

Development of building materials based on a high content of fly ash and polycondensation products from Chinese heat and power plants

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The addition of fly ash from thermal power plants (TPP) and chemical additives, such as polycondensation products of acetone and formaldehyde, is an effective and economical method for increasing the strength and durability of building materials, in particular concrete. Fly ash added to the concrete and mortars at 10%–15% does not reduce their technical properties, while polycondensation products of acetone and formaldehyde plasticize and accelerate the hardening process of concrete. The study aims to substantiate the possibility of obtaining concrete on dense aggregates with a high content of fly ash through the use of polycondensation products of acetone and formaldehyde as additives, which are highly soluble in water and polyfunctional. The strength indicators were determined using standard methods. The study has shown quantitative changes in the properties of the fly ash-concrete mixture, improvement of its physical and technical characteristics, and durability of the fly ash concrete with the addition of polycondensation products of acetone and formaldehyde. In the present study, the authors determine the effect of fly ash addition on the properties of a concrete mixture, as well as the maximum possible content of fly ash that can be added to concrete to maximize certain properties required of the mixture; additionally, the same action is conducted with the addition of polycondensation products of acetone and formaldehyde concomitant with the addition of fly ash.

Keywords: *acetone-formaldehyde resins, ash-and-slag mixture, concrete properties, fly ash, mineral additive*

1. Introduction

Given today's growth of urban areas and industrial sectors, it is possible to state that concrete has become a "vital" construction material. Nowadays, it is difficult to imagine the construction of bridges, pavements, and buildings without concrete. Therefore, its production is increasing day by day. At the same time, the increased competitiveness in the market leads to inflated requirements for the technical, physical, and performance qualities of construction materials.

Cement is another crucial component of construction activity. Even though it is difficult to overestimate the role of cement in construction, its production entails numerous environmental problems. The indispensable condition for manufacturing cement is extremely high temperature, which, in turn, causes high energy consumption and generates large quantities of carbon dioxide [\[1\]](#page-16-0). Consequently, it is possible to state that the cement manufacturing process significantly increases greenhouse gas emissions. The high ash content (30%– 50%) of the coal in the world even worsens the environmental problem. Safe disposal of fly ash is a complicated challenge since it should not have an adverse effect on the environment.

In addition to the enhancement of technical, physical, and performance properties of materials and ecologically friendly production, modern construction activity faces one more challenge, namely the cost reduction of building materials. It is possible to solve this and other issues by the additional introduction of additives. The introduction of fly ash as an additive to cement concrete mixtures can solve several tasks at once: substantial savings in the consumption of cement; an economic and useful avenue for disposal of fly ash; the reduction in

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the cost of building materials; the enhancement in physical and technical characteristics [\[2\]](#page-16-1). Therefore, the introduction of inexpensive but effective additives such as ash-and-slag wastes in materials is quite a relevant area of research.

Thermal power plants (TPP) use anthracite, hard and brown coal, slates, and peat as fuel. Their reserves are significant, and the residuals of their combustion are fly ash consisting of a noncombustible mineral part of coal and organic fuel particles. Its fine particles have binding properties, contain active mineral additives, and are an effective way to save cement $[3]$. The high degree of scientific know-how and technological sophistication required to put fly ash to a productive use, and high cost of these technologies, limit their adoption in the present era. Modern coal generation technologies provide for the formation of solid waste, and their tailings harm the environment [\[4\]](#page-16-3).

At high temperatures $(1,300^{\circ}C-1,600^{\circ}C)$ of the process involved in coal combustion, the mineral part of the fuel, being non-combustible, is obtained as a residue, which is usually left behind in forms such as ash, lumpy slag, and ash-and-slag mixtures (ASMs). The main reason for the infrequent use of ash-and-slag waste (ASW) is the imperfect regulatory framework in terms of their use in various areas of economic activity. A prominent national concern is to increase the use of industrial wastes as secondary raw materials [\[5\]](#page-16-4). Since construction and the production of building materials are material-intensive, these two processes are the ones that offer the maximum (and most promising) scope for the mass involvement of ASW usage as secondary raw materials. Ash and slag can be a source material in the production of cement and heat-insulation materials or an aggregate with the replacement of sand and gravel in construction, including road and railway construction, where their share of usage is very high [\[6\]](#page-16-5).

The utilization of ASW is extensive in construction and it represents the second most essential component of building materials after cement. ASW is also used for manufacturing cement itself and products based on it, such as brick, cinder block, and aggregate for concrete. It is possible to use ASW for landscaping, general construction works, embankments, or backfilling of trenches. They also represent an insulating material at landfills for solid household waste. Moreover, ASW is a widely used construction component when it comes to road bases and pavements [\[7\]](#page-16-6). The utilization of ASW for manufacturing basic building materials such as cement, concrete, dry mixtures, or paving slabs makes it possible to reduce costs by $12\% - 25\%$ [\[8\]](#page-16-7).

In addition, individual components contained in these wastes, such as aluminosilicate microspheres, are used for manufacturing paints, highquality heat-insulation materials, etc. [\[9\]](#page-16-8). Many countries develop various methods and technologies for ASW utilization and put them into practice, making it possible to offer an alternative to natural raw materials in manufacturing building materials. At the same time, China is the most promising market for the use of cement in the world. Thus, it is the most promising to introduce technologies that reduce the cost of building materials, including the use of ASMs. It is advisable to use local ASWs from Chinese TPPs in order to avoid logistical costs.

Despite all these advantages, the introduction of fly ash has one significant disadvantage. The high ash content raises the water demand of cement and reduces strength under initial and design conditions. The authors of this article argue that the use of polycondensation products of acetoneformaldehyde resin in an optimum amount compensates for water consumption and evens out the strength characteristics of cement under both initial and design conditions. A group of researchers from Kalasalingam Academy of Research and Education also highlights the enhancement properties of acetone-formaldehyde resins, such as increased workability and higher mechanical strength [\[10\]](#page-16-9).

Thus, the novelty of this study lies in its examination of the combined use of ash and additives of acetone-formaldehyde resins in cement concrete mixtures and the substantiation of its contribution to the formation of an additional amount of hydrates and compaction of the cement stone, reduction in the carbonization rate, and thus increase in the durability of concrete. The study aims to substantiate the possibility of obtaining concrete on

Basic oxides	Coal ash $(\%)$			Shale ash $(\%)$	Peat ash $(\%)$	
	Anthracite	Stone	Brown		Siliceous	Calcified
Si ₂ O ₃	$16-50$	$22 - 60$	31-58	20-60	42	15
Al_2O_3	5-37	$15 - 43$	$20 - 45$	$5-20$	16	10
Fe ₂ O ₃	$5-40$	$3 - 32$	$2 - 45$	$2 - 10$		15
CaO	Up to 3	Up to 5	Up to 50	$10-15$	18	50

Table 1. Chemical composition of ashes

Source: Compiled by authors according to Big Chemical Encyclopedia [\[12\]](#page-16-10).

dense aggregates with a high content of fly ash by using polycondensation products of acetone and formaldehyde as additives. It is necessary to fulfill the following objectives to achieve the aim set:

- 1. to determine the effect of fly ash addition on the properties of a concrete mixture, as well as the maximum possible content of fly ash that can be added to concrete to maximize certain properties required of the mixture; and to conduct the same investigation with the addition of polycondensation products of acetone and formaldehyde concomitant with the addition of fly ash;
- 2. to define the features of the ash-concrete technology incorporating the addition of polycondensation products of acetone and formaldehyde and substantiate the technical and economic efficiency of the practical use of this technology; and
- 3. to establish quantitative changes in the properties of the fly ash concrete mixture and improve its physical and technical characteristics and durability of the fly ash concrete with the addition of polycondensation products of acetone and formaldehyde.

2. Theoretical overview

Since it is costly to use pure polycondensates, the addition of production waste (ASW with fly ash produced in China) can help reduce the cost of building materials' production technology. In China, such a solution is reasonable and finds motivation in both economic and in scientific terms. In order to define the possibility of its use as an additive in concrete, it is necessary to determine the content of harmful components in ash, which can be stated as follows: sulfur content (defined in this research); unburned particles of coke and semicoke coal determined by weight loss on ignition; free calcium and magnesium oxide; alkali metal oxides. They worsen the physical and mechanical properties of concrete and its technical and performance indicators and sometimes lead to concrete deterioration [\[11\]](#page-16-11).

The main properties of fly ash are chemical and phase-mineral composition, relative and volume weight, grain size composition, and specific surface area. Fly ash has a diverse chemical composition, as presented in Table [1.](#page-2-0) Table [1](#page-2-0) does not include other components (oxides) of fly ash since their content does not significantly change its characteristics.

The authors use low-calcium (CaO $<$ 20%) fly ash from coal combustion since Trykoz et al. [\[13\]](#page-16-12) note that this is a preferable practice when it comes to fly ash utilization in concrete. The used ash had a mixed chemical composition, as indicated in Table [1,](#page-2-0) after the combustion of anthracite and coal. These ashes have less active hydraulics than high-calcium ashes, their chemical and grain compositions are relatively more stable and uniform, and they do not adversely affect cement soundness [\[14\]](#page-16-13). Studies on the shrinkage of ash concrete showed that the introduction of ash reduces volumetric changes in concrete, and so the shrinkage of ash concrete (30% ash content) was 10%–20% lower than the control and did not depend on the quality of the concrete. Lower water-binding ratio and decreased heat release of ash concrete compared to concrete on pure Portland cement lead to a decrease in shrinkage [\[15\]](#page-17-0). Sokolenko et al. [\[16\]](#page-17-1) highlight that the use of an optimum dose of ash (150–200 kg) and plasticizing additive makes it possible to save up to 90–120 kg of cement per 1 m^3 of concrete, and the introduction of ash also serves as a sand substitute in construction works involving heat-insulation (e.g., when there is a need for heat-insulating concrete).

Studies conducted in Japan have found that ash used in concrete (especially in large quantities) reduces frost resistance and durability [\[17\]](#page-17-2). The low frost resistance and durability are due to the reduced content of entrained air (by 1.5–3 times), depending on the carbon content in the ash [\[18\]](#page-17-3). It proves the efficiency of using ash in concrete under complex plastification. Japanese researchers have reported decreases in the density of ash concrete and, accordingly, in water resistance [\[19\]](#page-17-4). An increase in water absorption by 1.5%–4% compared to traditional cement concrete confirms the deterioration of the ash-concrete structure. Thus, the use of ash mortars and concrete can be rendered more effective by introducing complex additives, which have an air-absorbing and plasticizing action and accelerate concrete hardening.

The analysis of the research results and the practice of using additives shows that complex additives are the most convenient to use since they can influence several characteristics of concrete at once and are often not related to each other. They also significantly strengthen and deepen any maximum effect of a single-component additive $[20]$. The introduction of complex additives can reduce or eliminate the undesirable side effects of each component of a complex additive. For example, the use of plasticizing additives for concrete mixtures, introduced at one-tenth of a percent, simultaneously slows down the binder hydration and, in turn, adversely affects the workability of precast concrete [\[21\]](#page-17-6).

It is possible to eliminate this deficiency by using an electrolyte and additive for plasticizing a concrete mixture. This additive also eliminates an undesirable side effect of an electrolyte – increased concrete hygroscopicity [\[22\]](#page-17-7). Another advantage of complex additives is the possibility of achieving their more universal action, which means almost absolute independence of the obtained effect from a chemical and mineralogical composition of cement and partial independence from concrete mixture composition [\[23\]](#page-17-8).

There are two categories of additives. The first one includes additives that consist of two or more additives of the same class. The second one involves combinations of additives of different classes. Studies on the effect of additives that belong to the first category have shown that their introduction makes it possible to change the intensity of structure formation and dispersion of newly formed structures. When introducing such additives, it is necessary to remember that their acceleration, in some cases, can be less than expected under the additivity principle [\[24\]](#page-17-9).

The authors of this article conducted the literature overview on the combustion products used to design the performance properties of building materials and found that the synergy of components was largely found to prevail in the literature. Thus, combustion products (slags and ash) are used in combination with modifying components (additives of various kinds, including derivatives of the combustion products themselves).

3. Materials and methods

In the course of experimental work, the authors used cement, ash, sand, and crushed stone in combination with acetone-formaldehyde resins in order to obtain and study the ash-concrete mixture. The authors used cement brands 400 and 500 as binders. Notably, when indicating the cement brands 400 or 500, the authors imply the brand of Portland cement (since cement used in classical construction mainly contains cement clinker in the alite phase and does not have a narrower name). These cement brands have almost the same chemical composition, and their main characteristics are as follows:

- normal density of cement paste -26.0% ;
- initial setting time -1 h 30 min;
- final setting time -7 h 35 min;
- body spreading 112 mm;
- relative weight -3.20 g/cm³;
- volume weight -1.20 g/cm³.

Hardening age (days)					28	
Brand	Compression	Bending	Compression	Bending	Compression	Bending
	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)	(MPa)
500	20.5	⊥.∠	27.2	6.0	50.2	6.1
400	15.9	3.0	24.2	4.8	39.8	52 ے ۔ ب

Table 2. Cement activity

Table 3. Compositions of heavy concretes

Concrete grade				
Brand	Cement (g/cm^3)	Sand (g/cm^3)	Crushed stone $(g/cm3)$	Water (g/cm^3)
200	280	630	1,300	150
300	370	610	.250	175
400	430	580	1.160	200

The authors present necessary data on the difference in cement activity between brands 400 and 500 in Table [2;](#page-4-0) other criteria are not affected by the difference in brands.

The cement contains an active mineral additive, a phosphorus slag in a quantity of 5%–15%. The authors selected compositions of heavy concrete of brands 200, 300, and 400 according to modern methods (Table [3\)](#page-4-1).

In this research, for the first time, the authors have studied how the increased content of ashes affects the structure formation of the ash-cement stone in the presence of the acetone-formaldehyde resins. The authors changed ash content from 0% to 40% and acetone-formaldehyde resin content from 0.1% to 0.2% of the cement weight on a dry basis. They were introduced into batched water when preparing a concrete mixture, and the ash was used under normal conditions in a binder-aggregate mixture using traditional concrete technology.

For physical and chemical analysis, the authors prepared samples of cement and ash-cement stone having a size of 2 cm \times 2 cm \times 2 cm and with a water-cement ratio (W/C) of 0.4. After manufacturing, these samples retained in the mold for a day under air-humid conditions, and then, after molding, the samples were stored in water. After a curing period, the samples were dried at 80◦C for 4 h and crushed to pass through a screen of 008. The samples were dehydrated with absolute ethanol. The obtained powder was filtered 1 day later and dried to a constant weight. The test tube with the

ground sample was covered with a wax plug and stored in the desiccator above calcium chloride for further physical and chemical research. The temperature of the furnace rose at a rate of $6°C/min$.

The authors determined the normal density, setting time, and strength of the samples of ashcement stone in accordance with the guidelines for designing protection against corrosion of concrete and reinforced concrete constructions [\[25\]](#page-17-10). The change in the strength of the samples was determined by testing after 1 day, 3 days, 7 days, and 28 days of curing. The kinetics of structure formation of cement paste during the setting time was determined using the Rebinder's conical plastometer. Since the initial kinetics of structure formation of cement mortars significantly depends on batch time, the preparation of the ash-cement paste took 6 min. The authors then examined the effect of ash and acetone-formaldehyde resin additives on the workability of the concrete mixture and the strength of the concrete samples. The authors determined the initial concrete mixture mobility by the slump of the reference cone. The kinetics of the cement workability over time was measured at normal and elevated temperatures (40° C– 45° C) of the mixture and the environment.

Test determination of the deformation properties of ash concrete was carried out at samples of prisms of 100 mm \times 100 mm \times 400 mm 3 days, 7 days, and 28 days after. Deformations were determined under short-term loading. Total deformation was defined as the sum of elastic and plastic

		Content of the components of the cement paste $(\%)$	Normal density of the cement paste $(g/cm3)$	Setting time (h:min)		
Cement	Ash	Acetone-formaldehyde resins		Start	Finish	
100	$\overline{}$		27.5	1:30	7:15	
100	$\overline{}$	0.15	23.5	2:15	6:00	
90	10		28.0	2:00	8:25	
90	10	0.15	26.0	2:40	6:45	
80	20		29.0	2:40	8:45	
80	20	0.15	27.0	2:55	7:00	
70	30		30.0	2:55	8:55	
70	30	0.15	27.5	3:15	7:20	
60	40		30.5	3:20	9:15	
60	40	0.15	28.0	3:35	7:35	

Table 4. Normal density and setting time of cement paste

components. The authors selected a stepped type of loading of the concrete sample. The deformations that occur at the time of application of the load are elastic, and the deformations that occur during the holding period under the load are plastic. During the static testing of concrete, the authors used dial indicators to determine its deformation characteristics for short-term loading. The modulus of elasticity of concrete samples was determined at the age of 14 days and 28 days.

Water absorption of ash concrete with the addition of acetone-formaldehyde resin was determined in accordance with current standards. The samples were placed in a vessel with water so that water covered them with a layer of 5 cm in height. Weight measurements were taken on a conventional balance every day until the weight gain was less than 0.1% of the initial weight. The saturation of samples with water was after their drying.

The water resistance of ash concrete with the addition of acetone-formaldehyde resin was determined according to the current standards. The authors made samples-cylinders with $D = H =$ 150 mm, kept them in a moisture chamber, and preserved them for a day in the air before testing. Molten paraffin filled the gap between the samples and the mold. The ends of the samples were cleaned with a steel brush. The prepared samples were placed in a test apparatus, which brought the sample to its lower surface at a specified iodine pressure. Observing the upper surface, the authors recorded the moment when water began to seep through the concrete. The tests began at a pressure of 0.1 MPa, and then the pressure increased by 0.1 MPa every 8 h. The authors determined water absorption of the concrete by the pressure at which no water seepage through the concrete was yet detected on the surface of four of the six samples.

The frost resistance of ash concrete with the addition of acetone-formaldehyde resin was determined according to the current standards. The samples were frozen in a freezer at temperatures below 15◦C–20◦C for 4 h in a water-saturated state. The samples were thawed in water at a temperature of 15° C–20 $^{\circ}$ C for 4 h. The distance between the cubes was 5 cm during freezing and thawing. The number of cycles depended on the brand of concrete used, and it was determined based on whether that particular brand of concrete was designed with the capability for providing significant frost resistance.

According to modern concepts, composite materials should include materials based on inorganic binders and mineral fillers characterized by an inhomogeneous structure with evident phase separation [\[20\]](#page-17-5). In accordance with the theory of polystructures, it is necessary to study the structure of such materials at two levels – the microstructure and macrostructure. The first results from the curing of a mixture of a binder with mineral and other modifying additives. The second is a cured mixture of microstructures and coarse aggregate of the appropriate density [\[26\]](#page-17-11).

		Content of the components of the cement paste	Strength of a sample of cement stone,					
$(\%)$			MPa in a day					
Cement	Ash	Acetone-formaldehyde resins		3		28		
100			31	52	72	78		
100		0.15	35	65	83	88		
90	10		25	44	70	75		
90	10	0.15	33	53	78	80		
80	20		22	43	60	64		
80	20	0.15	31	52	72	79		
70	30		20	37	49	60		
70	30	0.15	29	47	67	76		
60	40		18	29	39	50		
60	40	0.15	25	42	54	62		

Table 5. Strength gained with time

4. Results

There were two stages in developing the composition of ash-cement concrete. First, the authors determined the effect of ash content and the addition of acetone-formaldehyde resin on the properties of the microstructure, normal density, setting time, and structure formation of cement stone in the initial and subsequent curing periods. These results are presented in Table [4.](#page-5-0)

As noted above, the introduction of ash in the increased amount significantly slows down the structure formation of cement stone, and the obtained data on the setting time of cement paste also confirm this. Therefore, the authors studied the kinetics of the structure formation of cement stone in the initial curing periods according to the change of plastic strength during the setting time of cement paste of the normal density. Partial replacement of the cement with ash leads to a proportional slowdown in the structure formation, as evidenced by the plastograms of the structure formation of cement stone in the first 6–9 h of curing at normal temperature. An increase in the percentage of ash in the binder extends the so-called induction period when there is no formation of structural bonds in the cement paste [\[27\]](#page-17-12). Osman and Al-Masry [\[28\]](#page-17-13) believe that the cement paste has thixotropic properties, retains its plasticity, and does not set during this period.

The rate of structure formation of cement stone decreases with the increase of ash additive since the dilution of cement with finely dispersed additive affects the structure formation and the kinetics of clinker hydration in the binder. Acetoneformaldehyde resin introduced to the cement paste can both slow down and speed up the structure formation [\[28\]](#page-17-13). The study shows the concentration of the additive in the cement paste determines its effect. At a low dosage (0.05% by weight of cement), the addition of acetone-formaldehyde resin slightly increases the induction period without changing the rate of structure formation. An increase in the dosage of the additive to 0.15% decreases the equal plasticity of the paste, extends the induction period, and accelerates the structure formation in the first hours after setting.

An increase in the dosage rate of acetoneformaldehyde resin to 0.3%, despite a further decrease, extends the induction period and decreases the rate of structure formation. Therefore, the authors investigated the structure formation of the ash-cement paste with the addition of 0.15% acetone-formaldehyde resin. If the resin content is optimum (0.15% of cement consumption), the replacement of the clinker part of cement with ash (up to 30%) does not reduce the rate of the structure formation, which should also have a positive effect on the strength of cement stone. The authors kept the samples of cement stone in water during the research. Table [5](#page-6-0) presents the results of the experiments in terms of the strength that the samples of cement stone gained with time.

As a result of testing, the deviation from the

Content		W/C	Mobility, cm (numerator)/cone spread, mm (denominator)							
of additive $(\%)$		$(\%)$								
Ash	SAFA		0 min	30 min	60 min	90 min	120	150	180	$\overline{210}$
							min	min	min	min
At normal temperature										
Reference		0.56	4.3/142	3.8/129	3.4/125	3.0/118	\blacksquare	$\overline{}$	$\overline{}$	$\overline{}$
mixture										
Without	0.15	0.56	5.5/160	5.2/155	4.7/148	4.3/140	3.9/132	3.5/125	$\overline{3.0/119}$	
additives										
20		0.56	5.2/155	5.1/152	5.0/150	4.8/147	4.2/140	3.8/132	3.5/125	3.0/120
30		0.56	4.8/149	4.6/147	4.5/145	4.2/142	3.8/136	3.4/129	3.0/122	
40		0.56	4.6/147	4.5/145	4.4/143	4.0/140	3.6/133	3.3/127	3.0/120	
20	0.15	0.56	6.8/170	6.7/168	6.6/165	6.3/163	6.0/160	5.5/154	5.0/149	4.2/140
30	0.15	0.56	6.3/167	6.1/165	5.9/163	5.6/160	5.3/156	5.0/150	4.4/143	3.8/135
40	0.15	0.56	5.6/165	5.5/163	5.3/161	5.0/158	4.7/155	4.2/147	3.8/142	3.2/134
Ash	SAFA	$\overline{\text{W}/\text{C}}$	0 min	15 min	30 min	45 min	60 min	75 min	90 min	
		$(\%)$								
At 50° C										
Without	0.15	0.58	4.0/140	3.5/128	3.0/121	\blacksquare	$\overline{}$	\blacksquare	\blacksquare	
additives										
20		0.58	5.0/155	4.5/150	4.0/145	3.5/130	3.0/125		$\overline{}$	
30		0.58	4.5/142	4.2/138	3.8/135	3.3/130		$\overline{}$		
40		0.58	4.2/140	3.8/135	3.3/132	3.0/128				
20	0.15	0.58	6.0/158	5.5/152	4.7/147	4.2/141	3.6/134	3.5/134	3.2/129	
30	0.15	0.58	5.6/156	5.1/150	4.7/145	4.2/137	3.6/134	3.2/128	2.9/122	
40	0.15	0.58	5.4/154	5.0/148	4.5/142	4.0/135	3.4/131	3.0/124		

Table 6. Indicators of the workability of the mortar mix

SAFA, Supplement acetone-formaldehyde additive.

Fig. 1. Calcium hydroxide binding process. SAFA, Supplement acetone-formaldehyde additive

shown result did not exceed 5% when considering five samples of each group. The decrease is proportional to the ash content, and the qualitative changes are linear. An increase in ash content from 10% to 40% decreases the strength indicators by 20%–41% at the initial stage decrease, and the grade strength drops by 4%–34%. The addition of acetone-formaldehyde resin and 30% ash content facilitates alignment during initial and subsequent curing periods. Thus, the combined use of ash and acetone-formaldehyde resin additives has a positive effect on the kinetics of the structure formation and strength of the cement stone both in the early and in the design periods. The authors studied the kinetics of cement hydration in the presence of increased content of the active mineral and the addition of acetone-formaldehyde resin in order to confirm the quantitative regularities of structure formation.

Ash does not significantly change the kinetics of the hydration of clinker minerals in its early stages. Pectous hydrated newly formed structures in cement are equalized (400◦C) by the 7th day of hydration, although the total loss by burning remains at a higher level in plain cement, and its

	Content of additive $(\%)$	W/C $(\%)$		Strength indicators (MPa)					
Ash	Acetone- formaldehyde additive		In 3 days hardening at 25° C		In 7 days hardening at 25° C		In 28 days hardening at 25° C		
			Bend	Compression	Bend	Compression	Bend	Compression	
	Reference composition	0.56	2.3	9.8	3.8	16	5	22	
Without	0.15	0.51	2.7	12	4.1	22	5.5	27.5	
additives									
20		0.54	1.3	8	3.2	14	6	20.5	
20	0.15	0.50	2.6	10	3.4	16.8	5.3	24	
30		0.56	1.2	6.5	3	12.8	4.8	19.5	
30	0.15	0.51	2.4	9.5	3.2	15.6	5.2	22.5	
40		0.56	0.8	5.4	2.6	9.6	5.8	18	
40	0.15	0.51	1.9	7	2.7	12.8	5	19.5	

Table 8. Results of tests on the strength of samples made of a mixture of normal temperature

evening out comes due by the age of 28 days. This is due to the interaction of ash with calcium hydroxide to form hydrated silicates, hydrated aluminates, and hydrated aluminum silicates [\[29\]](#page-17-14). It is possible that a change in the kinetics of crystallization of calcium hydrosulfoaluminate, which is a highly hydrated compound, can have a particular influence. The binding process of calcium hydroxide begins at an early age of cement hydration with the addition of ash and is noticeable throughout the study period (Figure [1\)](#page-7-0).

Content of additive $(\%)$		W/C	Strength indicators (MPa)						
		\mathscr{G}_o							
Ash	SAFA			In 3 days hardening		In 7 days hardening	In 28 days hardening		
			at 25° C		at 25° C		at 25° C		
			Bend	Compression	Bend	Compression	Bend	Compression	
Reference composition		0.58	1.8	8.8	2.4	13	3.9	19.2	
Without additives	0.15	0.52	2	10.2	2.8	15.9	4.4	24.2	
20		0.57	1.3	7	1.8	11.8	3.3	18	
20	0.15	0.51	1.5	8	2.2	13.5	3.8	22	
30		0.58	1.0	5.7	1.6	10	3	17	
30	0.15	0.53	1.3	7.2	2	11.5	3.2	20.5	
40		0.58	0.8	5.2	1.3	6.8	2.8	15	
40	0.15	0.53	1	5.3	1.7	8.7	3	18	

Table 9. Determination of the strength of samples made of a mixture of normal temperature

SAFA: Supplement acetone-formaldehyde additive.

Table 10. Results of tests on the strength of samples made of a mixture of an elevated temperature

	W/C Content of additive $(\%)$		Strength indicators, MPa, in a day hardening at 50° C						
		\mathscr{G}_o							
Ash	SAFA			In 3 days hardening		In 7 days hardening	In 28 days hardening		
			at 25° C		at 25° C		at 25° C		
			Bend	Compression	Bend	Compression	Bend	Compression	
Reference composition		0.58	2.1	11	4	17	5.2	23.5	
Without additives	0.15	0.53	3.4	13.2	4.8	18.8	6	30.5	
20		0.56	1.9	9	3.5	15.8	5	21.5	
20	0.15	0.52	3.4	12.8	4.2	19	5.6	26.5	
30		0.57	1.7	8	3.2	14	4.9	20	
30	0.15	0.53	3	10	3.6	16.8	5.4	24	
40		0.57	1.5	5	2.8	13.5	4.9	18.5	
40	0.15	0.53	2.4	8	3	14.5	5.2	20.5	

SAFA: Supplement acetone-formaldehyde additive.

The authors determined the total CaO content according to step responses at 440◦C–510◦C and 740 $°C$ –780 $°C$ peculiar to Ca(OH)₂ and CaCO₃, respectively. Within 28 days of hydration, the binding process occurs between 30% of ash and about 8% of CaO (given the ratio of cement components). Thus, most ash has already reacted with calcium hydroxide during this period. Less carbonization of cement stone with ash added is noticeable. Thus, this additive proves to increase the density of the cement structure. The introduction of a plasticizing additive of acetone-formaldehyde resin that reduces the water demand for cement plays a particular role [\[30\]](#page-17-15).

During the experiments, the authors studied the influence of ash and acetone-formaldehyde resins on the mortar workability over time at the same water content and for mixtures of equal mobility. In order to obtain objective indicators of the effects of addition of ash and acetone-formaldehyde resins, the workability of the mortar mix was assessed based on the mobility and spreading of the cone. In this case, the authors tested the mortar mix until it spread evenly over coarse-grained sand (Table 6).

Table [6](#page-7-1) shows that the highest workability at normal temperature is possible with 20% ash content, while a further increase evens out the mobility and spreading of the cone. Notably, the introduction of ash significantly improves the spread rate of the mixture due to the rounded and vitrified surface of the particles of the mineral additive. The spread

Ash content	Content of acetone-formaldehyde additive $(\%)$			Compressive strength
in concrete mix $(\%)$				of concrete brands (MPa)
		200	300	400
θ	Ω	20.3	30.2	40.5
	0.1	22.5	34.9	47.5
	0.15	24.8	36.5	50.2
	0.2	21	30	39.6
20	Ω	15.9	24.5	32.5
	0.1	17.8	28.7	38.1
	0.15	19.2	31	40
	0.2	14.7	22.7	31.7
30	Ω	14.5	22	30
	0.1	16.5	25.6	35.7
	0.15	17.9	27.5	38
	0.2	13.1	20.7	29.5
40	Ω	12.5	18.3	24.5
	0.1	13.7	19.8	28.7
	0.15	14.5	21	30.2
	0.2	11.7	17.8	23.5

Table 11. Strength of ash concrete

Table 12. The experimental results

	Concrete brand Strength indicators of concrete, ordinary concrete (numerator)							
		and ash concrete (denominator)						
		After steaming Over 28 days of curing						
200	14.8/12.6	19.6/19.2						
300	22.4/19.4	29.8/29						
400	28.1/26.5	39.5/39.1						

Table 13. The results of the determination of water resistance

Note: The letter B stands for compression grade of concrete.

rate of the reference mixture lasts for 90 min. If there is an increase in ash from 20% to 40%, the workability lasts for 180–210 min. The reasons are the extension of the induction period of the structure formation and the water-retaining capacity of

the ash.

The addition of acetone-formaldehyde resins more significantly increases (by 23–28 mm) the spread rate of the ash-cement mixture, and the workability remains almost at the level of the ref-

erence mixture even after 210 min from the start of preparation. The plasticizing air-absorbing effect of acetone-formaldehyde resin is even more evident in the workability of the heated mortar mix [\[26\]](#page-17-11). For example, the workability of the reference mixture lasts only 30 min. If there is an addition of ash, it lasts 45 min, and acetone-formaldehyde resins increase the duration of mobility by 2.5–3.

In order to determine the maximum content of ash that could be added to a cement–sand mortar concomitant with the addition of acetoneformaldehyde resins in the amount of 0.15%, together with retaining the properties of the resultant samples at acceptable levels, the authors examined the change in compressive and bending strength of samples with a mineral additive in the amount of 20%–40%, selected equally from slump mixtures at 25◦C and 50◦C, and varying relationally with the size of the sand particles. (Table [7\)](#page-8-0).

Tables [8](#page-8-1) and [9](#page-9-0) show the results of tests on the strength of the samples made of a mixture of normal temperature, and Table [10](#page-9-1) presents results of the same tests but for samples made of a mixture of an elevated temperature.

The analysis of the obtained data shows that the combined use of ash and acetone-formaldehyde resins positively affects the strength of the mortar on both ordinary and fine-grained sand. For example, 20%–40% ash content reduced the strength by 10%–20%, and it dropped by 40% with the introduction of 0.15% acetone-formaldehyde and 30% ash content into the reference mixture. Notably, mineral and chemical additives significantly increase the bending strength of the mortar samples due to the increased contact density and strength. The kinetics of changes in the strength of mortar samples over time is similar to the previously obtained data on the strength of the ash-cement stone. The qualitative changes in the strength of samples cured at normal and elevated temperatures are the same, while quantitative indicators differ due to the intensifying effects of the curing conditions. The experimental studies involved examining the effects of ash additives (20%–40%) and acetoneformaldehyde resins $(0.1\% - 0.2\%)$ on the strength of ash concrete (Table [11\)](#page-10-0).

The introduction of acetone-formaldehyde resin

into the ash-concrete mixture increases the strength of the concrete, which is due to the deflating and dispersing effect of this additive on the grains of cement and cement stone. The result is the increased density and strength of concrete. Thus, the combined use of mineral and chemical additives makes it possible to replace 30% of cement without decreasing the strength of concrete.

Given the multifunctional effect of the addition of acetone-formaldehyde resins, the authors tested and studied the strength of ash concrete depending on the water demand at an ash content of 30% and at an acetone-formaldehyde resin content of 0.15%. The experiments have shown that the reference mixture can obtain the same mobility index if the water content of the ash-concrete mixture is reduced by 16%, 10%, and 11% for cement brands 200, 300, and 400, respectively. Reducing the water demand within the specified limits made it possible to obtain an additional increase in the strength of concrete, ensuring the achievement of design indicators of the reference samples and increasing the uniformity of the concrete structure.

Lyashenko et al. [\[31\]](#page-17-16) state that the plastification of the concrete mix can adversely affect the strength of the steamed concrete and lengthen the steam curing period of precast concrete and ash-concrete products. As noted earlier, although the combined use of ash (30%) and acetoneformaldehyde resins (0.15%) slows down the structure formation during the induction period, it does not reduce the strength of the cement stone in the initial curing period. This suggests that the strength indicators will correspond to the calculated data if there is steam curing of ash-concrete with the addition of acetone-formaldehyde resins. Thus, the authors carried out tests on steam curing of ash-cement samples of brands 200, 300, and 400 with 0.15% acetone-formaldehyde resins. During the tests, the authors adopted the standard mode for steam curing of concrete: $2 + 3 + 6 + 2$ h at 80 $^{\circ}$ C and 13 h of total duration (Table [12\)](#page-10-1).

Table [12](#page-10-1) shows that the strength of steamed samples of ordinary concrete is $71\% - 75\%$ and that of ash concrete 65%–68%, which practically meets the regulatory requirements. Thus, the need for the introduction of high ash content and for the addition of acetone-formaldehyde resins in the production of precast concrete products is substantiated. Since data on the influence of plasticizing additives on the deformation of concrete and the initial elastic module were contradictory, it was interesting to study the effect of acetone-formaldehyde resins on the deformation of ash concrete. The research focused on the concrete brand 300 (reference mixture) with 30% ash and 0.15% acetoneformaldehyde resins.

The experiments involved determining the kinetics of plastic, elastic, and total deformations of concrete under short-term loading. When 30% ash is introduced into concrete, this significantly reduces deformation properties and increases plastic, elastic, and total deformations. The introduction of 0.15% acetone-formaldehyde resins into ash concrete helps to reduce deformations both at 7 days and 28 days of age, and these results are largely consistent with those observed in the case of the reference concrete samples. The authors examined water resistance, swelling, and frost resistance in order to establish the effect of the increased content of ash (20%–40%) and acetone-formaldehyde resins (0.15%) on the durability of concrete (Table [13\)](#page-10-2).

According to the data obtained, the water resistance of concrete without ash doubles with the introduction of an additive. Table [13](#page-10-2) demonstrates that the water resistance of ash concrete decreases. However, the optimum amount of additive introduced and correction of its composition in terms of water content make it possible to increase the water resistance grade by three times. This confirms a lower degree of carbonization and, consequently, a denser structure. Since chemical additives reduce the pores in concrete, and since the solid body surfaces bordering the pores are partially water-repellent, the water penetration into the material is slower, and the rate and degree of swelling are decreased compared with the reference sample of only-cement concrete. Studies have shown that when the ash content of concrete is 20%, 30%, and 40%, the degree of swelling increases by 7%, 15%, and 18%, respectively.

The introduction of 0.15% acetoneformaldehyde resins into the composition of ash concrete decreases the degree of swelling of the concrete with 30% ash content by 10%, 16%, and 20%, respectively, and the indicators reach the level of reference samples. Studies of the frost resistance of concrete with the addition of acetone-formaldehyde resins showed that concrete samples that had no additives withstood 150 cycles with a loss of strength of 5%–12%. The samples with acetone-formaldehyde resins withstood 300 cycles and lost 10%–15% in strength. It is known that the frost resistance of concrete increases with the decrease of the water–cement ratio [\[32\]](#page-17-17). These facts suggest that ash concrete can be of the required frost resistance grade if there is an optimum amount of additive: 0.15% acetone-formaldehyde resins by cement weight.

The main task in preparing concrete mixtures is to ensure the uniform distribution of the components during the mixing process, achieving the necessary homogeneity of the mixture. It is necessary to create the possibility of maximum wetting of the surface of individual cement grains with water and coating with cement paste. Simultaneously with the mixing of the components, multiple complex physical and chemical processes rapidly develop. The increased hydrophilicity of aluminate minerals, gluing clinker components of cement such as two-calcium and tricalcium silicate, ensures the active penetration of water into microc-racks of cement grains [\[26\]](#page-17-11). The resulting hydration causes the chemical dispersion of grains of polymineral cement. This process is facilitated by the phenomenon of adsorption weakening of cement grains as a result of surface wetting. The cement paste is enriched by colloidal particles as soon as these materials begin to come into contact with each other.

The smallest cement grains, following the laws of molecular attraction, tend to unite in floccules in the water. The distribution of water is far from uniform within and between individual floccules. A more uniform coagulation structure of the cement paste is possible with the increase in the uniformity of the water distribution in cement and the separation of the sticky grains of cement at mixing time. It ensures the best technological properties of the concrete mixture and higher quality of the hardened concrete [\[19\]](#page-17-4). Crystallization of hydrate embryos of newly formed structures and the dissolution of mineral clinkers coincide in time.

The hydrophilic surfaces of quartz sand grains are the most active substrate for the growth of newly formed crystals. The contact of sand grain surfaces with cement hydration products largely determines the strength of hardened concrete. The thinnest layers of dust and clay cover sand grains, which prevent the formation of solid contact layers. The area of the layers, which prevent the formation of contacts, narrows due to repeated collisions of particles caused by mixing the concrete mixture and the friction of their surfaces against each other in the water. It improves the physical and mechanical characteristics of hardened concrete.

Thus, the mixing process cannot be considered only as a mechanical connection between the content of its components in the volume of the finished material. This condition is not enough to obtain materials with reinforced properties. Mixing should provide optimum conditions for the physical and chemical processes involved in the formation of the technological properties of the concrete mixture and its physical and mechanical properties during hardening. If the mixing time is insufficient, the homogeneity of concrete deteriorates, the coefficient of variation increases, and strength decreases [\[33\]](#page-17-18). Increasing the mixing time of the concrete mixture can lead to its segregation. All results were obtained by testing five forms of each sample. Since the scatter of readings (error) did not exceed 5%, the authors applied average values for informational content. Based on this, the work investigated the effect of the additive on the duration of mixing, uniformity and mobility of the concrete mixture, and the compressive strength of concrete samples.

5. Discussion

The analysis of the research results and the practice of using additives shows that complex additives are the most convenient to use since they can influence several characteristics of concrete at once and are often not related to each other. They also significantly strengthen and deepen any maximum effect of a single-component additive. The introduction of complex additives can reduce or eliminate the undesirable side effects of each component of a complex additive. For example, the use of plasticizing additives for concrete mixtures, introduced at one-tenth of a percent, simultaneously slows down the binder hydration and, in turn, adversely affects the workability of precast concrete [\[21\]](#page-17-6).

It is possible to eliminate this deficiency by using an electrolyte and additive for plasticizing a concrete mixture. This additive also eliminates an undesirable side effect of an electrolyte – increased concrete hygroscopicity. Another advantage of complex additives is the possibility of achieving their more universal action, which means almost absolute independence of the obtained effect from a chemical and mineralogical composition of cement and partial independence from concrete mixture composition [\[30\]](#page-17-15). There are two categories of additives. The first one includes additives that consist of two or more additives of the same class. The second one involves combinations of additives of different classes [\[22\]](#page-17-7). Studies on the effect of additives that belong to the first category have shown that their introduction makes it possible to change the intensity of structure formation and dispersion of newly formed structures. When introducing such additives, it is necessary to remember that their acceleration, in some cases, can be less than expected under the additivity principle [\[34\]](#page-17-19).

It is possible to achieve the plasticizing effect at the optimum dosage of additives, and then electrolytes can provide a sufficiently high rate of concrete hardening. Additives for plasticizing a concrete mixture are ineffective when high-alumina cement is used as a plasticizer, but gypsum additives can restore their plasticizing effect [\[35\]](#page-17-20). In addition, the introduction of plasticizing and electrolyte additives into concrete mixtures improves the physical–mechanical properties of cement stone, and more complex compositions consisting of several electrolytes and additives act in the same way $[36]$.

Experimental and theoretical studies on the effect of complex additives on the properties of individual clinker materials and various types of cement made it possible to determine the role and significance of additives in the structure formation of concrete mixes and hardened concretes. Given the specific properties of thin layers of liquid located between solid dispersed particles in cement systems, it is possible to attribute the cement paste to a plastic and viscous body.

It is possible to purposely influence the structure of cement stones in concrete with the help of complex additives containing a plasticizing or airentraining additive in addition to the electrolyte. Since the electrolyte affects the submicroscopic level of the structure of cement stone, and the plasticizing or air-entraining additives influence the micro- and macro-levels of the structure, their combination makes it possible to obtain a denser, highly dispersed cement stone while maintaining the nature of the distribution of air bubbles in it. As a result, it is practically possible to increase the frost resistance of concrete [\[24\]](#page-17-9).

Numerous studies of properties of concretes with complex additives show that additives consisting of plasticizers and hardening accelerators are most effective for hardening concrete under normal conditions and during steaming. A complex additive consisting of a preservative and a hardening accelerator additive, calcium chloride with sodium nitrate, has the following effects:

- it retains the mobility of the concrete mixture;
- it increases the strength characteristics of monolithic concrete;
- calcium chloride accelerates hardening and increases the concrete strength in the initial curing stages;
- sodium nitrite plays the role of a corrosion inhibitor [\[6\]](#page-16-5).

Studies showed that a complex additive based on green liquor, introduced into the concrete mixture in the amount of 0.05% of the cement weight, could increase the frost resistance of concrete by 2.5–3 times compared to the composition without additives. It could also double the workability of concrete mixtures and reduce their water demand. The combination of black sulfate liquor with a superplasticizer is the most effective for manufacturing densely reinforced structures [\[37\]](#page-18-1). The introduction of a complex additive consisting of calcium chloride, zinc chloride, and phthalic acid increases the strength of concrete by 20%–40%.

The study established that the introduction of 20%–40% ash into concrete increased the mobility and strength of concrete under the condition of extended mixing. The introduction of 0.15% acetoneformaldehyde resins into ash concretes with an optimal ash content (30%) makes it possible to reduce the mixing time by an average of 1.5–2 times without changing the grade strength. This is due to the additive, which increases the homogeneity of the ash-concrete mixture and improves its thixotropic properties.

The authors introduced the additive of acetoneformaldehyde resins into the concrete mixture in the amount of 0.15% of the weight of the binder. First, the authors adapted the composition to the water content until they obtained the required mobility, and made structures from a concrete mixture of the adjusted composition with an additive. Pursuant to the introduction of additives, the mobility of the concrete mixtures containing cement brands 200 and 300 increased by 1.5 and 1.8 times, respectively. The strength of ash concrete with the optimum amount (30%) of ash and additives (0.15%) had the same indicators as the reference composition while testing reference samples. When correcting the composition, the authors determined the quanta of consumption of components of the concrete mixture having a 30% ash content and a 0.15% additive content. Results of the reference samples manufactured at the test site confirmed the data of the mentioned laboratory studies.

The ash-concrete mixture was prepared in a concrete mixer with a capacity of 700 L. The authors supplied cement and ash from the bunkers and added the additive with a dosing device into the mixing water. The experimental work was carried out at a temperature of 14◦C–16◦C and a humidity of 75%–80%. The experiments showed that the compressive strength of samples of ash concrete and the reference composition of brand 300 increased after 1 day, 2 days, and 4 days, amounting to 16–18%, 24–28%, 46–48%, respectively, of the calculated value. Since the stripping strength of concrete should be at least 50% of the grade strength, the curing time before stripping should be 4–5 days when using ash concrete with the addition of acetone-formaldehyde resins for monolithic structures. The study also showed that it would be possible to reduce the duration of the masonry and compaction processes by 30% when concreting monolithic foundations.

In the context of manufacturing monolithic bridge pillars, the question of the duration of hardening of ash concrete before crumbling is of interest. For this purpose, the authors performed tests to determine the strength of ash concrete in the initial curing periods after 1 day, 2 days, and 4 days after manufacturing the samples. It is possible to regulate the technological properties of the concrete mixture and the characteristics of the ready mixed concrete if there is an introduction of new complex polymer additives. These additives are polyconjugated oligomeric hydroxylated substances of aromatic nature, containing 4–12 elementary links. The use of complex additives with a high content of lignosulfonates and electrolytes makes it possible to increase the durability of reinforced concrete structures by 3–5 times. These additives used in concrete mixtures of equal mobility increase the concrete strength by 30%–40% and thus help to save cement.

6. Conclusion

Increased dispersion of cement stone structure with complex additive causes the adsorption modification of hydration products of cement systems, which leads to an increase in the strength of hardened cement systems and improves their properties of resistance to deterioration when subject to water and freezing exposures. Complex additives allow the following:

- to increase the mobility of the concrete mixture and thus save 12%–15% cement;
- to increase the productivity of processing lines by 20%–40% due to the acceleration of concrete hardening; and

• to decrease fuel costs and increase the durability of the product by reducing the isothermal time or the steaming temperature by 15° C -20° C.

The introduction of a complex additive consisting of two or more components into the concrete mixture is quite difficult to implement in production conditions. Partial replacement of cement with ash content of up to 30% reduces the rate of early structure formation in cement stone samples and improves the kinetics of hydration of clinker minerals. However, the increased ash content raises the water demand of the cement test by 13% and reduces strength under the initial and design conditions by 20%–41% and 4%–34%, respectively. The interaction of ash with calcium hydroxide causes a decrease in the concentration of $Ca(OH)_2$ in the cement stone, acceleration of crystallization of hydrosulfoaluminate, and an increase in the content of pectous hydrated newly formed structures (hydrated silicates, hydrated aluminates, and hydrated aluminum silicates).

The use of acetone-formaldehyde resin in an optimum amount compensates for water consumption and evens out the strength characteristics of cement stone under both the initial and design conditions. The combined use of ash and additives of acetone-formaldehyde resins contributes to the formation of an additional amount of hydrates and compaction of the cement stone, reduces the carbonization rate, and thus increases the durability of concrete. The authors established the following in an experimental study:

• The addition of the acetone-formaldehyde resin positively influences the technological and strength properties of the cement–sand mortar with ash content of up to 30%, and this holds true with sand of standard and fine fractions, and under the normal and elevated temperatures of the mixture's formative conditions. The plasticizing effect is evident since the water demand of the mixture decreases by 10%–12% under the same mobility, and the durability grows by 1.5–2 times.

- The use of the additive of acetoneformaldehyde resins in an optimum amount (0.15%) makes it possible to replace up to 30% of cement with ash without reducing the strength characteristics of the ashcement mortar.
- The use of acetone-formaldehyde resins has a positive effect on the structure formation of the ash-concrete mortar and provides the required strength indicators. In this case, the contact zone of the binder and filler is characterized by a more homogeneous and finely dispersed structure with simultaneous modification of plate crystals, probably $Ca(OH)_2$, into cubic form.
- The strength of concrete decreases linearly with an increase in the ash content from 20% to 40%. The introduction of the additive of acetone-formaldehyde resins ensures the replacement of up to 30% of cement with ash and evens out the strength of ash concrete within the estimated time during hardening both under normal conditions and steam curing.

The combined plasticizing effect of mineral and chemical additives is characterized by an average two-fold increase in the mobility of the concrete mixture and a decrease in water demand for obtaining mixtures of equal mobility of brands 200, 300, and 400 by 16%, 10%, and 11%, respectively. The addition of acetone-formaldehyde resins positively affects the deformation properties, water resistance, and frost resistance.

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