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ОБЗОР СПОСОБОВ И МЕТОДОВ ПЕРЕРАБОТКИ ОТВАЛА ЛЕЖАЛЫХ КЛИНКЕРОВ ЦИНКОВОГО ПРОИЗВОДСТВА

Ж.У. Гаппаров[1], С. Сырлыбеккызы[1], А.Э. Филин[2], А.С. Колесников[3], Е.Т. Жатканбаев[3]

[1] Каспийский университет технологий и инжиниринга им. Ш. Есенова, Актау, Республика Казахстан
[2] НИТУ МИСИС, Москва, Россия
[3] Южно-Казахстанский университет им. М. Ауэзова, Шымкент, Республика Казахстан, e-mail: kas164@yandex.kz

Аннотация: Представлен анализ методов и подходов к утилизации техногенных отходов – лежалых клинкеров цинкового горнорудного производства, которые накоплены в достаточно больших объемах в техногенных отвалах по всему миру. Подробно рассматриваются исследования ученых ряда стран относительно способов и методов их переработки. В результате проведенного анализа было установлено, что среди множества описанных способов и методов утилизации техногенного отхода горнорудного производства, в частности применительно к лежалым клинкерам Ашисая, нет ни одного комплексного технологического решения, отличающегося полнотой переработки или утилизации, все попытки по переработке отвальных клинкеров цинкового производства подразумевали повторное обогащение или доизвлечение с образованием вторичных отвалов и хвостов обогащения. Лежалые клинкеры начали образовываться и накапливаться в отвалах, начиная с 1927 по 1995 гг. Так, на открытой горной местности в Туркестанской области близ поселка Ашисай на настоящий момент лежалые клинкеры остаются складированными в отвале в объеме 7–7,5 млн т, оказывая негативное воздействие на окружающую среду. На основе проведенных исследований был определен элементно-химический состав техногенного отвала, по результатам которого было установлено, что лежалые клинкера (шлаки) цинкового производства содержат в своем химическом составе соединения кальция, кремния, железа, алюминия, углерода и тяжелые цветные металлы, такие как цинк и свинец. По результатам исследований сформулировано заключение о поиске технологических решений утилизации техногенного отвала путем их комплексной переработки в качестве вторичного минерального сырья, в частности, в качестве железо-алюмо-силикатного компонента сырьевой смеси при получении портландцементного клинкера.

Ключевые слова: отвалы горнорудного цинкового производства, техногенное сырье, лежалый клинкер, цинк, свинец, антропогенное воздействие, пылевое загрязнение, техносферная безопасность, защита и охрана окружающей среды.

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Overview of techniques and methods of processing the waste of stale clinkers of zinc production

Jaken Gapparov[1], Samal Syrlybekkyzy[1], Alexander Filin[2], Alexandr Kolesnikov[3], Yergazy Zhatkanbayev[3]

[1] Sh. Yesenov Caspian University of Technology and Engineering, Aktau, Kazakhstan
[2] NUST MISIS, Moscow, Russia
[3] M. Auezov South Kazakhstan University, Shymkent, Kazakhstan,
e-mail: kas164@yandex.kz

Abstract: This article presents an analysis of methods and approaches to the disposal of man-made waste — stale clinkers of zinc mining, which are accumulated in sufficiently large volumes in man-made dumps around the world. The research of scientists from a number of countries on the methods of their processing is considered in detail. As a result of the analysis, it was found that among the many described methods of recycling man-made waste from mining, in particular with regard to stale Ashisai clinkers, there is not a single integrated technological solution that differs in the completeness of processing or disposal. All attempts to process zinc clinker waste products involved re-enrichment or additional extraction with the formation of secondary dumps and tailings of enrichment. Stale clinkers began to form and accumulate in dumps from 1927 to 1995. Thus, in an open mountainous area in the Turkestan region near the village of Ashisai, stale clinkers remain stored in the dump in the amount of 7–7.5 million tons, having a negative impact on the environment. Based on the conducted research, the elemental and chemical composition of the technogenic dump was determined, according to the results of which it was found that the stale clinkers (slags) of zinc production contain in their chemical composition compounds of calcium, silicon, iron, aluminum, carbon and heavy non-ferrous metals such as zinc and lead. Based on the results of the research, a conclusion was formulated on the search for technological solutions for the utilization of man-made waste by its complex processing as secondary mineral raw materials, in particular as an iron-aluminum-silicate component of the raw material mixture in the production of Portland cement clinker.

Key words: zinc mining dumps, man-made raw materials, stale clinker, zinc, lead, anthropogenic impact, dust pollution, technosphere safety, protection and environmental protection.

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Introduction

In the modern world, every year there is a decrease in the amount of metals in ores due to intensive mining and processing of ores rich in non-ferrous metals. The amount of waste from various industries is growing every year, and they contain both valuable metals and useful compounds. Their quantitative content often exceeds the content of certain metals in the extracted ores. [1]. Man-made waste from the zinc mining industry, located in dumps and containing various chemical compounds such as silicon, calcium, aluminum, iron and heavy metals, have a negative impact on the environment, including the well-being of human life, fauna and vegetation [2].

However, their chemical composition makes them a profitable raw material, and they can be used as auxiliary artificial raw materials in a number of different industries, such as metallurgy or chemical engineering. In Kazakhstan, starting from the 20s of the last century and up to the present, in a number of areas, including the East Kazakhstan and Turkestan regions, during the extraction of difficult-to-enrich zinc-containing ores and their use in the Weltz process, a significant amount of waste in the form of clinker was created, which is in the dump. These wastes were formed using mining plants for hard-to-enrich ores, and today these dumps are stored in the open air, which leads to contamination of soils, surface and groundwater, as well as the climate and the natural environment in general under weather and climatic conditions [3—5]. The predominant metals from these slags enter the body of living beings and humans both along the food chain and in the process of inhaling dust, having a harmful toxic effect [6—8]. In the Welz process of various natural and man-made materials, such as oxide ores, caps and slags, clinker slag is formed. Its value depends on the type of material being processed.

For example, clinkers slags obtained from zinc production cakes contains 1.2—2.1% Zn, 0.9—3.02% Cu, 0.4—0.78% Pb, as well as significant amounts of Ag and Au (0.3—0.6 kg/t). Clinker slags obtained during the Welz slag process of mine smelting of lead agglomerates contain 0.89—1.02% Zn, 0.12—0.35% Pb, 0.49—1.2% Cu and noble metals. clinkers slags of zinc production from Ashisai contain less copper and precious metals (“traces”), 0.59—0.72% Zn and 0.12—0.22% Pb. All clinker slags contain Si (from 11 to 18%) and Fe [9]. At the moment, according to various sources, the dumps contain from 7 to 7.5 million tons of technogenic waste from the process of Waelz processing of difficult-to-process zinc-containing oxide ores, which are located in Ashisai (Kazakhstan) [10, 11]. In this regard, the purpose of this study is to review the analysis of world scientific research in the field of methods and methods for complex disposal and processing of accumulated waste from the zinc mining industry.

Results and discussion

Analysis of existing methods of processing stale clinker (slags) from the processing of various industrial products

Clinker slag processing methods can be divided into two types:

1. Complex processing of stale clinker-slag obtained during the Welz processing of zinc-containing materials, with the extraction of non-ferrous and precious metals, a carbon component using iron-containing materials and non-metallic materials;

2. The use of clinker-slag in the Welz processing of zinc-containing materials as a raw material in various pyrometallurgical processes.

In the practice of copper production, a method for processing clinker slag with copper-zinc concentrates by agglomeration and mine smelting is known. The addition of 18—24% clinker slag to the charge leads to the formation of poor matte. An increase in the proportion of clinker slag in the sintering charge to 29—34% increases the temperature of the exhaust gases and complicates the operation of turbo extractors. This method has another drawback — the “smearing” of zinc and lead on processed products. For example, when processing clinker slag with copper concentrate at the Karabash copper plant, only 30% of Zn and 60% of Pb are converted to sublimations.

On an industrial scale, tests were carried out on the processing of copper-zinc concentrates and slags of current intake and dumps in a CALCET unit containing 2.23—5.75% copper, 0.67—1.68% lead, 0.81—1.37% zinc, 16.18—27.68% carbon, 26.61—30.47% iron, 5.47—6.05% sulfur, 14.08—15.1% silica, 1.41—2.37% lime. With the addition of 10% slag, the extraction of copper in matte was 91.21%. Zinc sublimates contained 72.17—72.97% zinc, 4.74—5.12 lead, 1.31—1.37 iron. Σ silica + lime — 0.98—1.3%, cadmium — 0.18—0%. However, despite this, the residual zinc content in the slags was 9.5—10.59%, lead — 0.29—0.42%.

Balkhashmed JSC carried out work on the use of clinker zinc instead of coal in the melting of low-sulfur concentrators in the Vanyukov furnace [12]. Elemental iron and carbon contained in clinker slag act as energy reagents in the liquid phase, which, when interacting with oxygen, release heat necessary for the melting process. In the presence of clinker slag, the copper content in the slag decreases, and zinc peroxidation is limited. Replacing coal with clinker slag does not cause technological difficulties in the operation of the Vanyukov furnace and helps to reduce the temperature of the exhaust gases in the recovery boiler and gaseous waste (by reducing or completely eliminating the burning of volatile coal components).

A method of melting slag after welding with the supply of an oxygen-containing blast to the slag layer and into the melt in a certain amount for the predominant oxidation of solid carbon in the slag in the first case and for the oxidation of metallic iron in the slag, excessive for binding sulfur in the slag and dissolving it in the matte phase, in the second case [13]. The disadvantages of this method are: — a small release of heat in the melt during the oxidation of excess metallic iron slag, which requires the use of external heat sources to maintain the melt in working condition. There is also a fundamental limitation on the maximum enrichment of the obtained matte with copper, and the impossibility, as a result, to obtain conditioned matte suitable for individual conversion when processing slag with a copper content of less than 2.49%. The share of such slag is more than 50% of all produced and accumulated in dumps. The latter circumstance significantly limits the possibilities of industrial application of this method.

Clinker slag from the Ukrzink plant was used as fuel in the processing of poor lead matte. As a result of melting the charge in a blast furnace, a matte with a content of 14.1—16 was obtained.2% copper and 9.1—10.3% lead, as well as distillates containing 16.8—26% zinc and 36% lead. However, the content of the sum (zinc, lead, copper) in the slag was 2.49—2.89% [14].

A raw material mixture has been developed for processing zinc-containing materials using clinker slag from the calcination of such materials and consisting of, by mass fraction: a carbon-containing reducing agent 5—9; calcium oxide 2—6; slag from calcination 3—10, and a material containing zinc — the rest. [15]. In this raw material mixture, slag plays the role of a reducing agent and at the same time a source of iron. Using this mixture allows you to: reduce coal consumption from 12—17% to 5—9%; increase zinc extraction by 0.8% from 96.1% to 96.9%; reduce electricity consumption by 50—110 kWh/t of zinc from 3710 kWh/t to 3650—3590 kWh/t and increase productivity by almost 10%.

Among the proposals for the independent processing of zinc slags and Achisai ore, it is worth noting the work of VNIITSVETMET, which conducted large-scale laboratory studies on the enrichment of clinker to produce coal concentrate with a carbon content of 51.5—60.2% when it is extracted by 86—95%. Magnetic separation of non-ferrous metal slag has also been studied. The indicators of the production of high-quality coal concentrate and the maximum extraction of iron into

Comparative data on the composition of slag and stone casting
Сравнительные значения состава шлакового и каменного литья

Parameters	Slag casting	Stone casting
Chemical composition:		
SiO_2	50 – 55	47 – 48
Al_2O_3	7 – 9	5 – 16
Fe_2O_3+FeO	7 – 8,5	15 – 16
CaO	12 – 14	11 – 12
MgO	5 – 6	6 – 7
Na_2O+K_2O	1 – 2	2 – 4
Cr_2O_3	0,5 – 1	–
TiO2	0,5 – 0,7	–
Mineralogical composition	Pyroxene augite	Pyroxene magnetite
Density, g/cm^3	2,8 – 3,0	2,9 – 3,0
Water absorption, %	0,003 – 0,1	0,16
Compressive strength, MPa	3 – 3,5	2,5
Abrasion, g/cm^3	0,05 – 0,08	0,08
Softening temperature, °C	1050	1000
Acid resistance,% H_2SO_4 HCl	98,8 – 99,3 99,5 – 99,7	99,8 99,3
Heat resistance	15 – 20	10 – 12
Crystal size, microns	50 – 80	150 – 200

a magnetic concentrate are optimal when using flotation in combination with magnetic separation. They contain up to 7% Fe and 2.3. C and are recommended for use in the production of building materials.

Clinker slag of zinc production is a waste product of processing oxidized zinc raw materials in rotating welz furnaces. When lead slags and oxidized zinc ores are formed, and in foreign practice, zinc slags of copper production, as well as presumably KIVCET slags, a clinker is formed that does not contain noble metals and other valuable components. According to VNIITSVETMET, the processing of clinker of this type can be reduced to: carbon extraction to return it to the Welz furnace as a fuel or reducing agent; extraction of metallic iron for use as a cementing agent or reducing agent for fusing furnaces and processing of the residue into building materials [14]. It is possible to melt clinker with fluxes due to the heat of combustion of carbon in cyclone or flare furnaces on a heated or oxygen-enriched blast, to produce foundry slag. At α = 1.15, the carbon burned out almost completely. With a carbon content of 25 – 27% in the non-magnetic fraction, the process is autogenic. Comparative data are given in Table [14].

Thus, the task of complex clinker processing can be solved, but with the need to take into account the specifics of the technology of a particular enterprise and the region of its location. When welding zinc cakes or raimovka, the processing of clinker is complicated by the need to extract precious metals and copper, the cost of which is several times higher than the cost of other components.

In this case, the clinker is processed in furnaces of copper smelters or lead plants without preparation or preliminary separation of carbon is carried out, which is used as fuel for welz furnaces.

An experiment was carried out at the Electrozinc plant in Russia to speed up the process and use less fuel. They tried blowing air to remove carbon and using a coal-air mixture in the furnace. This sped up the process (about 10%) and used less fuel, but also made the product less pure. Therefore, the experiment had to be stopped. The clinker is processed at the same plant by means of chloride distillation firing using residual coke as fuel.

Almalyk MMC is interested in processing clinker from baking cakes because it contains valuable non-ferrous and precious metals. [14]. It is a fine bulk material with a grain size of 15—20% of class +10 mm, 45—50% of class –10—5 mm, 8—10% of class –5 +3 mm, 40—45% of class –3 mm, containing on average, %: Cu — 0.5—5; Pb — 0.1—1; C (coke) — 15—25; Au — 0.5—6.5 g/t; Ag — 100—450 g/t.

The total production of copper-containing clinker of zinc production as of 1990—1992 in the CIS countries was about 450 thousand tons/year. In subsequent years, the clinker yield decreased by 15—20%, but at the beginning of this century this figure increased.

When processing clinker, it should be borne in mind that the fractional composition can be changed towards increasing the yield of large fractions when recycled clinker (small fractions) is introduced into the charge of welz furnaces as an additive. The process involves leaching with sulfuric acid at temperatures of 60—80 degrees. Au and Ag are then extracted from the cake using sorption technology and flotation to produce coke (C). The sorption process includes cyanidation, which is its disadvantage. Instead, it is proposed to use sodium bisulfite to extract Au and Ag. This technology makes it possible to obtain Cu powder with a purity of 90.1—94.8%, Dore alloy with 84—91% Au and 55—65% Ag, and coal concentrate with 95% C content.

During the extraction of non-ferrous metals from stale clinkers slags of zinc production of UCCC and CHPP in GINTSVETMET, a method of chloride distillation in a fluidized bed furnace was developed [15]. The method was tested on an enlarged semi-industrial plant with a capacity of 165 kg of raw materials per hour.

When processing clinkers slags of zinc production of UCCC containing 1.89% copper, 2.43% zinc, 0.87% lead, 0.25 kg/t silver and 00.5 kg/t gold, the degree of chloride sublimation of non-ferrous metals was: copper — 86.5%, zinc — 79%, lead — 93.2%, silver — 93.8%, gold — 88% at a temperature of 1223—1273 K. Despite the fact that KAVKAZGIPROTSVETMET has developed a technical project for the construction of a clinkers slags of zinc production processing plant at the Elektrozink plant, the method has not received practical application due to its disadvantages, such as high energy consumption and complexity of hardware design. The chloride distillation method has several disadvantages that prevent its widespread use. Firstly, it requires a large amount of concentrated calcium chloride solution, which is 30% of the ore weight. This leads to an increase in the cost of reagents and electricity. Secondly, the process takes a long time — about 5.5 hours. Thirdly, after processing, a significant amount of metals remains in the stub, especially zinc and copper, at the level of 0.5% and 0.25%, respectively. All these factors make chloride sublimation of non-ferrous metals less attractive compared to other metal extraction methods.

At CIS plants, clinker is processed by adding agglomerate of copper and lead concentrates to the charge, however, when adding 1.3—1.8% clinker to the charge, the agglomeration process is unsatisfactory. Direct processing of clinker in mine furnaces leads to an increase in dust removal (up to 10%) and the production of metallized matte, which complicates its release (CMC). Hot briquetting of clinker has been mastered at the BCZ, however, the resulting briquette is poorly processed in mine furnaces due to increased metallization of matte.

At AGMK and JGMK (Kazakhstan), work was carried out on processing clinker in converters. Positive results were obtained in reducing the copper content in converter slags and increasing the processing of revolutions [12, 14]. However, at the same time, the wear of the lining increased and the SO_2 content in the exhaust converter gases decreased. In addition, the removal of pulverized carbon-containing clinker fractions led to their combustion in flues.

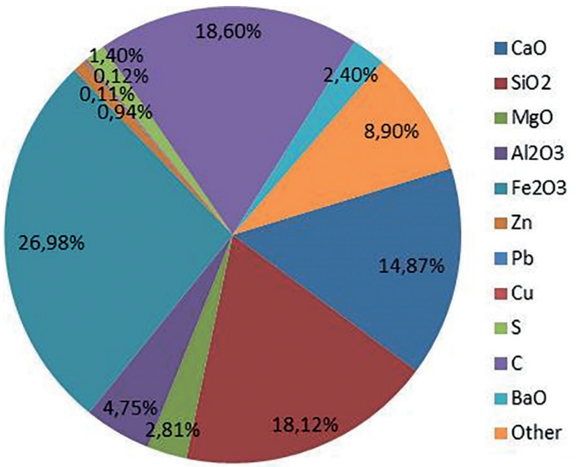
The processing of clinker in the PV and CFP furnaces at BGMK and AGMK [14, 16] gave a positive result, but this method is more acceptable for PV furnaces, since it reduces fuel consumption, since additional heat generation reduces furnace performance for CFP.

The Unipromed Institute, together with MMSK and KaMK, has developed a technology for processing clinker in mine furnaces. The development was preceded by a large amount of theoretical work [14, 17], during which it was established that the main task of smelting is the deep oxidation of carbon and iron in order to prevent the formation of crust.

The oxidation sequence of the charge components is as follows: coke — dissolved carbon — metallic iron — sulfides. The interaction of metallized matte with oxygen begins with the oxidation of the metal, in which sulfides are not oxidized. Therefore, varying the ratio of metal and sulfides can minimize desulfurization and produce exhaust gases with a content of 0.1 — 0.5% SO_2. Based on this, the technology of melting clinker briquettes with dust and recycled materials and melting clinker with matte recycling was introduced at KaMK and MMSK, which made it possible to increase the processing of clinker and obtain furnace gases with a content of 0.1 — 0.5% SO_2.

The technology of slag processing proposed by scientists from Shymkent University in the early 2000s was as follows, тhe developed technology made it possible to drive off lead-zinc sublimations (up to 98%) and isolate iron and silicon into a low-grade ferrosilicon alloy. However, this process is energy-intensive and led to the formation of a large amount of waste (more than 50%), which was proposed to be crushed into slag rubble. These studies have been tested on a pilot industrial base, but have not been further implemented in industry.

Despite the variety of different methods and techniques for processing, the dump of mining stale clinkers (welz slag) of zinc production has not been disposed of at present, and still has a negative impact on the environment of the region and the population, as evidenced by an interview with the head of the Department of Ecology for the Turkestan region K. Kalmakhan spoke about the environmental problems of the Turkestan region to the newspaper "South Kazakhstan" dated April 25, 2022, during which he, in particular, noted the following: "The main problems in our region are related, including with the placement of large-tonnage mining waste — these are dumps, tailings dumps. These wastes must be recycled and the territory must be cleared of them. Subsequently, the released lands can be given away for other needs. There is a stale clinker-slag dump in Ashisai, which is also located along the river, and there are dumps of phosphorite ores in the Suzak district. There are 44 dumps and tailings dumps in the region. For example,



Chemical composition of the studied stale clinker slag of zinc production
Химический состав исследуемого лежалого шлакового клинкера цинкового производства

in dumps and tailings dumps, according to scientists, there is a large volume of zinc and other non-ferrous metals that can be processed and extracted."

In this regard, we conducted slag sampling from the processing of non-ferrous metal ores using a manual sampling method in accordance with GOST 28192-89. 5 point samples were taken along the perimeter of the tailings dump and combined into one combined sample, which was then delivered to the laboratory for analysis. The samples were analyzed using physicochemical methods, including chemical and electron microscopic analyses. The stale clinker-slag under study is represented by the following chemical composition, which is shown in Figure.

We conducted a chemical analysis of toxic slag in order to dispose of it by complex processing as an iron-containing additive in the production of cement clinker.

The sample of the test was analyzed on an electronic scanning microscope in order to identify its chemical element structure and the content of various elements. According to the analytical results of electron scanning microscopy, it was found that the present slag in its structure has the following elements as Ca (calcium), Si (silicon), O (oxygen), Fe (iron), Al (aluminum), the presence of compounds demonstrated by the previously conducted chemical analysis results [18 – 21], and which make it a secondary raw material for the chemical industry [22 – 26].

Conclusions

Based on the review of Ashisai stale clinkers (slags) processing technologies, chemical and electron microscopic analyses, the following conclusions can be drawn:

- the Ashisai stale clinker dump contains such important substances as aluminum, iron, calcium, silicon and a small amount of heavy metals, clinker dumps can be used as raw materials for the production of various industrial products such as cement.
- there are various ways of processing stale clinker dumps, for example, the use of chloride, its melting and other methods.
- the way waste is handled today, in particular with the stale clinker dump, can lead to the consumption of a large amount of energy and the formation of even more waste. Therefore, it is necessary to look for ways and methods for their complex processing and disposal;

• despite the availability of technologies for processing stale clinker dumps, many of them have not been widely used due to various factors, including economic and environmental ones;

• for many years, the stale clinker slag dump in the village of Ashisai continues to have a polluting effect on the environment and the population under the influence of natural and climatic factors;

• it is necessary to conduct further research to optimize the processes of processing stale clinker waste and increase their environmental, socio-economic efficiency.

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ИНФОРМАЦИЯ ОБ АВТОРАХ

Гаппаров Жакен Унарбой угли[1] — докторант PhD,
e-mail: zhakengapparov.94@mail.ru,
ORCID ID: 0009-0000-0597-489C,
Сырлыбеккызы Самал[1] — д-р PhD, профессор,
e-mail: Samal.Syrlybekkyzy@yu.edu.kz,
ORCID ID: 0000-0002-0260-0611,
Филин Александр Эдуардович — д-р техн. наук,
профессор, НИТУ МИСИС, e-mail: aleks_filin@bk.ru,
ORCID ID: 0000-0002-3015-5965,
Колесников Александр Сергеевич[2] — канд. техн. наук,
профессор, e-mail: kas164@yandex.kz,
ORCID ID: 0000-0002-8060-6234,
Жатканбаев Ергазы Тлеубаевич[2] — канд. техн. наук,
доцент, e-mail: ergazy-1980@mail.ru,
ORCID ID: 0000-0001-6955-806X,
[1] Каспийский университет технологий и инжиниринга
им. Ш. Есенова, Актау, Республика Казахстан,
[2] Южно-Казахстанский университет им. М. Ауэзова,
Шымкент, Республика Казахстан.
Для контактов: Колесников А.С., e-mail: kas164@yandex.kz.

INFORMATION ABOUT THE AUTHORS

Jaken Gapparov[1], PhD Student,
e-mail: zhakengapparov.94@mail.ru,
ORCID ID: 0009-0000-0597-489C,
Samal Syrlybekkyzy[1], PhD, Professor,
e-mail: Samal.Syrlybekkyzy@yu.edu.kz,
ORCID ID: 0000-0002-0260-0611,
Alexander Filin, Dr. Sci. (Eng.), Professor,
NUST MISIS, 119049, Moscow, Russia,
e-mail: aleks_filin@bk.ru,
ORCID ID: 0000-0002-3015-5965,
Alexandr Kolesnikov[2], Cand. Sci. (Eng.),
Professor, e-mail: kas164@yandex.kz,
ORCID ID: 0000-0002-8060-6234,
Yergazy Zhatkanbayev[2], Cand. Sci. (Eng.),
Assistant Professor, e-mail: ergazy-1980@mail.ru,
ORCID ID: 0000-0001-6955-806X,
[1] Sh. Yesenov Caspian University of Technology
and Engineering, 130002, Aktau, Kazakhstan,
[2] M. Auezov South Kazakhstan University,
160012, Shymkent, Kazakhstan.
Corresponding author: Alexandr Kolesnikov, e-mail: kas164@yandex.kz.

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