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Influence of industrial waste on the structure of environmentally friendly cement clinker

[1,2]Zhanikulov N.N., [3*]Kolesnikov A.S., [3]Taimasov B.T., [4]Zhakipbayev B.Y., [3]Shal A.L.

[1]Academician E.A. Buketov Karaganda University, Karaganda, Kazakhstan
[2] M.V. Lomonosov Moscow State University, Moscow, Russian Federation
[3]M. Auezov South Kazakhstan University, Shymkent, Kazakhstan
[4]Bolashak University, Kyzylorda, Kazakhstan

*Corresponding author`s e-mail: kas164@yandex.kz

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ABSTRACT

The main problem of cement production is to reduce energy consumption and reduce the amount of greenhouse gases CO_2 emitted into the atmosphere. The prerequisite for solving the problem is: the use of man-made waste and unconventional raw materials in the composition of the charge and the reduction of limestone content from 85% to 70-75% as a source of CO2 gas. The article presents the results of the studied technogenic waste and the suitability of non-traditional raw materials such as tefritobazalt, coal mining waste, and lead slag. The chemical and mineralogical composition of the materials and their suitability for obtaining environmentally friendly clinker have been established. The possibility of obtaining cement clinker using low-energy resource-saving technologies is shown. Clinker formation processes in the developed mixtures are completed at a temperature of 1350 °C, which, with the coal contained in the waste, will reduce the consumption of fuel injectors and reduce CO_2 emissions into the atmosphere. In the developed low-energy mixtures, the specific consumption of raw materials for obtaining 1 ton of clinker is reduced to 1516-1525 kg, which is significantly lower than in traditional raw mixtures. Alite crystals are large, reaching 100-140 microns. The content of clinker minerals is C_3S-57.88%, C_2S-18.82%, C_3A-6.46% and C_4AF-11.61%. Disposal of large-tonnage waste will reduce environmental pollution and improve the environmental situation in the region.

Keywords: clinker, microstructure improvement, waste disposal, temperature reduction, mineralizer.

Information about authors:

Zhanikulov Nurgali Nodiruly	*Doctor of Ph.D., assistant professor, Karagandy university of the name of academician E.A. Buketov, Universitetskaya street, 28, 100028, Karaganda, Kazakhstan; e-mail: nurgali.zhanikulov@mail.ru, ОРСИД: 0000-0002-0750-9753; Scholarship holder of the Bolashak program of the M.V. Lomonosov Moscow State University, Moscow, Russian Federation*
Kolesnikov Alexandr Sergeevich	*Professor, Candidate of Technical Sciences, M. Auezov South Kazakhstan university, Avenue Tauke-Khan, 5, 160012, Shymkent, Kazakhstan, e-mail: kas164@yandex.ru, ОРСИД: 0000-0002-8060-6234*
Taimasov Bahitzhan Tamasovich	*Professor, Doctor of technical sciences, M. Auezov South Kazakhstan university, Avenue Tauke-Khan, 5, 160012, Shymkent, Kazakhstan, e-mail: taimasovukgu@mail.ru, ОРСИД: 0000-0002-1844-4932*
Zhakipbayev Bibol Yermuratovich	*associate professor, Doctor of PhD, Bolashak University, Avenue Abay, 31A, 120000, Kyzylorda, Kazakhstan, e-mail: bibol_8484@mail.ru, ОРСИД: 0000-0002-1412-7796*
Shal Aizhan Leskyzy	*master student, M. Auezov South Kazakhstan university, Avenue Tauke-Khan, 5, 160012, Shymkent, Kazakhstan, e-mail: shalaizhan@mail.ru, ОРСИД: 0000-0002-0291-6894*

Introduction

Much attention is paid to the production of building materials, including Portland cement, in the Republic of Kazakhstan. The production of cement and a number of other building materials is constantly growing. According to the statistical agency of the Republic of Kazakhstan, in 2021 cement production amounted to more than 12.65 million tons, which is 15% more than in 2020 [[1]. [2], [3]].

Exports of cement to neighboring countries increased. The production of mixed binders and dry building mixtures for finishing works is also increasing. At the same time, we must remember that large enterprises in metallurgy, chemical industry, thermal power plants, cement production, etc. cause significant harm to the environment, ecology, and health of the population [[4], [5]].

The Concept for the Transition of the Republic of Kazakhstan to a Green Economy, approved by the Decree of the President of the Republic of Kazakhstan dated May 30, 2013, No.577 [6], lays the foundations for deep systemic transformations in order to transition to a new economy by increasing the welfare, quality of life of the population of Kazakhstan and countries among the 30 most developed countries in the world while minimizing the burden on the environment and degradation of natural resources. The concept of a green economy sets ambitious goals to achieve the level of pollutant emissions at the level of the participating countries of the Organization for Economic Cooperation and Development and other developed countries [[7], [8]].

Cement production refers to facilities that have a significant negative impact on the environment and are required to obtain integrated environmental permits to carry out their activities in accordance with the environmental legislation of the Republic of Kazakhstan [[9], [10]].

Portland cement is a large-tonnage and energy-intensive product.

The production of 1 ton of cement requires about 5 tons of various materials: raw materials, additives, fuel, water, air, refractories, grinding bodies, etc. In the Republic of Kazakhstan, 3 plants are still operating in a wet way. Specific fuel consumption for the wet method per 1 ton of clinker is 220-240 kg, for the dry one - 100-120 kg. Therefore, the issues of energy saving, especially with the wet method of obtaining clinker, remain important and relevant [[11], [12], [13]].

To increase the energy efficiency of the clinker burning process, and reduce fuel consumption, we have developed low-energy-intensive compositions of raw mixtures, where traditional natural raw materials are completely or partially replaced by technogenic ones [[14], [15]].

In this study, we propose to partially replace the traditional carbonate and aluminosilicate components of the raw charge (loess) with waste from coal mines in Lenger, magmatic rock – tephrite basalt of the Daubaba deposit in South Kazakhstan, and lead slags.

Experimental

Limestone is mainly composed of $CaCO_3$. The content of oxides SiO_2, and Al_2O_3 is low, and the content of Fe_2O_3 is very low - 0.57%. The MgO content is insignificant - 0.88%. The limestone is pure and highly basic, and the CaO content is more than 52%. The alkali content is within the normal range.

Coal mining wastes are composed of clay, carbonate minerals, and carbon. The content of silicon oxide is more than 55%, Al_2O_3 -10.6 %, they can replace the aluminosilicate component of the raw mixture in the raw mixture. The carbon content in coal mining waste is more than 15%.

Tephrite basalt contains SiO_2 - 45.54 %, Al_2O_3 - more than 10 %, Fe_2O_3 - about 8.5%. The magnesium content is increased - 6.95 %, which is undesirable. It contains a significant amount of alkalis: (K_2O + Na_2O) is 2.54%, which is also a lot.

Lead slags consist mainly of fayalite Fe_2SiO_4, melilite, wustite, small amounts of iron, lead and copper sulfides, and zinc spinel. Lead slag contains up to 37-40% iron oxides and can replace a corrective additive. In addition, lead slags contain up to 15% CaO and partially replace the carbonate component [[16], [17]].

For the preparation of the raw batch for low-energy clinkers, we used limestone of the Sastobe deposit, tephrite basalt, technogenic raw materials - lead slags, and coal mining waste, the chemical composition of which is given in Table 1.

Table1 - Chemical composition of mineral and technogenic raw materials

Natural and technogenic raw materials	Chemical composition, wt%							
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	loss on ignition	Sum
Limestone	3.87	1.04	0.57	52.83	0.88	0.10	40.71	100.0
Cinders	17.81	4.25	62.23	4.21	3.71	-	7.79	100.0
Lead slag	25.94	6.44	37.25	14.71	6.15	0.04	0.1	90.63
Tephrite basalt	45.54	10.7	8.53	10.66	6.95	0.2	7.92	90.50
Coal mining waste	55.50	10.6	2.01	3.21	0.7	0.79	24.08	96.89

Table 2 - Composition of low-energy raw mixes and clinkers based on technogenic and non-traditional raw materials

Mixes	Composition of the raw mixture, wt%					Specific consumption of raw materials, t/t of clinker					SC	Modules		Mineralogical composition of clinker, wt%			
	Lime stone	coal mining waste	tephrite basalt	cinders	lead slag	Lime stone	coal mining waste	tephrite basalt	cinders	lead slag		n	p	C_3S	C_2S	C_3A	C_4AF
1	75.76	11.69	11.68	0.86	-	1.158	0.179	0.178	0.013	-	0.85	2.7	1.50	48.85	30.29	6.44	11.55
2	76.67	10.92	10.91	1.50	-	1.174	0.167	0.167	0.023	-	0.90	2.5	1.26	58.61	19.05	3.11	16.38
3	76.94	11.13	11.12	0.81	-	1.182	0.171	0.171	0.012	-	0.90	2.7	1.51	59.62	19.38	5.19	13.0
4	76.64	10.34	10.33	2.69	-	1.171	0.158	0.158	0.041	-	0.92	2.2	0.97	60.90	19.81	7.76	8.72
5	77.12	10.21	10.2	1.47	-	1.183	0.164	0.165	0.023	-	0.92	2.5	1.26	61.99	14.86	1.47	18.88
6	77.39	10.92	10.91	0.79	-	1.191	0.336	0.168	0.168	-	0.92	2.7	1.51	62.85	15.07	3.1	16.17
7	76.22	19.61	-	-	4.17	1.186	0.305	-	-	0.065	0.85	2.7	1.31	48.43	30.03	6.65	11.28
8	76.93	18.03	-	-	5.05	1.196	0.280	-	-	0.078	0.90	2.5	1.13	59.00	19.19	5.41	12.67
9	77.38	18.67	-	-	3.95	1.209	0.292	-	-	0.062	0.90	2.7	1.32	59.69	19.41	6.49	10.88
10	76.56	16.54	-	-	6.90	1.181	0.255	-	-	0.106	0.92	2.2	0.89	61.97	14.80	3.48	15.65
11	77.37	17.68	-	-	4.94	1.204	0.275	-	-	0.077	0.92	2.5	1.13	63.27	15.18	5.38	12.49
12	77.82	18.32	-	-	3.87	1.218	0.287	-	-	0.061	0.92	2.7	1.33	64.00	15.35	6.43	10.7
13	74.34	10.36	10.37		4.93	1.119	0.156	0.156		0.074	0.85	2.2	0.98	45.97	28.49	4.58	15.35
14	75.56	11.47	11.48	-	1.49	1.152	0.175	0.175	-	0.023	0.85	2.7	1.51	47.43	29.41	7.73	10.18
15	76.32	10.55	10.55	-	2.57	1.164	0.161	0.161	-	0.039	0.90	2.5	1.27	57.88	18.82	6.46	11.61
16	76.75	10.93	10.93	-	1.39	1.176	0.167	0.168	-	0.021	0.90	2.7	1.52	58.50	19.03	7.57	9.82
17	76.01	9.68	9.68	-	4.63	1.152	0.147	0.147	-	0.070	0.92	2.2	0.99	60.89	14.60	4.46	14.65
18	76.78	10.35	10.35	-	2.52	1.173	0.158	0.158	-	0.039	0.92	2.5	1.28	62.08	14.89	6.41	11.46
19	77.20	10.72	10.72	-	1.36	1.185	0.164	0.165	-	0.021	0.92	2.7	1.52	62.75	15.06	7.47	9.70

Results and Discussion

We have developed various options for low-power compositions of raw materials, designed for the production of clinkers of various mineralogical composition, including sulfate-resistant and road Portland cements using limestone from the LLP Standard Cement (Table 2).

Calculations of charge compositions were made according to the formulas of S.D. Okorokov [18]. Several series of calculations of raw materials charges with natural limestone, unconventional and technogenic raw materials, located in close proximity (5-40 km) to the existing cement plants in South Kazakhstan, have been performed. Delivery of waste to the plant can be carried out by road and rail.

Based on the calculations, the compositions of the three- and four-component charges were selected:

- Limestone + coal mining waste + tephrite basalt + cinders;
- Limestone + coal mining waste + lead slag;
- Limestone + coal mining waste + tephrite basalt + lead slag.

When selecting the compositions of the raw mixtures, the saturation coefficient (SC) was changed from 0.85 to 0.92, and the silicate modulus (n) from 2.2 to 2.7. In this case, the value of the alumina modulus (p) varies depending on the type of components, silicate modulus, and SC from 0.97 to 1.52 [19].

Regularities of changes in modular characteristics and mineral composition of clinker from the type and amount of technogenic raw materials in the composition of the charge have been established. At SC= 0.92, with an increase in silicate modulus from 2.2 to 2.7, the content of lead slag in the charge decreases from 4.63 to 1.36%, and the alumina modulus increases from 0.99 to 1.52, the content of alite and C_3A increases, and the proportion of C_4AF decreases. The strength and speed of hardening of such cement will be higher.

Table 3 - Influence of mixture compositions and temperature on the process of binding calcium oxide

Raw mix composition, wt%				SC	Modules		Content of CaO free,%, at burning temperature, °C			
Limes tone	coal mining waste	tephrite basalt	lead slag		n	p	1300	1350	1400	1450
77.02	9.79	9.78	3.41	0.92	2.2	1.15	3.4	2.1	1.9	0.6
76.72	9.10	9.11	5.06	0.94	2.0	0.95	1.3	0.2	-	-

In the 1st series of raw mixtures "Limestone + coal mining waste + cinders" the content of coal waste is 20-23%. In this case, 3-4% of coal is added to the composition of the raw mixture. This will reduce the fuel consumption for burning clinker in the kiln. The cinder content is 0.86-2.69%. An increase in the silicate modulus from 2.2 to 2.7 leads to a gradual decrease in the proportion of pyrite cinders from 2.69 to 0.49%. The magnitude of the p modulus increases from 0.89 to 1.52. The resulting clinkers contain 3.1-7.76% tricalcium aluminate. These clinkers are suitable for the manufacture of low-aluminate sulfate-resistant clinkers and cement.

In the second batch of charges, scarce pyrite cinders were replaced by lead slags. The iron content in slags is slightly lower than in cinders. Therefore, the content of lead slag in the charge increases and reaches 3.95-6.9%. Similarly to series 1 of mixtures, an increase in the modulus n leads to a gradual increase in the alumina modulus from 0.89 to the optimal 1.12-1.33. The specific consumption of lead slag increases to 78-106 kg per 1 ton of clinker.

In the third series, coal mining waste and tephrite basalt are used as the aluminum-containing component, lead slags are the correcting additive. Slags, together with tephrite basalt, add a sufficient amount of iron oxides to the charge composition.

In the developed low-energy-intensive mixtures, the specific consumption of raw materials for obtaining 1 ton of clinker is reduced to 1516-1525 kg, which is much lower than in traditional raw mixtures. The mass of the material that must be heated in the furnace to the clinker formation temperature of 1300-1350°C is reduced, which makes it possible to further reduce fuel consumption. The content of lead slag in the mixture is from 70 to 106 kg, due to which from 10 to 15 kg of non-carbonate lime is added per 1 ton of clinker, which improves the heat balance of the burning process.

The effect of temperature on the clinker burning process is shown in Table 3.

As can be seen from the data in Table 3, depending on SC, n, and the content of lead slag, the processes of clinker formation in the developed low-energy-intensive raw mixtures are completed at 1300 or 1350°C. The content of unbound CaO is 1.3 and 2.1%, respectively. A raw mixture with a higher silicate modulus n=2.2 and a lower lead slag content of 3.41% is burned more difficult - the content of CaO free at 1300°C is 3.4%. An increase in the content of lead slag to 5.06% leads to a decrease in the silicate modulus to 2.0, an acceleration of the clinker formation process, and an improvement in sintering.

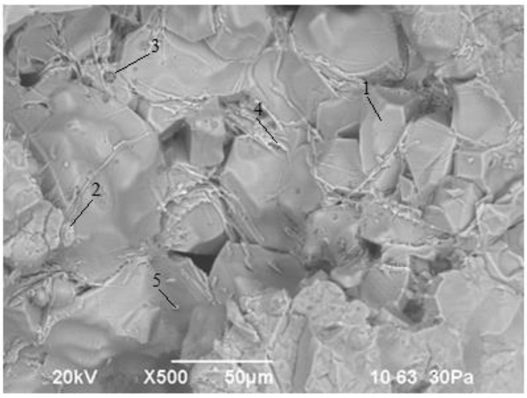
The lead slag contains 4.34% ZnO, 0.52% PbO, 1.1% CuO, which have a mineralizing effect on the sintering of the charge. Zinc oxide is a catalyst for the formation of minerals in the production of clinker. Zn^{2+} ions replace Ca^{2+} in the crystal lattices of minerals with the formation of solid solutions. The solubility of zinc oxide in C_3S is slightly more than 2% [20]. Differential thermal analysis of limestone with additions of 3-10% lead slag showed that the maximum of the $CaCO_3$ decarbonization process decreases by 30-50°C. The above will allow completing the processes at low temperatures, reducing the heat costs for clinker burning, increasing the furnace productivity, and reducing the consumption of nozzle fuel [21].

The developed raw mixes contain two technogenic products - coal mining waste and lead slag, as well as a low-melting igneous rock – tephrite basalt. Their total content is about 23-26% [22].

The synthesized clinker was subjected to microscopic examination. Using an electron microscope in the field of binders, one can study the following issues: the shape and size of individual crystals: the processes of growth and destruction of crystals, processes occurring at the grain boundary, and a number of other more particular problems [23].

The dissertation studied the microstructure of synthesized clinkers. Samples for research were cleaved from synthesized clinker.

Analysis of micrographs from a clinker chip obtained from the traditional raw mix "Limestone + coal mining waste + cinders" is shown in **Figure 1.**



1 - C_3S; 2 – C_2S; 3 – C_3A; 4 - C_4AF; 5 – CaO.

Figure 1 - Micrograph of a clinker chip obtained from traditional raw materials

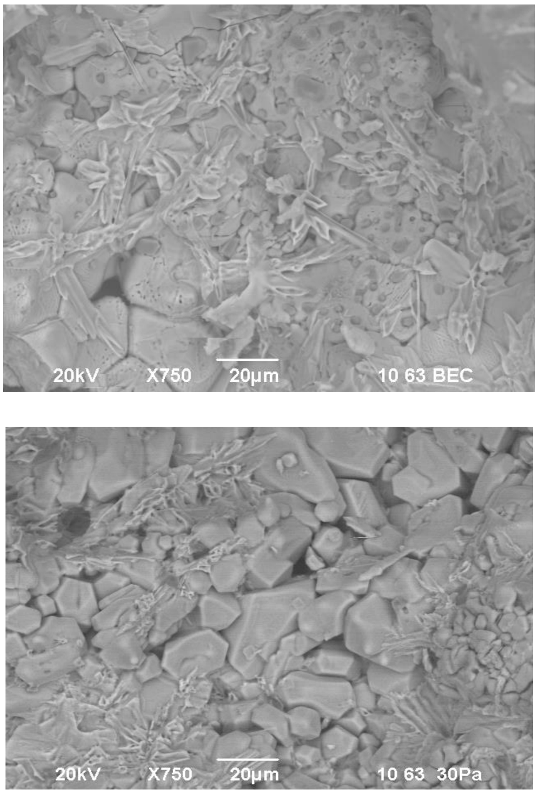


Figure 2. Micrographs from clinker chips obtained from the raw mixture Limestone + coal mining waste + tephrite-basalt + lead slag

Micrograph of clinker allows us to conclude that crystallization is relatively clear, the distribution of minerals is not uniform. Along with areas of relatively well-formed alite crystals, there are areas with crystals of an indefinite shape, passing into intergrowths of alite crystals.

Belite inclusions are observed in large crystals of alite. Belite is represented by small round and oval crystals. The amount of belita is much less than that of alita. The micrograph also shows a slight accumulation of crystals of free calcium oxide (dark rounded crystals). On the surface of crystals and between them, an alumoferite phase (light intermediate substance) is clearly visible. The presence of the aluminate phase in this micrograph is not determined [24].

Analysis of micrographs from a clinker chip obtained from an unconventional energy-saving raw mixture of Limestone + coal mining waste + tephrite-basalt + lead slag **(Figure 2)**.

Figure 2 shows a photomicrograph from cleavage of clinker. Analysis of micrographs allows us to conclude that the crystallization of minerals is clear, but their distribution is uneven. Along with areas where alite has a regular geometric shape, there are areas with an indefinite crystal shape. Belite is represented by crystals of round and oval shape; cracks are observed on the surface of belite granules.

Alite crystals of large size, reaching 100-140 microns. A small amount of lead slag (2.57%) introduced into the mixture has a mineralizing effect. This, together with basalt tephrite, reduces the burning temperature from 1450 °C to 1350 °C, the CaOf content decreases to 1.39%. There is a lot of intermediate phase and it is represented mainly by calcium aluminoferrite. Calcium alumoferrites are represented by light needle-like crystals. The content of clinker minerals is C_3S — 57.88%, C_2S — 18.82%, C_3A — 6.46%, and C_4AF — 11.61%.

The presence of three cement plants in the South Kazakhstan region: LLP Standard Cement, JSC Shymkentcement and LLP Sastobe Technologies with a total capacity of ~ 3.5 million tons would allow for a short period of time to completely utilize lead slag and coal waste in cement production. This would help to reduce the anthropogenic load on the environment, eliminate major sources of its pollution [[25], [26]].

Conclusions

1. Experiments have shown the possibility of using technogenic waste as part of the charge: coal mining waste - 9.79%, lead slag - 3.41-5.06%, and tefrito bazalt - 9.78%, that is, to dispose of up to 25%.

2. To improve the clinker formation process and the microstructure of the main minerals of alite

due to the introduction of lead slag. In the composition of lead slag, zinc oxide performed the task of a mineralizer and accelerated the process of clinker formation at 1350°C. The free calcium oxide in the composition of clinker was 0.2 % at a temperature of 1350°C.

3. With coal mining waste, from 2 to 3.5% of coal is introduced into the incinerated charge, which made it possible to reduce heat costs and nozzle fuel consumption for clinker combustion. Significantly reduced greenhouse gas CO_2.

Thus, the developed low-energy resource-saving compositions of raw materials for the production of cement clinkers using man-made raw materials will increase the efficiency of cement production, reduce environmental pollution and improve the ecology of the region.

Conflicts of interest. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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[1,2]Жаникулов Н.Н., [3*]Колесников А.С., [3]Таймасов Б.Т., [4]Жакипбаев Б.Е., [3]Шал А.Л.

[1]Академик Е.А. Букетов атындағы Қарағанды университеті, Қарағанды, Қазақстан
[2]М.В. Ломоносов атындағы Мәскеу мемлекеттік университеті, Мәскеу, Ресесй Федерациясы
[3]М.Әуезов атындағы Оңтүстік Қазақстан университеті, Шымкент, Қазақстан
[4]Болашақ университеті, Қызылорда, Қазақстан

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ТҮЙІНДЕМЕ
Цемент өндірісінің негізгі проблемасы - энергия шығынын төмендету және атмосфераға шығарылатын CO_2 парниктік газдар мөлшерін азайту. Мәселені шешудің алғышарты: техногендік қалдықтар мен дәстүрлі емес шикізатты шикіқұрамның құрамында қолдану және CO_2 газын шығару көзі әктастың мөлшерін 85% - дан 70-75% - ға дейін төмендету. Мақалада техногенді қалдықтарды және тефритобазальт, көмір өндірісінің қалдықтары және қорғасын шикіқұрамы сияқты дәстүрлі емес шикізат материалдарын физика-химиялық зерттеу нәтижелері келтірілген. Материалдардың химиялық және минералогиялық құрамы және олардың экологиялық таза клинкер алуға жарамдылығы анықталды. Энергияны аз қажет ететін ресурстарды үнемдейтін технологияларды қолдана отырып, цемент клинкерін алу мүмкіндігі көрсетілген. Әзірленген қоспалардағы клинкер процестері 1350°С температурада аяқталады, бұл қалдықтардағы көмірмен бірге бүріккішті (форсунка) отын шығынын азайтады және атмосфераға CO_2 шығарындыларын азайтады. Әзірленген аз энергиялы қоспаларда 1 тонна клинкер алуға арналған шикізаттың меншікті шығыны 1516-1525 кг дейін төмендейді, бұл дәстүрлі шикізат қоспаларына қарағанда айтарлықтай төмен. Алынған клинкердің минералогиялық құрамы C_3S — 57,88%, C_2S — 18,82%, C_3A — 6,46% және C_4AF -11,61%. Көп тоннажды қалдықтарды кәдеге жарату қоршаған ортаның ластануын азайтуға және өңірдегі экологиялық жағдайды жақсартуға мүмкіндік береді.
Түйін сөздер: клинкер, микроқұрылымды жақсарту, қалдықтарды жою, температураны төмендету, минерализатор.

Авторлар туралы ақпарат:

Жаникулов Нургали Нодырұлы	*PhD докторы, профессор ассистенті, Академик Е.А. Букетов атындағы Қарағанды университеті, Қарағанды, Қазақстан; e-mail: nurgali.zhanikulov@mail.ru; ОРСИД: 0000-0002-0750-9753; Болашақ бағдарламасының стипендиаты, М.В. Ломоносов атындағы Мәскеу мемлекеттік университеті, Мәскеу, Ресей Федерациясы*
Колесников Александр Сергеевич	*профессор, техника ғылымдарыңң кандидаты, М.Әуезов атындағы Оңтүстік Қазақстан университеті, Шымкент, Қазақстан, e-mail:kas164@yandex.ru; ОРСИД: 0000-0002-8060-6234*
Таймасов Бахитжан Таймасович	*профессор, техника ғылымдарыңң докторы, М.Әуезов атындағы Оңтүстік Қазақстан университеті, Шымкент, Қазақстан, e-mail: taimasovukgu@mail.ru, ОРСИД:0000-0002-1844-4932*

Жакипбаев Бибол Ермуратович	*Қауымдастырылған профессор м.а., PhD докторы, Болашақ университеті, Қызылорда, Қазақстан, e-mail: bibol_8484@mail.ru; ОРСИД: 0000-0002-1412-7796*
Шал Айжан Лесқызы	*магистрант, М.Әуезов атындағы Оңтүстік Қазақстан университеті, Шымкент, Қазақстан, e-mail: shalaizhan@mail.ru, ОРСИД: 0000-0002-0291-6894*

Влияние промышленных отходов на структуру экологически чистого цементного клинкера

[1,2]Жаникулов Н.Н., [3*]Колесников А.С., [3]Таймасов Б.Т., [4]Жакипбаев Б.Е., [3]Шал А.Л.

[1]Карагандинский университет имени академика Е.А. Букетова, Караганда, Казахстан
[2] Московский государственный университет им. М.В. Ломоносова, Москва, Российская Федерация
[3]Южно-Казахстанский университет им. Ауэзова, Шымкент, Казахстан
[4]Университет Болашак, Кызылорда, Казахстан

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АННОТАЦИЯ
Основной проблемой производства цемента является снижение энергозатрат и снижение количества парниковых газов CO_2, выбрасываемых в атмосферу. Предпосылкой решения проблемы является: применение техногенных отходов и нетрадиционного сырья в составе шихты и снижение содержания известняка с 85% до 70-75% как источника выделения газа CO_2. В статье представлены результаты физико-химических исследований техногенных отходов и пригодность нетрадиционных сырьевых материалов таких, как тефритобазальт, отходы угледобычи и свинцовый шлак. Установлен химический и минералогический состав материалов для получения экологического чистого клинкера. Показана возможность получения цементного клинкера с использованием малоэнергоемких ресурсосберегающих технологий. Установлено, что процессы клинкерообразования в разработанных смесях с углесодержащими отходами завершаются при температуре 1350 °С, что, способствует снижению расхода форсуночного топлива и уменьшению выбросов CO_2 в атмосферу. В разработанных малоэнергоемких смесях удельный расход сырья на получение 1 тонны клинкера снижается до 1516-1525 кг, что значительно ниже, чем в применении традиционных сырьевых смесей. Определен минералогический состав получаемого клинкера, который представлен C_3S- 57,88%, C_2S- 18,82%, C_3A- 6,46% и C_4AF- 11,61%. Утилизация крупнотоннажных отходов позволит снизить загрязнение окружающей среды и улучшить экологическую обстановку в регионе.
Ключевые слова: клинкер, улучшение микроструктуры, утилизация отходов, снижение температуры, минерализатор.

Информация об авторах:

Жаникулов Нургали Нодырұлы	*Доктор PhD, ассистент профессор, Карагандинский университет имени академика Е.А. Букетова, Караганда, Казахстан; e-mail:nurgali.zhanikulov@mail.ru, ОРСИД: 0000-0002-0750-975; Стипендиат программы "Болашак", Московский государственный университет имени М.В. Ломоносова, Москва, Российская Федерация*
Колесников Александр Сергеевич	*Профессор, кандидат технических наук, Южно-Казахстанский университет им. М.Ауэзова, Шымкент, Казахстан, e-mail: kas164@yandex.ru; ОРСИД: 0000-0002-8060-6234*
Таймасов Бахитжан Таймасович	*Профессор, доктор технических наук, Южно-Казахстанский университет им. М. Ауэзова, Шымкент, Казахстан, e-mail: taimasovukgu@mail.ru; ОРСИД: 0000-0002-1844-4932*
Жакипбаев Бибол Ермуратович	*И.о. асоц. профессор, доктор PhD, Университет Болашак, Кызылорда, Казахстан, e-mail: bibol_8484@mail.ru; ОРСИД:0000-0002-1412-7796*
Шал Айжан Лесқызы	*Магистрант, Южно-Казахстанский университет им. М. Ауэзова, Шымкент, Казахстан, e-mail: shalaizhan@mail.ru; ОРСИД: 0000-0002-0291-6894*

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