering, air and fire shrinkage, and fire resistance [[18], [19]].

The plastic properties of the studied raw materials were evaluated by the Atterberg plasticity number (GOST 21216-2014), which is the difference in the percentage of water at the upper and lower limits of plasticity, i.e., at the boundaries of the transition of clay paste from plastic to fluid and semi-solid condition. The plastic properties of the studied clays are given in Table. 2.

Table 2 - Plasticity of the investigated raw materials

Raw material	Limit fluidity, %	The border rolling, %	Number plasticity	Classification according to GOST 9169-2021
AC	33.33	20.00	13.33	moderately plastic
BC	33.41	15.35	18.06	medium plastic
AK	25.34	12.01	13.27	moderately plastic

AC – Arkalyk clay BC – Berlin clay; AK – Alekseevskiy kaolin

From the analysis of the data in Table 2, it can be seen that Berlin clay belongs to plastic raw materials, and Arkalyk clay and Alekseevsky kaolin belong to moderately plastic raw material.

In the production of products based on clay raw materials, drying is the most important technological factor. The choice of the drying mode is largely determined by the properties of the mass, the shapes, and the sizes of the products.

The determination of the sintering properties of the studied clays was carried out according to GOST 21216-2014. From an average test sample with a particle size of less than 1 mm, mixed with water to the state of a working dough, samples were made in the form of tiles 50 × 50 mm and cubes 25 × 25 mm in a plastic way, dried to an air-dry state, after which they were fired in a laboratory furnace with silicate heaters from 1000 to 1550 C with an interval of 50 C and holding at the final firing temperature for 2 hours.

Table 3 - Technological properties of the investigated aluminosilicate raw materials

Properties	AC	BC	AK
Number plasticity	14	24	5.0
Connectivity, MPa	5.0	5.6	3.5
Binding capacity (% standard sand)	45	65	-
Sensitivity to drying, Kch	1.95	3.4	0.55
Air shrinkage, %	1.4	2.5	0.3
Sintering start temperature, C	1350	1200	1350
Fire resistance, °C	> 1720	1630	>1720

In addition, one of the most important indicators of the quality of refractory clays is fire resistance, and technological properties - binding capacity, sensitivity to drying, and air shrinkage [[20], [21]].

Under certain test conditions, refractoriness depends only on the chemical composition and

partly on the mineralogical composition, that is, it characterizes the purity of the raw material under study. The results of calculating the refractoriness and some technological properties of the studied clay raw materials are given in Table. 3.

Conclusions

Features of granulometric (high content of fine particles), chemical and mineralogical compositions, as well as the state of the structure (a disorder of kaolinite) will favorably affect the sintering of the studied aluminosilicate raw materials. At the same time, Arkalyk clay can be recognized as the most promising in the technology of superdense aluminosilicate refractories as a raw material to produce fireclay, the monomineral composition of which and the disorder of kaolinite will favorably affect both its rheological and ceramic properties. The high plasticity of Berlin clay allows it to be used as raw material, both to produce fireclay and for use as a binder clay.

Thus, according to the totality of the considered properties, Arkalyk and Berlin clays, as well as Alekseevsky kaolin, are of practical interest for the production technologies of aluminosilicate refractory materials. At the same time, Arkalyk clay and Alekseevsky kaolin are promising in the technology of aluminosilicate refractory products as a raw material to produce fireclay, and Berlin clay can be used in the technology of aluminosilicate refractories as a binder.

Conflict of interest

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

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Қазақстан Республикасының отқа төзімді шикізатын зерттеу

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ТҮЙІНДЕМЕ

Отандық металлургия мен машина жасаудың бәсекеге қабілеттілігін арттырудың маңызды аспектілері жылу қондырғыларының ұзақ мерзімділігін арттыру және өндіріске дәстүрлі емес, арзанырақ энергия ресурстарын тарту болып табылады. Осы ресурстардың бірі алюминий өнеркәсібінің көміртекті қалдықтары – электрод сынықтары мен электролизерлердің көміртекті қаптамасының қалдықтары. Оларды металлургия, машина жасау және энергетика салаларында қатты отынның (кокс көмірі) алмастырғыштары ретінде пайдалану мәселесі термиялық қондырғылардың дәстүрлі төсемдерін бұзатын, оларды сіңіретін фтор және сілті бар тұздар болып табылады. Құрамында фтор және сілті бар коррозияға (балқымалар мен газдар) төзімді, тиімді, отқа төзімді материалдарды әзірлеу арзанырақ энергия тасымалдаушыларды пайдалану арқылы да, олардың науқанының ұзақтығын ұлғайту арқылы да жылу қондырғыларының бәсекеге қабілеттілігі мен тиімділігін арттыруға мүмкіндік береді. Қауіпті өндірістік қалдықтарды бір мезгілде көму және Қазақстан Республикасының экосферасына экологиялық жүктемені азайту маңызды аспект болып табылады. Құрамында фтор және сілті бар коррозияға төзімді жаңа отқа төзімді материалдарды әзірлеудің негізі Қазақстан Республикасының қолданыстағы шикізат базасын талдау және химиялық тұрақтылығы жоғары алюмосиликатты отқа төзімді материалдарды алуға мүмкіндік беретін материалдарды таңдау болып табылады. Жұмыста Қазақстан Республикасының отқа төзімді саздары мен каолиндерінің фазасы мен химиялық құрамы зерттеліп, олардың реологиялық және термофизикалық қасиеттері зерттелді. Тығыз алюмосиликатты отқа төзімді бұйымдарды алу технологиясын жасау үшін іріктелген шикізат.

Түйін сөздер: алюмосиликатты отқа төзімді заттар, отқа төзімді саздар мен каолиндер, шамот, пластикалық, агломерация, ашық кеуектілік, суды сіңіру.

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АННОТАЦИЯ

Важными аспектами повышения конкурентоспособности отечественной металлургии и машиностроения является повышение стойкости тепловых агрегатов и вовлечение в производство нетрадиционных, более дешёвых энергоресурсов. Одним из таких ресурсов являются углеродистые отходы алюминиевой промышленности – электродный бой и отходы углеродистой футеровки электролизеров. Проблемой их применения в качестве заменителей твердого топлива (угля кокса) в металлургической, машиностроительной и энергетической промышленности являются пропитывающие их фтор- и щелочесодержащие соли, разрушающие традиционную футеровку тепловых агрегатов. Разработка эффективных огнеупоров, стойких к воздействию фтор- и щелочесодержащих корродиентов (расплавов и газов) позволяет повысить конкурентоспособность и экономичность тепловых агрегатов как за счёт использования более дешёвых энергоносителей, так и за счёт повышения длительности их кампании. Немаловажным аспектом является одновременная утилизация вредных промышленных отходов и снижение экологической нагрузки на экосферу Республики Казахстан. Основой разработки новых огнеупорных материалов стойких к воздействию фтор- и щелочесодержащих корродиентов, является анализ существующей сырьевой базы Республики Казахстан и выбор материалов, позволяющих получить алюмосиликатные огнеупоры с повышенной химической стойкостью. В работе исследованы фазовый и химический состав огнеупорных глин и каолинов РК, исследованы их реологические и теплофизические свойства. Выбраны сырьевые материалы для разработки технологии производства плотных алюмосиликатных огнеупорных изделий.

	Ключевые слова: алюмосиликатные огнеупоры, огнеупорные глины и каолины, шамот, пластичность, спекаемость, открытая пористость, водопоглощение.
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