ГИАБ. Горный информационно-аналитический бюллетень /
MIAB. Mining Informational and Analytical Bulletin, 2022;(10-1):103–115
ОРИГИНАЛЬНАЯ СТАТЬЯ / ORIGINAL PAPER

УДК 622 DOI: 10.25018/0236_1493_2022_101_0_103

ОПТИМИЗАЦИЯ СЫРЬЕВОЙ СМЕСИ С ИСПОЛЬЗОВАНИЕМ ТЕХНОГЕННЫХ ОТХОДОВ ДЛЯ ПРОИЗВОДСТВА ЦЕМЕНТНОГО КЛИНКЕРА

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Аннотация: представлены результаты возможности использования техногенного сырья путем переработки его как вторичного минерального сырья, в частности, техногенных хвостов Надеждинского металлургического комбината, шлаков медного производства горно-металлургического комбината «Норильский никель» путем переработки их как вторичного минерального сырья с целью снижения их антропогенное воздействие на окружающую среду арктического региона. В частности, были проведены исследования по оптимизации состава сырьевой смеси и химико-минералогического состава цементного клинкера. Оптимизация проводилась с помощью программного комплекса «ROCS», предназначенного для расчета и оптимизации цементных сырьевых смесей в зависимости от коэффициента насыщения, с определением покомпонентного химического состава сырьевой смеси и цементного клинкера, минералогического состава цементного клинкера, теплового эффекта образования клинкера (FEC) и расхода топлива на обжиг (Gfuel). В ходе проведенных исследований установлено, что: техногенное сырье в виде отвальных хвостов и шлаков производства меди может быть использовано в качестве вторичного минерального сырья для производства цементного клинкера; в зависимости от коэффициента насыщения возможно получение цементного клинкера определенного минералогического состава, как низкоосновного (белит), так и высокощелочного (алит); в зависимости от минералогического состава клинкера оптимальный состав сырьевых смесей для белитового (низкоосновного) цементного клинкера-известняка – 72,97%; отвальные хвосты – 0,71%; шлак – 26,32%, а для алита (высокопрочный) – известняк – 81,71%; отвальные хвосты – 3,52%; шлак – 14,77%.

Ключевые слова: цементный клинкер, техногенные отходы, оптимизация.

Для цитирования: *Колесникова О., Васильева Н., Колесников А., Золкин А.* Оптимизация сырьевой смеси с использованием техногенных отходов для производства цементного клинкера // Горный информационно-аналитический бюллетень. – 2022. – № 10-1. – С. 103–115. DOI: 10.25018/0236_1493_2022_101_0_103.

Optimization of raw mix using technogenic waste to produce cement clinker

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Abstract: This article presents the results of the possible utilization of technogenic raw materials, in particular, technogenic tailings of the Nadezhdinsky Metallurgical Plant, slags of copper production of the mining and metallurgical plant Norilsk Nickel by processing them as secondary mineral raw materials in order to reduce their anthropogenic impact on the environment of the Arctic region. In particular, studies were conducted to optimize the composition of the raw material mix and chemical and mineralogical composition of cement clinker. optimization was carried out using the software package "ROCS", designed for the calculation and optimization of cement raw mixes depending on the saturation coefficient, with determining the component chemical composition of the raw mix and cement clinker, mineralogical composition of cement clinker, the thermal effect of clinker formation (FEC) and fuel consumption for firing (Gfuel). In the course of the research it was found that: technogenic raw materials in the form of tailings and slags of copper production can be used as secondary mineral raw materials for cement clinker production; depending on the saturation coefficient, it is possible to obtain cement clinker of a certain mineralogical composition, both low — base (belite) and high-base (alite); depending on the mineralogical composition of clinker, optimal compositions of raw mixtures for belite (low-base) cement clinker-limestone are as follows: 72.97%; dump tails-0.71%; slag-26.32%, and for alite (high — base) — limestone-81.71%; dump tailings-3.52%; slag-14.77%.

Key words: cement clinker, industrial waste, optimization.

For citation: Kolesnikova O., Vasilyeva N., Kolesnikov A., Zolkin A. Optimization of raw mix using technogenic waste to produce cement clinker. *MIAB. Mining Inf. Anal. Bull.* 2022;(10-1): 103—115. [In Russ]. DOI: 10.25018/0236_1493_2022_101_0_103.

Introduction

The Russian Arctic zone today is one of the priority areas of development in economic and environmental terms. Over the years of industrial development of the Arctic zone of Russia, a significant number of industrial enterprises of chemical oil, gas, mining and processing, mining and metallurgical, and a number of other industries have been put into operation. [1—16].

Today, the Arctic produces products that provide about 11% of Russia's national income (with a population of 1% living there) and account for up to 22% of all-Russian exports. The region has a multidisciplinary industrial and social infrastructure, which consists mainly of the raw materials sector of the economy, as well as military-industrial and transport (the Northern Sea Route — NSR) complexes [1—4, 17].

Most types of specialized products of the North have no alternative in terms of possible production in other regions of the country or import purchases. In fact, no branch of the Russian economy and social sphere can function without fuel, energy and other resources extracted and produced in the northern regions. At the same time, the development of a significant number of fields in the Arctic zone creates many problems and requires significant economic investments. In addition, new mining and transportation technologies are needed to ensure the preservation of the Arctic environment [2—4, 17].

In the Russian part of the Arctic zone, 27 sites have been identified (11 on land, 16 in the seas and the coastal zone), which have received the name "impact sites" and which represent the four main centers of anthropogenic tension — the Murmansk region (with a total emission of pollutants — 10%), the Norilsk industrial area (just over 30%), areas of development and operation of oil and gas fields in Western Siberia (just over 30%), and the Arkhangelsk region, where a high degree of pollution by specific substances is observed) [2—4, 18].

In these regions, the anthropogenic impact on the environment has already led to serious violations of the natural geochemical background, significant pollution of air, soil, subsoil, vegetation, penetration of harmful substances into the food chain and growth of morbidity of the living population [19—22, 57].

An extremely acute problem for the Arctic is the disposal of accumulated industrial waste, which has accumulated in huge quantities and continues to accumulate around industrial enterprises in the form of dumps, tailings ponds, sludge reservoirs, etc. [2—11, 23—30].

Today, the accumulated industrial waste is a constant source of environmental threat objects of pollution in the Arctic territories. One of the most dangerous are wastes and abandoned areas of mining and industrial production, especially those associated with the extraction of raw materials for non-ferrous metallurgy and with the use of amalgamation method in the technological cycle of their production [1—4, 23—36].

The Arctic, due to its natural and climatic conditions at the present stage of its development, is not able to recycle huge amounts of accumulated waste from various industries and enterprises even over hundreds of years [1—4, 23—38]. In this regard, today the problem of processing [39] and disposal [40] of daily generated industrial and accumulatedindustrial waste, which is the main problem of pollution of the natural environment of the region is acute.

A similar technogenic source of pollution of the Arctic environment is the accumulated dump tailings of the Nadezhdinskiy Metallurgical Plant (NMZ) and slags from copper production at the Norilsk Nickel Mining and Metallurgical Plant. They have accumulated in huge quantities, amounting to tens of millions tons and occupy large areas of land for waste dumps and tailings, while polluting the environment, having negative impact on the air, soil, surface and underground water, flora and fauna, and, of course, negatively affecting the lives and health of people living there. Currently functioning industrial enterprises and production facilities of Norilsk mining and metallurgical Plant are foci of anthropogenic pollution of the natural environment of the city of Norilsk and the Krasnoyarsk Krai.

Having considered [41—45] and studied the chemical composition of the dump tailings of the Nadezhdinsky Metallurgical Plant, which are a fine powder, wefound that they contain compounds of oxides of silicon, aluminum, calcium, iron, magnesium, the presence of which makes it possible to use them as secondary mineral raw materials in the chemical industry, as an iron-containing component in the raw material charge for the production of cement clinker. Similar to the waste tailings of the NMZ, the slag of the copper production of the Norilsk Nickel mining and Metallurgical combine also contains in its chemical composition such useful compounds as compounds of silicon, aluminum, iron, calcium, and magnesium oxides [46—50] and is able to act as a silicon-containing component of the raw charge for the production of cement clinker. Limestone of the Kalargonskoye deposit with CaO content from 46 to 52% is considered as the main component of the raw charge containing $CaCO_3$ [58—60].

In the standard technological scheme for the production of cement clinker, currently is used a three-component charge mixture of limestone, loess clay and iron ore. In this case, limestone, clay and iron ore must be extracted in quarries which must be developed and maintained, and the raw materials extracted from them must undergo a multi-stage crushing and grinding to prepare the component of the raw material mixfor clinker, which requires a huge financial investment, which in the future will affect the high cost of obtaining cement clinker [61 – 63].

In this regard, the use of waste, which by its chemical composition can replace the two classical components (clay and iron ore), as well as save financial resources intended for the development of clay and iron ore quarries and their preparation in the form of crushing, grinding and grinding, which is beneficial in terms of finance — the cost cement clinker will be significantly reduced, while at the same time recycling waste from the mining and metallurgical industry, and, accordingly, reducing the anthropogenic load on the environment. Therefore, at present, research aimed at reducing the anthropogenic load on the environment, reducing the cost of mining and preparing mineral raw materials, and recycling industrial waste by involving it in the production cycle as secondary mineral raw materials is relevant.

The purpose of the research was to calculate and optimize the raw mix with the involvement of technogenic waste as components of the raw mix for the production of cement clinker, using the software package "ROCS" [51], designed to calculate and optimize multi-component raw mixes of mineralogical composition of Portland cement clinker for cement production.

Materials and Methods

The "ROCS" program, in comparison with all existing in our country and abroad programs and methods [52 – 56], has a number of new features and allows [51]:

– calculate mixes with any number of components

– take into account introduction of an unlimited number of additives (components with specified consumption) into the raw mix or directly into the furnace);

– calculate special cements and perform calculations using various methods (e.g., those used in the UK or the USA);

– optimize the composition of the raw mix and clinker in terms of various characteristics, including the energy intensity of the resulting mixes

– make recommendations for the preparation of mixes based on the raw material base of a particular plant

– perform a graphical analysis of the characteristics of mixtures and clinker, including depending on the consumption of additives

– expand and customize the program to account for the raw material base of the plant, the products produced, the calculation of new types of clinkers and the application of new calculation methods.

Implementation of these differences became possible as a result of the fact that the program is based on fundamental scientific work — methods of calculation and optimization of multicomponent silicate-containing systems and raw mixes [28].

In the course of research by optimization method, a three-component mixture consisting of limestone of the Kalargonskoye deposit, dump tailings and slag of copper production was developed with the following chemical composition, given in Table 1.

Results and Discussion

Using the software package ROCS, in order to optimize the raw mix and mineral composition of clinker, we conducted a series of calculations with a different saturation coefficient (KN) in the range of 0.70 – 0.90 with a step of 0.02, with a constant silicate modulus (n), which was taken in all calculations equal to 1.8.

Table 1
Chemical composition of raw materials intended for optimization in the production of cement clinker

Compounds	Components, %		
	Limestone	Dump tails	Slag
SiO_2	6.67	15.28	54.75
Al_2O_3	2.73	4.46	13.45
Fe_2O_3	0.38	49.73	9.38
CaO	50.12	10.25	18.3
MgO	0.21	0.85	1.2
SO_3	0.1	1.58	0.25
Na_2O	0.52	0.23	–
K_2O	0.38	0.12	–
Losses	38.63	11.46	–
Other	0.76	6.39	2.67

At the same time, the alumina modulus (p) during optimization, depending on KN, varied from 1.154 to 1.,270. Of all the calculations carried out, we took and presented three calculations, in particular, for KN equal to 0.70 (Figure 1), for KN equal to 0.80 (Figure 2) and for KN=0.90 (Figure 3). Optimization was carried out according to the following parameters: mineralogical composition of clinker, in particular, the phase content of the main minerals in cement clinker; fuel consumption (G_{fuel}); thermal effect of clinker formation (FEC); composition of raw materials in the raw mixture.

From the results of optimization of composition of raw mix and mineralogical composition of cement clinker with saturation coefficient (KN) of 0.70, shown in Figure 1, it can be seen that, depending on the KN, with a silicate module of 1.8, it is possible to obtain cement clinker of the following mineral composition: C_3S (alite) – 8.90%; C_2S (belite) – 60.83%; C_3A (tricalcium aluminate) – 8.30%; C_4AF (four – calcium aluminoferrite) – 18.49%; $CaSO_4$ (gypsum)-0.47%; MgO (magnesium oxide) – 0.62%.

The predominance of the belite phase in the mineralogical composition means that the resulting cement clinker will be belite, which meets the requirements for cement clinkers according to GOST 31108–2016. The raw mix for the production of belite cement clinker is represented by the following percentage of the components of the raw material mixture, in particular: limestone-72.97%; dump tailings-0.71%; slag-26.32%.

At formation of the fuel and energy complex (thermal effect of clinker formation) equal to 289 kcal/kg, the consumption of the standard fuel equivalent for burning (G_{fuel}) is equal to 179 kg of conventional fuel/t of cl.

Fig. 2, which presents the results of the optimization of the composition of the raw mix and mineralogical composition of cement clinker at KN=0.80 and n=1.8. shows that the formation of cement clinker of the following mineral composition occurs: C3S – 33.43%; C2S – 37.94%; C3A- 8.39%; C4AF – 16.85%; CaSO4 – 0.45%; MgO – 0.58%. from which it follows that this cement clinker by its mineralogical composition does not meet the requirements of GOST 31108–2016. In this case, the thermal effect of clinker formation was 320.8 kcal/kg, and the consumption of conventional fuel equivalent for burning (G_{fuel}) was

Chemical composition of raw materials										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Limestone	6,67	2,73	0,38	50,12	0,21	0,10	0,52	0,38	38,63	0,26
Dump tails	15,28	4,46	49,73	10,25	0,85	1,58	0,23	0,12	11,46	6,04
Slag	24,75	13,45	9,38	18,30	1,20	0,25	-	-	-	2,67

Component chemical composition of the raw mixture										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Limestone	5,069	2,075	0,289	38,089	0,160	0,076	0,395	0,289	29,357	0,198
Dump tails	0,652	0,190	2,122	0,437	0,036	0,067	0,010	0,005	0,489	0,258
Slag	10,807	2,655	1,851	3,612	0,237	0,049	-	-	-	0,527
Raw mixture	16,53	4,92	4,26	42,14	0,43	0,19	0,40	0,29	29,85	0,98

Component chemical composition of clinker									
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Other
Limestone	7,225	2,957	0.412	54,293	0,227	0,108	0,563	0,412	0,282
Dump tails	0,929	0,271	3.025	0,623	0,052	0,096	0,014	0,007	0,367
Slag	15,404	3,784	2.639	5,149	0,338	0,070	-	-	0,751
Clinker	23,56	7,01	6,08	60,06	0,62	0,27	0,58	0,42	1,40

Chemical composition of the raw mixture and clinker										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Raw mixture	16,53	4,92	4,26	42,14	0,43	0,19	0,40	0,29	29,85	0,98
Clinker	23,56	7,01	6,08	60,06	0,62	0,27	0,58	0,42	-	1,40

Modules	Raw mixture	Clinker
KN (lime saturation coefficient)	0,7	0,7
n (silica module)	1,8	1,8
p (alumina module)	1,154	1,154
FEC (thermal effect of clinker formation, kcal/kg)	-	289,2
G_{fuel} (fuel consumption for firing, kg of conventional fuel/t of cl.)	-	179

Mineralogical composition						
Minerals	C_2S	C_3S	C_3A	C_4AF	$CaSO_4$	MgO
Mas.%	8,90	60,83	8,30	18,49	0,47	0,62

Content of components			
Materials	Raw mixture		Clinker
	kg/kg cl	%	%
Limestone	1,1141	72,97%	64,22%
Dump tails	0,0108	0,71%	1,04%
Slag	0,4018	26,32%	34,74%
Amount	1,5267	100,00%	100,00%

Fig. 1. Results of the calculation of the chemical and mineralogical composition of the raw mixture and clinker at KN=0.70 and n=1.80

186.1 kg of fuel equivalent/t of clinker. The components of the raw mixture are presented in the following percentage ratio: limestone-79.08%; dump tailings-3.86%; slag-17.06%.

From the results of the optimization and calculation of the chemical and mineralogical composition of the raw mix and clinker at KN = 0.90 and n = 1.80, shown in Figure 3, it can be seen that the mineralogical composition of cement clinker is represented by the main minerals in the following percentage ratio: C_3S-54.98%; C_2S-17.84%; C_3A-

Chemical composition of raw materials										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Limestone	6,67	2,73	0,38	50,12	0,21	0,10	0,52	0,38	38,63	0,26
Dump tails	15,28	4,46	49,73	10,25	0,85	1,58	0,23	0,13	11,46	6,04
Slag	54,75	11,45	9,38	18,30	1,20	0,25	-	-	-	2,67

Component chemical composition of the raw mixture										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Limestone	5,275	2,159	0,301	39,635	0,166	0,079	0,411	0,301	30,549	0,206
Dump tails	0,590	0,172	1,921	0,396	0,033	0,061	0,009	0,005	0,443	0,233
Slag	9,339	2,294	1,600	3,121	0,205	0,043	-	-	-	0,455
Raw mixture	15,20	4,63	3,82	43,15	0,40	0,18	0,42	0,31	30,99	0,89

Component chemical composition of clinker									
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Other
Limestone	7,644	3,128	0,415	57,436	0,241	0,115	0,596	0,435	0,298
Dump tails	0,855	0,250	2,783	0,574	0,048	0,088	0,013	0,007	0,338
Slag	13,533	3,324	2,318	4,523	0,297	0,062	-	-	0,660
Clinker	22,03	6,70	5,54	62,53	0,58	0,26	0,61	0,44	1,30

Chemical composition of the raw mixture and clinker										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Raw mixture	15,20	4,63	3,82	43,15	0,40	0,18	0,42	0,31	30,99	0,89
Clinker	22,03	6,70	5,54	62,53	0,58	0,26	0,61	0,44	-	1,30

Modules	Raw mixture	Clinker
KN (lime saturation coefficient)	0,8	0,8
n (silica module)	1,8	1,8
p (alumina module)	1,21	1,21
FEC (thermal effect of clinker formation, kcal/kg)	-	320,8
G_{fuel} (fuel consumption for firing, kg of conventional fuel/t of cl.)	-	186,1

Mineralogical composition						
Minerals	C_3S	C_2S	C_3A	C_4AF	$CaSO_4$	MgO
Mac.%	33,43	37,94	8,19	16,83	0,45	0,58

Content of components			
Materials	Raw mixture		Clinker
	kg/kg cl	%	%
Limestone	1,146	79,08%	70,33%
Dump tails	0,056	3,86%	4,96%
Slag	0,2472	17,06%	24,72%
Amount	1,4491	100,00%	100,00%

Fig. 2. Results of the calculation of the chemical and mineralogical composition of the raw mixture and clinker at KN=0.80 and n=1.80

8.47%; C_4AF-15.41%; $CaSO_4$ – 0.44%; MgO – 0.56%. based on which it follows that the resulting clinker is alitic due to the predominance of the mineral C_3S and highly basic in accordance with GOST 31108 – 2016, with a thermal effect of clinker formation – 348.5 kcal/kg and the consumption of fuel equivalent for burning (G_{fuel}) – 192.4 kg of fuel equivalent/t of clinker. The components

Chemical composition of raw materials										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Limestone	6,67	2,73	0,38	50,12	0,21	0,10	0,52	0,38	38,63	0,26
Dump tails	15,28	4,46	49,73	10,25	0,85	1,58	0,23	0,12	11,46	6,04
Slag	54,75	13,45	9,38	18,30	1,20	0,25	-	-	-	2,67

Component chemical composition of the raw mixture										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Limestone	5,450	2,231	0,310	40,953	0,172	0,082	0,425	0,310	31,564	0,212
Dump tails	0,537	0,157	1,749	0,361	0,030	0,056	0,008	0,004	0,403	0,212
Slag	8,088	1,987	1,386	2,704	0,177	0,037	-	-	-	0,394
Raw mixture	14,08	4,37	3,45	44,02	0,38	0,17	0,43	0,31	31,97	0,82

Component chemical composition of clinker									
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Other
Limestone	8,011	3,279	0,456	60,195	0,252	0,120	0,625	0,456	0,312
Dump tails	0,790	0,231	2,571	0,530	0,044	0,082	0,012	0,006	0,312
Slag	11,889	2,921	2,037	3,974	0,261	0,054	-	-	0,580
Clinker	20,69	6,43	5,06	64,70	0,56	0,26	0,64	0,46	1,20

Chemical composition of the raw mixture and clinker										
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Losses	Other
Raw mixture	14,08	4,37	3,45	44,02	0,38	0,17	0,43	0,31	31,97	0,82
Clinker	20,69	6,43	5,06	64,70	0,56	0,26	0,64	0,46	-	1,20

Modules	Raw mixture	Clinker
KN (lime saturation coefficient)	0,9	0,9
n (silica module)	1,8	1,8
p (alumina module)	1,27	1,27
FEC (thermal effect of clinker formation, kcal/kg)	-	348,5
C[illegible] (fuel consumption for firing, kg of conventional fuel/t of cl.)	-	192,4

Mineralogical composition						
Minerals	C_3S	C_2S	C_3A	C_4AF	$CaSO_4$	MgO
Mas.%	54,98	17,84	8,47	15,41	0,44	0,56

Content of components			
Materials	Raw mixture		Clinker
	kg/kg cl	%	%
Limestone	1,2010	81,71%	73,71%
Dump tails	0,0517	3,52%	4,58%
Slag	0,2171	14,77%	21,71%
Amount	1,4699	100,00%	100,00%

Fig. 3. Results of the calculation of the chemical and mineralogical composition of the raw mixture and clinker at KN=0.90 and n=1.80

of the raw mixture are presented in the following percentage: limestone- 81.71%; dump tailings-3.52%; slag-14.77%.,

Results and Discussion Conclusions

Thus, based on the research we can draw the following conclusions:

– Technogenic raw materials in the form of waste tailings and slags of copper production, can be used as secondary mineral raw materials in the production of cement clinker.

– Depending on the saturation coefficient, it is possible to produce cement clinker of a certain mineralogical composition, such as belite, with a fuel consumption of 179 kg of equivalent fuel/t of cl. and C2S content of 60.83%, and alite, with C3S content of 54.98% and a fuel consumption of 192.4 kg of equivalent fuel/t of clinker.

– In the course of optimization, depending on the mineralogical composition of cement clinker, optimal compositions of raw mixes for the belite-containing cement clinker were found in the following ratio: limestone-72.97%; dump tails-0.71%; slag-26.32%. and for the alite — containing limestone-81.71%; dump tails-3.52%; slag-14.77%.

– Organizing the processing of technogenic dump tailings of the Nadezhdinsky Metallurgical Plant, copper slag of the Norilsk Nickel Mining and Metallurgical Plant, and limestone to produce cement clinker using the above technology in the Arctic region, will improve the social, investment, economic and environmental climate, which in its turn will contribute to the development of the Arctic.

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Получена редакцией 20.03.2022; получена после рецензии 27.06.2022; принята к печати 10.09.2022.
Received by the editors 20.03.2022; received after the review 27.06.2022; accepted for printing 10.09.2022.