IOP Conference Series: Materials Science and Engineering

Quality evaluation of carbonaceous industrial by-products and its effect on properties of autoclave aerated concrete

Quality evaluation of carbonaceous industrial by-products and its effect on properties of autoclave aerated concrete

E V Fomina, V S Lesovik, A E Fomin, N I Kozhukhova, M S Lebedev

Belgorod State Technological University named after V.G. Shoukhov, 46 Kostukov St., 308012, Russia

Abstract. Argillite is a carbonaceous industrial by-product that is a potential source in environmentally friendly and source-saving construction industry. In this research, chemical and mineral composition as well as particle size distribution of argillite were studied and used to develop autoclave aerated concrete as partial substitute of quartz sand. Effect of the argillite as a mineral admixture in autoclave aerated concrete was investigated in terms of compressive and tensile strength, density, heat conductivity etc. The obtained results demonstrated an efficiency of argillite as an energy-saving material in autoclave construction composites.

1. Introduction

Development of advanced technologies for construction material production in order to achieve radically new characteristics in final products is an impossible task without application of new technological methods including alternative raw materials [1, 2]. Nowadays, the number of industrial by-products, secondary and wastes materials is constantly growing and accumulated in landfills. Irrational coal deposit development also initiates the increase of industrial waste.

Open waste storage leads to change the geomorphology of the Earth's surface, hydrogeological cycle of ground waters, waste dusting and, generally, environmental contamination [3, 4]. As a result of industrial pollution, there is an environmental depletion and rapid shortage of natural resources, and misuse of good quality secondary materials applicable in construction industry. This problem can be solved by more effective application of the waste as binder components, aggregates and mineral admixtures in construction materials [5–7].

Widely applied technology of autoclave material production allows a wide range application of the off-spec industrial and natural resources.

In this research, effect of the coal-bearing source on properties of autoclave aerated concrete were studied using by-products from the Korkin coal field. The Korkin coal field is one of the largest coal formation in Europe. The large area of the deposit zone is occupied by landfills. Technologies oriented towards the application of by-products from the Korkin coal field in roads and pavements construction are not able to complete waste utilization. These by-products and waste materials are represented by polymineral composition with different types of structural bonds and morphology [12].

One of the major components of by-products from the Korkin coal field is argillite. Argillite is overburden aluminosilicate rock containing coal impurity and could be applied for autoclave composite binders and products (Table 1). This study was focused on estimation of quality of argillite as a component in autoclave aerate concrete.

2. Material and methods

Polarizing microscope POLAM-312 in the reflected light mode was used for petrographic analysis of the argillite polished section. XRD-analysis was realized with diffractometer DRON-4 using Cu $\lambda_{K\alpha1,2}$

radiation and Ni-filter for reflection of β-component depression with a step angle of 0.05º. Duration of intensity measurement in scan point was 1 sec.

Scanning electron microscope Hitachi S-800 was used in this study.

Argillite was ground using a ball mill. A particle size distribution was accomplished using laser particle analyzer Analysette 22NanoTec plus and particle analyzer Microtrac S3500. The CaO_{free} content in argillite composition as an indicator of reactivity was detected by a titration method using ethyl-glycerate solution [13].

In this study CEM I 42,5N Portland cement (LTD Belgorodsky cement, Russia), quick lime (reactivity was 82 %), aluminum paste GPB-1; quartz sand (Bezludovsk deposit, Russia) were used as main components for autoclave aerated concrete production.

Mechanical tests have been carried out on 100×100×100 mm cubes. Autoclave treatment of the casted specimens was performed under the following curing regime: air-flush of the specimens in the curing chamber for 40 min; after that pressure was buildup by 10 atmospheres and temperature up to 183 °C during 1 hour. The curing process was performed at constant 10 atmospheres and 183°C for 5 hours followed by pressure release during 2 hours.

3. Experimental part

Genetic properties of silica for autoclave aerated concrete production are very important due to its reactivity to quick lime in binding system at early hardening period before autoclave curing. It is related to a decrease of $Ca(OH)_2$ solubility as the temperature during the autoclaving process increases. Typical silica in autoclave aerated concrete is quartz sand containing more than 90 % of SiO_2, as well as clay impurities up to 1,5 % [14]. The chemical composition of argillite mostly represented by the oxides of SiO_2 (67 %) and Al_2O_3 (21.55 %). LOI was 7.65 % confirming a high concentration of organic and clay impurities in the mineral (Table 1).

Table 1. Chemical composition of carbonaceous by-product from the Korkin coal field

Component	Oxides content, wt. %						
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	LOI
Argillite	67,09	21,55	0,02	0,78	0,28	-	7,65
Quartz sand	92,4	2,36	0,77	1,88	0,2	0,05	1,95

The studied argillite was presented by several crystal phases such as quartz, illite, feldspar, clay minerals (Table 2, Fig. 1).

Table 2. Mineral composition of argillite

Minerals	Content, %
Quartz	30.07
Illite	8.74
Kaolinite	4.28
Albite	3.34
Clinochlore	1.13
Biotite	1.62
X-ray amorphous phase	50.83

More than 50 % of argillite was X-ray amorphous (Table 2) containing nano-scaled silica polymorphous modification crystobalite and tridymite (Table 3).

IOP Conf. Series: Materials Science and Engineering **327** (2018) 042033 doi:10.1088/1757-899X/327/4/042033

Table 3. Mineral composition of argillite[1]

	Mineral	Content, wt. %	Size, nm
Argillite	crystobalite	46	1
	tridymite	20	1.6
	α-quartz	34	27

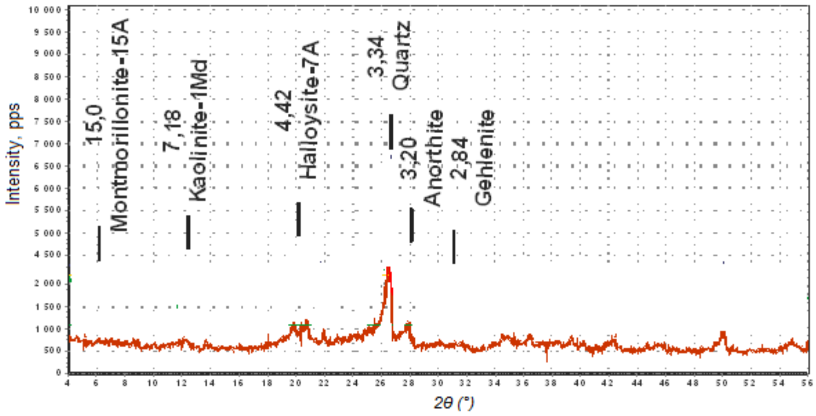


Figure 1. Quantitative XRD-analysis of argillite

SEM analysis of argillite demonstrated isometric plates, typical for layered aluminosilicate minerals. According to the argillite structure, it was formed during the katagenesis period when packing, dehydration and consolidation of clays took place [15]. A structural skeleton of argillite is represented by the aggregates of coagulated quartz grains and clay matrix (Fig. 2, b).

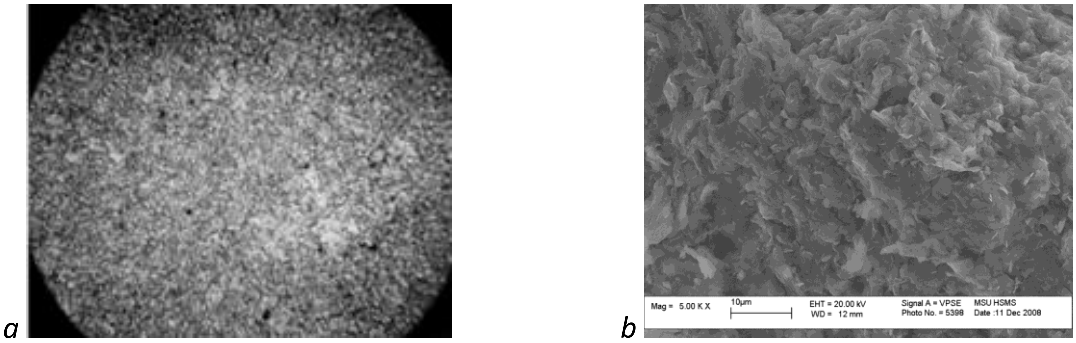


Figure 2. Petrographic (*a*) and SEM images (*b*) of argillite

Also the dark spots, identifying impurities associated with carbon component, were observed in the microstructure (Fig. 2, a).

High grindability of argillite is associated with lower hardness of consisting minerals vs. quartz (major component of quartz sand) and polymineral composition providing a significantly lower strength in interface zone vs. internal mineral strength.

[1]Data were calculated with quantitative full-profile XRD-analysis using a Rietveld algorithm. The Derivative Difference Minimization algorithm (DDM) software was used to quantify the X-ray amorphous phase

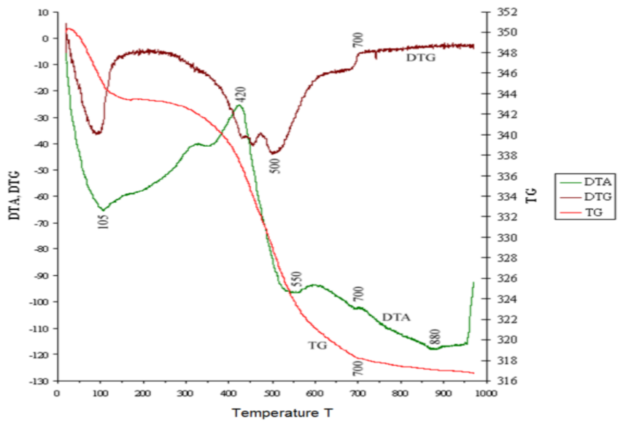
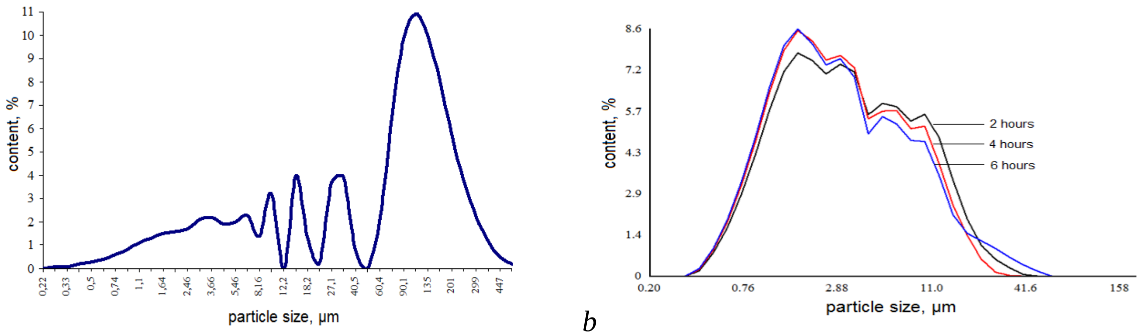


Figure 3. DTA-analysis of argillite

Table 4. Grindability of components of by-products from the Korkin coal field

Component	Specific surface area, m^2/kg, depending on milling time, hours							
	0.5	1	1.5	2	2.5	3	4	6
Quartz sand	190	244	296	354	417	497	520	640
Argillite	362	457	573	622	720	880	920	936

According to particle size destribution analysis, argillite particle-size distribution curve shifted towards the lower size range from 80–200 µm to 1–11µm after 2 hours of milling (Fig. 4, b).



a *b*

Figure 4. Particle size distribution analysis of argillite: a – before milling; b – after 2, 4 and 6 hours of milling

It was assumed that the size distribution range for argillite includes separate particles of clay minerals and conglomerates that can affect the fineness of argillite. Nano-sized crystobalite phase also significantly increases specific surface area due to its developed surface. Further grinding process (more than 2 hours) slightly effects on fineness of material (Fig. 4, b, particle-size distribution curves after 4 and 6 hours of grinding). The peak at 5–7 µm was slightly shifted to the area of 1–2 µm.

The mix of aerated concrete was designed to be applicable for a small-sized blocks cutting technology production to provide concrete density of 500–600 kg/m^3 and compressive strength 2,5–3.5 MPa [16].

The reference mix of aerated concrete had quick-lime – 11.6 %; quartz sand – 68.3 %; Portland cement – 20 %; aluminum paste – 0.1 %; «water-solid» ratio – 0.5.

Plastic strength is one of the basic characteristics in autoclave aerated concrete production which has to be considered before the cutting process and has to vary in the range of 0.03–0.08 MPa after 3-hours of pre-autoclave hardening.

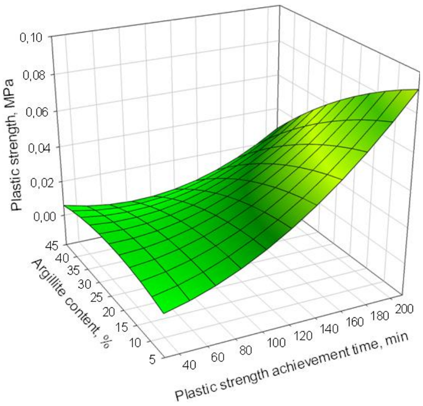


Figure 5. Effect of argillite incorporation on plastic strength of aerated concrete

In the experimental composition, quartz sand was partially replaced by argillite. The mix design of autoclave aerated concrete containing argillite was developed with experiment design using a regression equations method (Fig. 5).

The result of the test demonstrated that the most effective argillite concentration was 15 % providing shorter time of plastic strength development up to 90 min. A further increase of argillite in the system up to 35 % prolongs the strength development to 3 hours that identified a retarding effect of strength development.

Effectiveness of the hydration process of the argillite-contained binder was associated with improved reactivity of CaO_{free} to form hydrates at initial stage of the binder hardening before autoclave curing. According to the obtained data, specimens with 15 % of argillite had the highest binding degree of CaO_{free}.

Table 5. Content of CaO_{free} in the concrete mixture

№	Argillite content in silica component, %	Content of CaO_{free}, (wt. %)
1	0	27.83
2	5	24.78
3	10	21.58
4	15	20.37
5	20	21.56
6	25	22.13
7	30	22.79

Addition of argillite initiated variations in physical and mechanical properties of the binder. Replacement of 5–25 % of quartz sand by argillite led to the flowability increase of the concrete mixture favorable for a gas forming process. However, water demand of the mixture was also increased (Table 5).

Table 6. Properties of autoclave aerated concrete

Composition	Argillite content in silica component, %	«water-solid» ratio	Density, kg/m^3	Compressive strength, MPa	Tensile strength, MPa	Heat conductivity, W/(m·C°)	Water vapour permeability, mg/(m·h·Pa)	Viscosity, cm	Porosity, %
1	0	0.50	562	4.23	1.05	0,121	0.210	28	70
2	5	0.50	483	4.86	1.25	0,115	0.211	35	75
3	10	0.55	473	4.97	1.24	0,107	0.212	37	80
4	15	0.60	479	5.18	1.29	0,111	0.211	40	75
5	20	0.65	526	4.84	1.21	0,117	0.211	44	73
6	25	0.70	565	4.35	1.03	0,119	0.210	48	73
7	30	0.75	579	4.14	1.09	0,128	0.209	50	69

Replacement of 5−20 % of quartz sand by argillite provided the highest values of strength increase and density decrease. Higher concentration of argillite increased water demand leading to a massive shrinkage and strength decrease (Table 6, composition 7).

Results of physical and mechanical characteristics of the concrete specimens demonstrated the increase in compressive strength by 22.5%, tensile strength - by 23 % and water permeability - from 0.210 to 0.211 mg/(m·h·Pa) by replacement of 15 % of quartz sand by argillite. At the same time, with addition of argillite, the density as well as a heat conductivity reduction by 15 % and 8 % respectively was observed.

4. Summary
Agrillite from the Korkin coal field is alumosilicate carbonaceous industrial by-product with 50.83 % of X-ray amorphous phases like crystobalite and tridymite (polymorphous modifications of silica); crystal phases such as quartz and clay minerals as well as carbon. Argillite has a good grindability allowing production of particles with sizes of 1–11μm. In alkaline media argillite has an excellent reactivity stimulating hydration process.
An addition of argillite into the concrete mixture up to 25% increases water demand and mix flowability. At the same time, the incorporation of 15 %-concentration of argillite as a binder component provides compressive and tensile strength boost by 22.5 and 23 %, but leads to a density and heat conductivity reduction by 15 % and 8 %, respectively.

5. Conclusion
In the light of ecological aspect, this research study was focused on the analytical and experimental investigation of argillite characteristics as a resource for autoclave aerated concrete production. Replacement of 5–20 % of quartz sand by argillite helps to increase chemical interaction of the concrete component and improve its physical and mechanical performance. The study results demonstrated the efficiency of argillite application as a material- and energy-saving component in production of construction materials.

6. Acknowledgements
This research work is financially supported by the Ministry of Education and Science of the Russian Federation in the framework of State Assignment №11.9329.2017/8.9, the Russian Federation Ministry of Education and Science in the framework of the Presidential Scholarship № PS-2099.2015.1, using equipment of High Technology Center at BSTU named after V.G. Shoukhov.

References
[1] Lesovik V 2016 *Geonika (geomimetics). Implementation examples in construction material science: monography* (Belgorod: BSTU named after V.G. Shoukhov)
[2] Lesovik V S 2014 State and application potential of industrial raw materials *Scientific and engineering problem of construction and technological utilization of industrial wastes* (Belgorod: BSTU named after V.G. Shoukhov) pp 17–21
[3] Abdrarhimova E, Abdrakhimov V 2014 To the question about economic and ecological expediency uses of waste products coal enrichment coal argillites in manufacture heat isolation of materials on the basis of interslate clay *J. Ecological systems and machines* **1** 35–42
[4] Stolbushkin A, Akst D, Fomina O, Ivanov A, Syiromyasov V 2017 Estimation of by-product coal from enterprises of the South Kuzbass as raw component for production of ceramics materials *J. H-Tech technologies for development and introduction of mineral sources* **3** 335-340
[5] Talpa B, Kotlyar A 2015 Mineral-Raw Material Base of Lithified Clay Rocks of the South of Russia for Production of Building Ceramics *J. Construction materials* **4** 31–33
[6] Abdrakhimov V, Lobachev D, Abdrakhimova E 2016 Utilization of by-product coal when light-weight brick production *J. Biospheric compatibility: human, region, technologies* **3 (15)** 30–37
[7] Kairakbaev A, Abdrakhimov V, Kolpakov A, Abdrakhimova E 2015 Study of the effect of Al_2O_3 on acid and thermal shock resistance of acid-resistant refractories using a regression analysis method *J. Refractories and Industrial Ceramics* **56 (3)** 276–280
[8] Lesovik R, Nosova A, Savin A, Fomina E 2014 Assessment of the suitability of the opal-cristoballite rocks of korkinsk deposit in the construction industry *J. World Appl Sci.* **29 (12)** 1600–1604
[9] Tolstoy A, Kovaleva I, Novikov K 2016 Enhancement of composition and properties of powdered concrete containing industrial raw materials *J. Bulletin of BSTU named after V.G.*

Shoukhov **2** 19–24
[10] Lebedev M, Zhernovskiy I, Fomina E, Fomin A 2015 Features of Clay Rocks Application when Construction Material Production *J. Construction materials* **9** 67
[11] Lebedev M 2016 On structure formation of bitumen binder containing aluminosilicate fillers *International research and practice conference, devoted to the 70-Universe of professor Lesovik V S* (Belgorod: BSTU named after V.G. Shoukhov) pp 134–140
[12] Lebedev M, Zhernovsky I, Strokova V, Fomina E, Kozhukhova N 2014 Application of IR-spectroscopy for study of alumosilicate raw materials *J. Appl. Mech. and Mater.* **496–500** 96–101
[13] Butt Y, Timashev V 1973 *Practicum on chemical technology of binder materials* (Moscow: High School)
[14] Russian Standard 31359-2007 *Cellular autoclave curing concretes. Specifications*
[15] Rogozina E 1968 Porosity variation in argillite rock in katagenesis zone *J. Oil and Gas Geology* **11** 33–35
[16] Russian Standard 25485-89 *Cellular concretes. Specifications*