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Article

# Experimental Study of a Composite Modifying Additive Based on Industrial By-Products for Enhancing Durability of Portland Cement Concrete

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## Abstract

This article presents the results of tests evaluating the physical and mechanical properties of a modified hydraulic concrete formulation based on Portland cement, intended for use in general construction. The additive consists of post-alcohol distiller's grains (PaB), soapstock (Sp), caustic soda (NaOH), granite dust (Gr) and acrylic latex (Lx). These components contribute to transforming the strength characteristics of concrete in compression and bending, as well as its water absorption, water permeability and chemical resistance. Based on the results obtained, the effectiveness of the additive was assessed, as was the quantitative improvement in concrete properties, including an evaluation of the life cycle of reinforced concrete structures in aggressive environments. According to the research results, an optimal composition was obtained which increases compressive strength by 6.2%, flexural strength by 7.9%, decreases water absorption by 50.1%, decreases the filtration coefficient by 97.4%, and increases chemical resistance by 42.8%.

**Keywords:** hydraulic concrete; Portland cement; modifying additive; granite dust; soapstock; acrylic latex; caustic soda; water absorption; chemical erosion; durability; life cycle prediction

## 1. Introduction

The durability of concrete depends on many factors related to its operating conditions, including the chemical activity of the surrounding groundwater [1]. Previous studies in this area have focused on enhancing the corrosion resistance and durability of concrete through altering the binder composition and incorporating various mineral and polymer additives [2]. The destructive effect of aggressive environments on reinforced concrete structures significantly reduces the life cycle of buildings and is a major problem for their safe operation [3]. Aggressive environments affecting concrete and reinforced concrete structures can be classified according to several characteristics, including the degree of aggressive impact (non-aggressive, weakly aggressive, moderately aggressive or strongly aggressive), physical state (gaseous, liquid or solid) and chemical composition (organic or inorganic; sulphates; chlorides; acids; magnesium salts; alkalis; salts that crystallise with a large increase in volume; biologically active substances; and products of bacterial, fungal and fungal hyphal metabolism and plant root activity [4]).

The most common aggressive environments found in soil conditions are sulphate environments, chloride environments and acidic environments [5].

In a sulphate environment, gypsum and hydrosulfoaluminate compounds are formed within the concrete structure. This leads to internal stresses that can exceed the concrete's strength [6]. While reducing permeability offers some protection against the effects of sulphur oxide ions, the most

effective solution is to use sulphate-resistant cement, which contains low levels of tricalcium silicate and aluminates (up to 5%).

In a chloride environment, as it is chemically aggressive, not only concrete is susceptible to destruction, but above all reinforcement. When chloride penetrates concrete, it accelerates the depassivation and corrosion of steel reinforcement [7]. The main protective measures against chloride environments are reducing the permeability of concrete and applying protective solutions to reduce penetration by aggressive substances [8].

In acidic environments, the alkaline minerals in concrete react with acids to form calcium salts. The rate at which these salts form, and therefore the negative effect of the acids, depends largely on the acid concentration and the solubility of the calcium salts. The most common acidic environments include sulphuric and hydrochloric acid. The main measures of protection against such environments are reducing the permeability of concrete and providing insulating protection [9].

It should be noted that fresh environments can also be aggressive towards pozzolan cement. The solubility of calcium hydroxide in such environments leads to the destruction of silicates and aluminates, resulting in reduced permeability and, ultimately, reduced concrete strength [10].

Thus, the main reason for the destruction of concrete in the contact zone with groundwater is the destruction of the cement matrix of concrete, as a result of exposure to aggressive environments. This is due to the formation of calcium hydrosulfoaluminate and calcium salts, as well as the direct leaching of lime from the concrete. As the cement matrix weakens, the concrete's strength decreases and its water permeability increases. This leads to corrosion and the destruction of the steel reinforcement in reinforced concrete structures [11].

It has been proven that introducing mineral additives based on granite dust and acrylic latex improves concrete's chemical resistance in acidic and sulphate environments [12].

However, most existing studies focus on either concrete made with special types of cement or the use of high-tech composite additives. This limits their practical application in mass construction [13].

The above-mentioned problem of reinforced concrete structures in aggressive environments was the driving force behind this study, which aimed to determine the feasibility of using Portland cement-based concrete for general construction purposes in aggressive sulphate environments [14].

The main method of increasing the chemical resistance of concrete was to use a modifying additive that reduced its water permeability and increased its water-repellent properties [15]. This was achieved by increasing the density of the concrete to reduce water permeability, and by polymerising the cement matrix to increase water repellency.

## 2. Materials and Methods

The proposed additive modification comprises the following components: granite dust (GR), post-spirit distiller's grains (PaB), soapstock (Sp), caustic soda (NaOH) and acrylic latex (Lx).

Granite dust is a finely dispersed medium. Including it in the composition of the additive reduces concrete's water absorption capacity and water permeability by increasing its density. Distillers' grains are a waste product of alcohol production and are rich in organic substances. They are essentially surface-active substances (surfactants). In the proposed additive, they act as a modifier that affects the plasticity and moisture retention capacity of the concrete mixture. Using post-alcohol distillers' grains in the additive reduces its volume and gives the additive new properties, such as increased adhesion to various materials. Soapstock is a by-product of the fat processing industry. It has hydrophobic properties and is resistant to certain acids and alkalis, helping to increase the durability of concrete structures. When combined with distillers' grains, it increases strength due to its water-reducing effect. Acrylic latex is a colloidal polymer that improves the hydrophobicity of concrete by polymerising and blocking micropores and microcracks in the cement matrix.

Table 1 shows the types of samples to be tested (excluding water permeability and chemical resistance tests, which are detailed below) and their respective compositions. The granite-to-sand ratio is 1:2, 1:3, 1:4 and 1:5 by weight. The ratios of soapstock to caustic soda and cement, and of

caustic soda to soapstock, are 6%, 8%, 10%, 12%, 14% and 1% by weight, respectively. The ratio of post-alcohol distiller's grains to water is 2%, 4%, 6% and 8% by weight. The ratio of acrylic latex to soapstock and water is 0.1%, 0.2%, 0.3% and 0.4% by weight.

**Table 1.** Sample types and their compositions.

Sample type (Contents,%)	Component content by mass, g							
	Sand	Gr	Cement	Sp	NaOH	PaB	Water	Lx
Type 1 (RS-Reference sample)								
Type 2 (Gr1%)	1485	15	500	-	-	-	200	-
Type 3 (Gr2%)	1470	30	500	-	-	-	200	-
Type 4 (Gr3%)	1455	45	500	-	-	-	200	-
Type 5 (Gr4%)	1440	60	500	-	-	-	200	-
Type 6 (Sp6%)	1500		470	29.7	0.3	-	200	-
Type 7 (Sp8%)	1500		460	39.6	0.4		200	
Type 8 (Sp10%)	1500		450	49.5	0.5		200	
Type 9 (Sp12%)	1500		440	59.4	0.6		200	
Type 10 (Sp14%)	1500		430	69.3	0.7	-	200	-
Type 11 (PaB2%)	1500			500		4	196	
Type 12 (PaB4%)	1500			500		8	192	
Type 13 (PaB6%)	1500			500		12	188	
Type 14 (PaB8%)	1500			500		16	184	
Type 15 (Lx0.1%)	1500			500			199.6*	0.2
Type 16 (Lx0.2%)	1500			500			199.2*	0.4
Type 17 (Lx0.3%)	1500			500			198.8*	0.6
Type 18 (Lx0.4%)	1500			500			198.4*	0.8

\* the exact water-to-cement ratio is determined by testing the normal density of the cement, given the water-reducing effect of acrylic latex due to plasticisation.

The components were added to the additive in stages and in the following order: Gr, Sp, PaB and Lx. First, the finely dispersed filler was added, followed by the plasticisers and then the polymer component. It should be noted that each subsequent component was introduced into the optimal composition, which included the previous component under study and identified the optimal transformations of the physical and mechanical properties of concrete.

The optimal composition of the modified additive was determined through a series of studies that assessed the impact of each component on the quality indicators of the cement-sand mixture:

Stage 1: Assessment of the impact of granite dust on the physical and mechanical properties of concrete with varying compositions;

Stage 2: Assessment of the effect of soapstock on the physical and mechanical properties of concrete with varying compositions;

Stage 3: Assessment of the effect of post-alcohol distiller's grains on the physical and mechanical properties of concrete with varying compositions;

Stage 4: Assessment of the effect of acrylic latex on the physical and mechanical properties of concrete with varying compositions.

Table 2 shows the types of tests performed on each component, along with their sequence, according to the expected changes in concrete characteristics.

**Table 2.** Shows the test suite for the component being introduced.

Type of test	Additive component			
	Gr	Sp	PaB	Lx
X-ray diffraction analysis	Yes	Yes	Yes	Yes
Compressive strength	Yes	Yes	Yes	Yes
Flexural strength	Yes	Yes	Yes	Yes

Water absorption	Yes	Yes	Yes	Yes
Water permeability	Yes	No	No	Yes
Chemical resistance	Yes	No	No	Yes

The assessment of chemical resistance and water permeability was carried out when the first component (Gr, which has a compaction effect) and the last component (Lx, which has a volumetric hydrophobisation effect) were introduced. This assessment is regulated by reducing the permeability and volumetric hydrophobisation of concrete. It should be noted that, at the time of introduction of Lx, the samples already contained the optimal content of Gr, Sp and PaB. Therefore, when assessing chemical resistance and water permeability, samples with variable Gr can be considered as reference samples and samples with variable Lx as control samples. For the tests of water permeability and chemical resistance, type 1 samples correspond to RS; type 2-5 samples correspond to samples with variable Gr; and type 6-9 samples correspond to samples with variable Lx (see Figure 1).

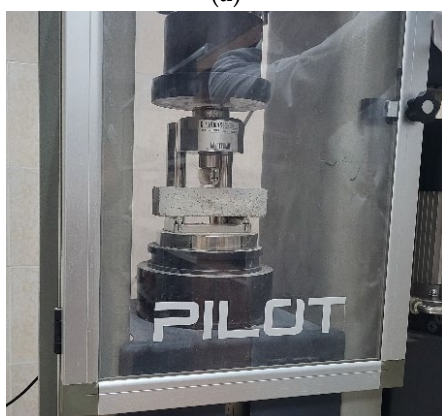
X-ray diffraction analysis was used to evaluate the mineralogical composition of the samples, determine the elemental and mineralogical composition, and assess the influence of the additive components on the phase composition and crystal structure of the cement material (Figure 1a). The strength characteristics of the samples were evaluated by compressing and bending the samples using the standard method specified in GOST 310.4 (Figures 1b and 1c). The water absorption capacity of the concrete samples was evaluated in accordance with GOST 12730.3 (Figure 1d). Water permeability was assessed in accordance with GOST 12730 (Figure 1e). Chemical resistance was assessed in accordance with GOST 58895 (Figure 1f).



(a)



(b)



(c)



(d)



Figure 1. Laboratory testing of concrete samples.

A device was used to supply water to the lower end surface of the samples at increasing pressure for water permeability testing. The water permeability rating was assessed using both the wet spot method and the filtration coefficient measurement method. The sample area was 100 cm<sup>2</sup>, and the sample height was 5 cm. Wet spot tests were performed with pressure increases in increments of 0.2 Mpa applied to the sample. The exposure time for each pressure step was at least 1 hour, until signs of filtration were observed. Signs of filtration capacity were visually assessed as water passing through the sample, which appeared as drops or a wet spot on the opposite surface. Filtration coefficient tests were performed at a pressure of 0.2 Mpa (previously determined by the wet spot method) for Gr samples and at a pressure of 2.0 Mpa (the maximum pressure) for Lx samples. Depending on the accumulation of filtrate, the exposure time was 12 hours. Water permeability is determined by the filtration coefficient according to the formula:

$$K_f = \frac{nQ\delta}{S\tau\rho} \quad (1)$$

where  $n$  is a coefficient that takes into account the viscosity of water; at a temperature of 22 °C, it is taken to be equal to 0.956 (according to GOST 12730, clause 5.4.2);

$Q$ —filter volume, cm<sup>3</sup>;

$\delta$ —sample thickness, cm;

$S$ —sample area, cm<sup>2</sup>;

$\tau$ —sample test time, during which the volume of the filtered liquid is measured, s;

$\rho$ —pressure in the installation, cm water column.

Chemical resistance tests were performed by exposing concrete samples to an aggressive environment and then measuring their flexural strength. A total of five beam samples of each type were tested under controlled conditions at 30-, 60-, 90-, 180-, 270- and 360-day intervals. There were 30 samples of each type of beam. The tests were carried out in an aggressive environment represented by a 10% aqueous solution of sulphuric acid. Based on the results of calculating the chemical resistance coefficients, a forecast was made for the life cycle of concrete structures during long-term operation in such environments. The chemical life cycle forecast essentially involves determining the decrease in the chemical resistance coefficient:

$$\begin{cases} \lg k_{GR} = a + b \times \lg \tau \\ a = (\lg \overline{k_{GR}} - b \times \lg \bar{\tau}) \\ b = \left( \frac{\sum (\lg \overline{k_{GRi}} - \lg k_{GRi}) \times (\lg \bar{\tau}_i - \lg \tau_i)}{\sum (\lg \bar{\tau}_i - \lg \tau_i)^2} \right) \end{cases} \quad (2)$$

Где  $k_{GR}$ —chemical resistance coefficient calculated by raising the value obtained in (2) to a power;

$\lg \overline{k_{GR}} = \frac{\sum k_{GRi}}{n}$ —average values of the logarithm of the chemical resistance coefficient;

$\lg \bar{\tau} = \frac{\sum \bar{\tau}_i}{n}$ —average values of the logarithm of the test time;

$lgk_{GRi}$  и  $lg\tau_i$ —logarithms of the chemical resistance coefficients and test times in the  $i$ -th series of samples corresponding to the intermediate (control) test periods;  
 $n$ —number of control tests.

### 3. Results

#### 1. X-ray structural analysis

Figures 2 and 3 and Table 3 show the results of X-ray structural analysis. Figure 2 shows microscopic illustrations of the samples, and Figure 3 shows the results. To demonstrate the dynamics of composition changes, the table shows the results for elements with the minimum and maximum content of a specific component.

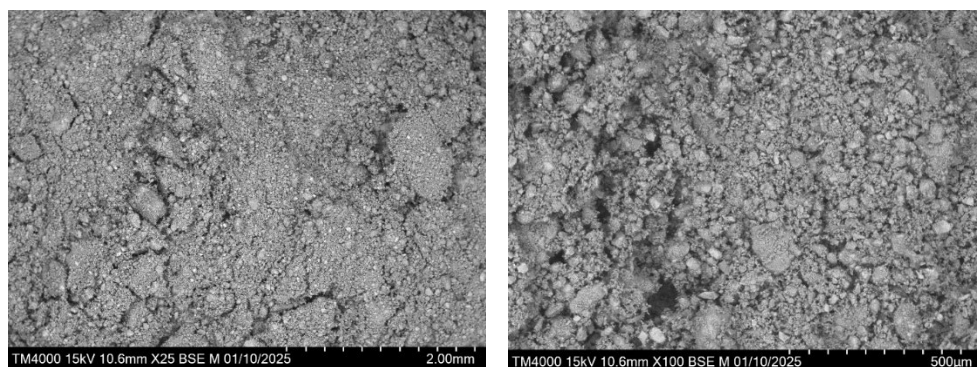


Figure 2. Microscopic photographs of the sample.

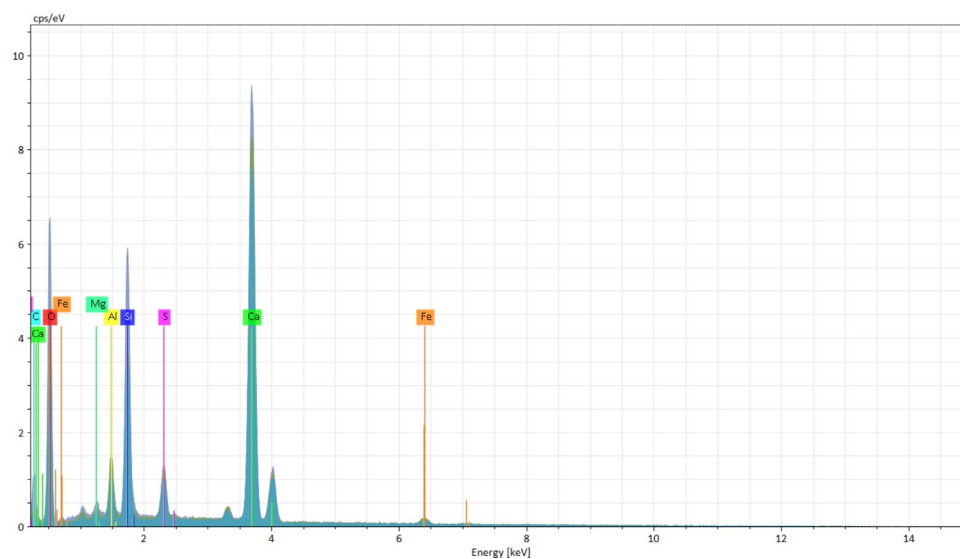


Figure 3. Elemental composition of reference sample.

Table 3. Elementary composition.

Types	O	Ca	Si	C	S	Al	Fe	Mg	K	Na
Type 1	46.43	36.66	8.39	2.72	2.78	2.01	1.02	0.00	0.00	0.00
Type 2	48.23	34.07	6.87	2.49	2.58	2.56	1.9	0.42	0.88	0.00
Type 5	45.68	37.77	9.39	2.38	1.78	2.13	0.87	0.00	0.00	0.00
Type 6	45.21	29.38	7.73	9.75	1.66	4.31	1.05	0.00	0.91	0.00
Type 10	41.66	24.85	6.85	18.64	2.13	2.37	1.62	0.00	0.00	1.88
Type 11	39.40	25.69	8.41	19.27	1.82	3	1.62	0.00	0.00	0.79
Type 14	37.52	23.46	6.35	25.65	2.56	2.42	1.15	0.00	0.00	0.89
Type 15	45.71	13.93	16.77	14.99	1.1	3.66	0.57	0.00	1.51	1.76
Type 18	42.13	27.38	6.35	17.89	1.82	1.9	1.36	0.60	0.00	0.57

A chemical analysis was conducted on the use of granite powder in combination with a cement binder. The results showed that, unlike the control sample, the silicon content changed insignificantly at a quantity of 1% of the sand mass, but a slight increase in aluminium was observed. This is due to the high silicon content and relatively low aluminium content of the granite powder. The presence of 48.23% oxygen indicates an increase in aluminium, silicon and iron oxides. Using granite powder at 4% by weight of sand increased the silicon content to 9.39%, which is 12% higher than the control sample. This increase in silicon oxides justifies the increased resistance to aggressive environments.

Adding soapstock at 6% of the cement mass results in a decrease in calcium, an increase in aluminium, and a slight increase in silicon. This shows that as calcium decreases, calcium carbonates increase, improving wetting, compaction and hydrophobicity. At a soapstock-to-caustic soda ratio of 3%, the carbon content increases by 9.7%, while the calcium content decreases to 29.38%. Adding 14% soapstock to cement results in a carbon content of 18.64% and a calcium content of 24.85%. The increase in carbon relative to calcium indicates that aluminium and silicon carbonates are also formed in addition to calcium carbonate. This has a positive effect on the hydration process, mixture compaction, hydrophobicity and the strength of the concrete.

Adding 2% post-alcohol distiller's grains by weight to water increases the oxygen content to 39.4% and the carbon content to 19.27%. This indicates an increase in carbonate content and a decrease in calcium oxides and hydroxides, which has a positive effect on concrete durability. Adding 8% post-alcohol distiller's grains by weight of water results in a slight decrease in oxygen to 37.52%, a decrease in calcium to 23.46% and a noticeable increase in carbon to 25.65%. These results demonstrate an increase in the activity of the resulting mixture, with the formation of a significant quantity of carbonates that contribute to the durability and resistance to aggressive environments of the concrete.

Adding 0.1% acrylic latex increases silicon by 16.77% and aluminium by 3.66%. This is because a small amount of acrylic latex creates a negatively charged polymer film within the cement stone structure. Since silicon and aluminium are positively charged, movement of these particles is stimulated, leading to changes in chemical reactions and a decrease in the amount of carbonates in the cement stone. This process occurs with a small amount of acrylic latex in the cement stone, but this effect decreases with a large amount. The most balanced composition is observed when 0.4% acrylic latex is added. In terms of chemical composition, this does not differ significantly from compositions without acrylic latex, confirming the influence of electrostaticity when a small amount of acrylic latex is present.

## 2. Compression and bending strength tests

Figure 4 shows the results of the compressive strength tests conducted on the samples. The serial numbers on the y-axis correspond to the sample types listed in Table 1. Figure 4a shows the curves for each individual component (dotted lines) and the strength growth curve, which illustrates the general trend of strength increase relative to the order in which each component is included at the optimum concentration. Figure 4b shows the coefficients of variation for six samples of each type.

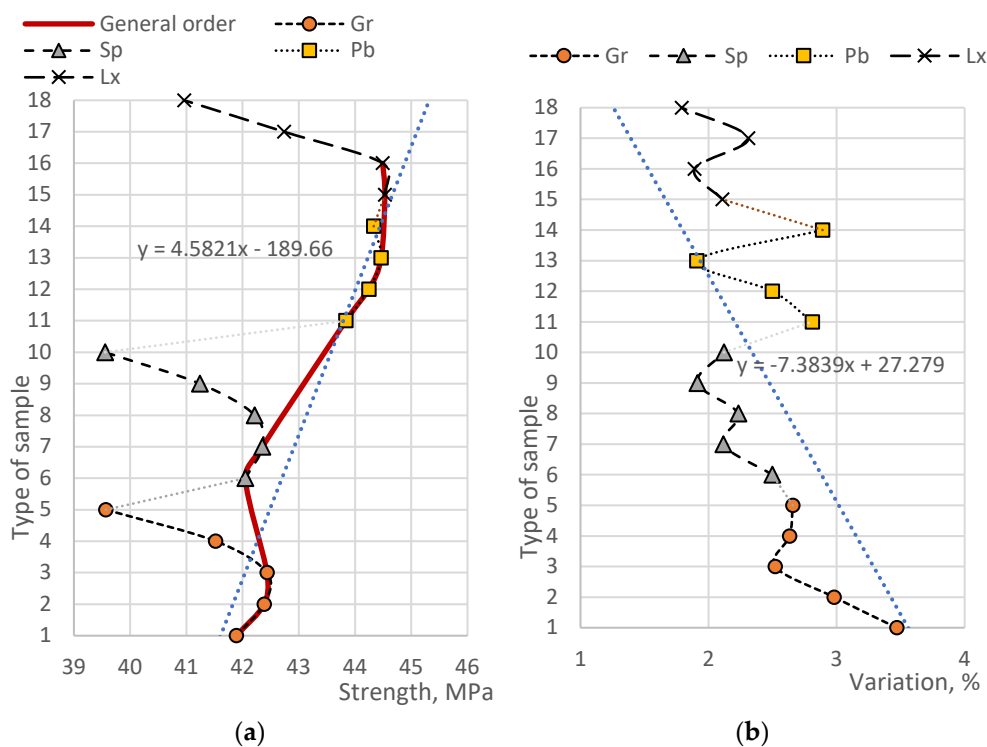


Figure 4. Determination of compressive strength.

According to the results of tests on reference samples (Type 1) under compression, the strength data points at 28 days and 100% strength gain vary from 41.09 to 42.14 MPa, with an average of 41.89 MPa. A close relationship was revealed between the strength data points of all Type 1 samples, with coefficients of variation not exceeding 3.5% (statistical error). Therefore, any change in strength exceeding 3.5% can be attributed to the influence of the introduced component on strength. The Gr curve shows a peak in strength change, with the average maximum strength data points for samples 2–5 being: 42.38, 42.43, 41.52 and 39.57 MPa. Changes in the strength of samples with Gr content of 1%, 2% and 3% RF are within the statistical error of RF and are: 1.17%, 1.29% and -0.88% (<3.5%). When the Gr content increases to 3%, there is a tendency for strength to decrease; however, the decrease only becomes noticeable in relation to the statistical error at 4%: -5.54% > |3.5%. Therefore, 2% is considered the optimal Gr concentration in concrete in terms of changes in compressive strength. Subsequent tests on samples 6-18 will include 2% granite dust in the RS composition.

Similar analyses were performed on the results for the other components of the additive, which also show a peak pattern of strength variation with component concentration. For the soapstone component, the error criterion was 1.54%, corresponding to the coefficient of variation of the samples at a 2% granite content. Changes in the strength of samples containing 6%, 8% and 10% Sp in addition to 2% RF and 2% Gr are within the statistical error of the RF+2%Gr sample, amounting to: -0.90%, -0.19% and -0.51%, which is less than the statistical error of 1.54%. However, a subsequent increase in Sp to 12% and 14% leads to a noticeable decrease in strength that exceeds the statistical error: -2.80% and -6.78%, respectively. Therefore, the optimal Sp concentration in the concrete composition, relative to changes in compressive strength, is 8%. Subsequent tests on samples 11–18 will include RS, 2% Gr and 8% Sp in the composition.

For the post-alcohol distiller's grains component, the error criterion (Type 7: RF + 2% Gr + 8% Sp) was 2.11%. Changes in the strength of samples for all PaB variations exceeded the statistical error RF+2%Gr+8%Sp by amounts of: 3.5%, 4.47%, 4.98% and 4.68%. However, when 8% PaB is added to the concrete composition, there is a tendency for strength to decrease; therefore, 6% PaB is considered the optimal concentration. Subsequent tests on samples 15–18 will include RS, 2% Gr, 8% Sp and 6% PaB in the composition.

For the acrylic latex component, the error criterion (Type 13—RF+2%Gr+8%Sp+6%PaB) was 1.91%. Changes in the strength of samples for Lx 0.1 and 0.2% do not exceed the statistical error RF+2%Gr+8%Sp+6%PaB, amounting to: 0.14 and 0.06% < |1.19%|. A subsequent increase in Lx to 0.3 and 0.4% leads to a noticeable decrease in strength exceeding the statistical error: -3.87 and -7.88% > |1.19%|. Thus, the optimal concentration of the modifying additive, relative to changes in compressive strength, is accepted as RS, 2% Gr, 8% Sp, 6% PaB, 0.2% Lx.

Figure 5 shows the results of the bending strength tests carried out on the samples. The data is presented in the same way as in Figure 1.

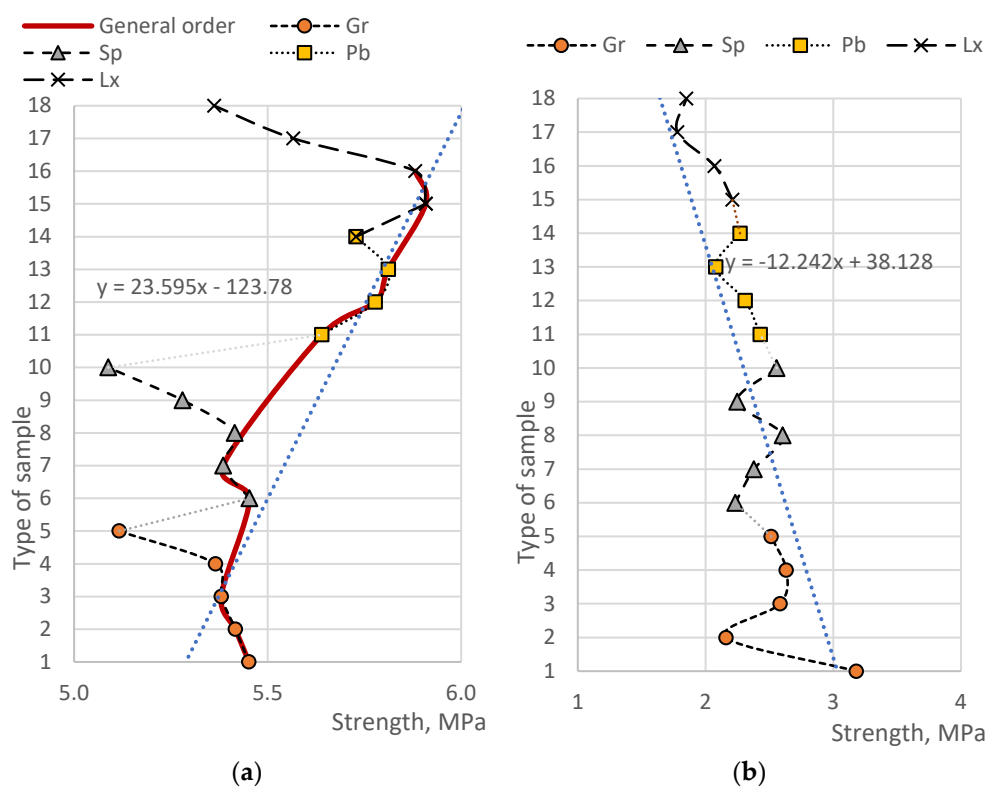


Figure 5. Determination of bending strength.

The results were analysed in the same way as those for compressive strength. All curves also exhibited a peak. The strength of the reference sample (Type 1) at 28 days and 100% strength gain varies from 5.33 to 5.66 MPa, with an average of 5.45 MPa. The coefficient of variation (RS) was 3.18% (statistical error). Changes in the strength of samples containing 1%, 2% and 3% RF are within the statistical error of RF and amount to: -0.64%, -1.31% and -1.59% (<3.18%). At any Gr content, there is a tendency for strength to decrease; however, at 4% Gr, the decrease is more significant in relation to the statistical error: -6.14% > |3.18%. As changes in strength of up to 3% can be attributed to statistical error, the optimal Gr concentration in concrete, in terms of changes in flexural strength, can be set at 2%.

For the soapstone component, the error criterion was set at 2.58%. Thus, the changes in strength of the samples containing 6%, 8%, 10% and 12% Sp in RF+2%Gr are within the statistical error margin and amount to: 1.36%, 0.09%, 0.65% and -1.85%, respectively. When 12% Sp is added to the composition, there is a tendency for strength to decrease; however, the 14% Sp concentration significantly exceeds the error criterion: -5.42 > 2.58%. As adding 12% Sp to the concrete composition decreases strength, the optimal concentration remains 6% Sp.

For the post-alcohol distiller's grains component, the error criterion was 2.38%. Changes in the strength of samples for all PaB variations exceeded the statistical error and amounted to: 4.73%, 7.30%, 7.92% and 6.38% (all greater than 2.38%). When 8% PaB is added to the concrete composition,

there is also a tendency towards a decrease in strength; therefore, the optimal PaB concentration remains unchanged at 6%.

For the acrylic latex component, the error criterion was 2.08%. Changes in the strength of samples for Lx 0.1% and 0.2% do not exceed the statistical error amounting to: 1.66% and 1.20%, which is less than the statistical error of 2.08%. However, a subsequent increase in Lx leads to a decrease in strength that exceeds the statistical error: -4.21% and -7.74%, which is greater than 2.08%. Thus, the optimal concentration of the modifying additive relative to changes in flexural strength remains unchanged: RS, 2% Gr, 8% Sp, 6% PaB and 0.2% Lx.

### 3. Water absorption tests

Figure 6 shows the results of the water absorption tests carried out on the samples. The data is presented in a similar way to that in Figure 1.

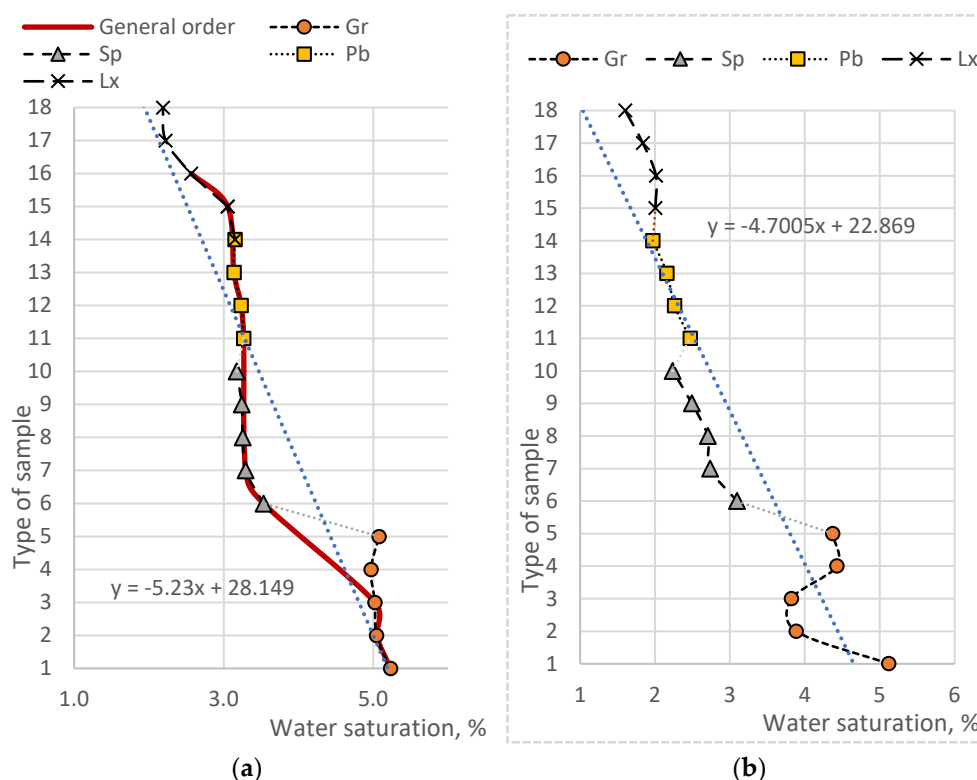


Figure 6. Determination of water absorption.

According to the test results, the water absorption value of the reference sample (Type 1) is 5.23%. For samples containing a granite additive, the same indicator was: Gr = 1%: 5.04%; Gr = 2%: 5.02%; Gr = 3%: 4.97%; Gr = 4%: 5.08%. There is a slight downward trend in the indicator in general, but it can be concluded that the additive has no effect on the water absorption capacity of concrete at any concentration, since the decrease does not exceed the statistical error RS, which is equal to 5.12%. 3.56%, 3.98%, 5.00% and 2.93%. The maximum concentration of Gr4% is the best in terms of reducing water absorption, but Gr2% remains optimal since an increase in concentration leads to a decrease in strength characteristics.

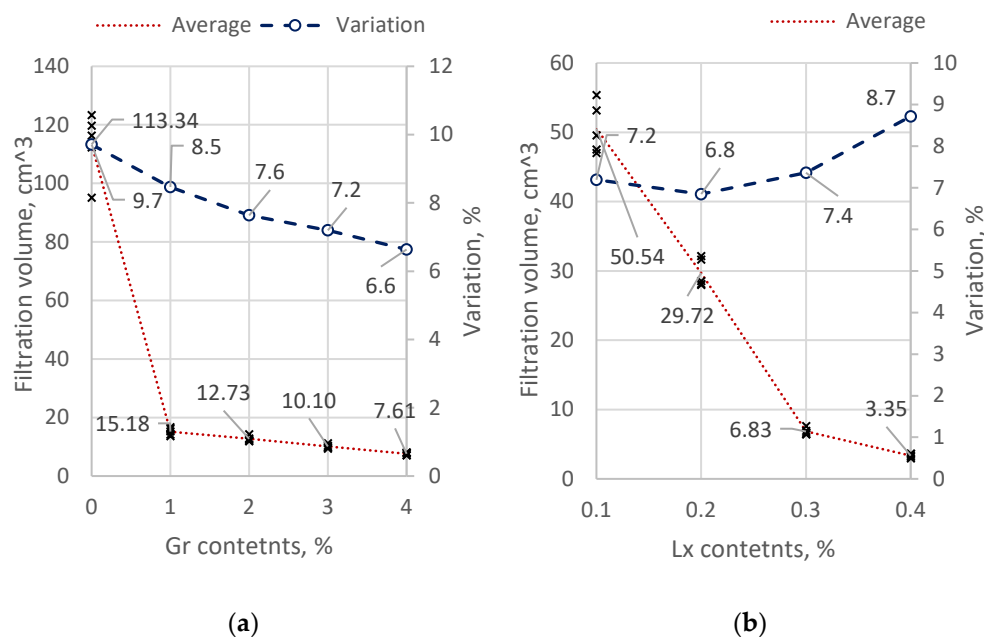
For the soapstock component, the error criterion was set at 3.82%, corresponding to the coefficient of variation of the Gr2% samples. Even with the initial introduction of Sp, there is a significant change in the water absorption of the samples, which exceeds the statistical error RF+2%Gr by a large margin: 29.7%, 34.4%, 35.2%, 35.4% and 36.7% >> 13.82%. As with Gr, each increase in Sp results in a decrease in water absorption. Therefore, the determining factor for the optimal Sp concentration is the concrete's strength. Therefore, the optimal concentration remains 2% Gr and 8% Sp.

For the post-alcohol distiller's grains component, the error criterion was set at 2.73%. Changes in the water absorption of the samples for PaB variations of 2% and 4% do not exceed the statistical error: 0.81% and 1.77% respectively, which is less than the 2.73% error criterion. When PaB is increased to 6% and 8%, there is a tendency for water absorption to decrease: 4.75% and 4.34%, respectively, which is greater than the error criterion of 2.73%. As with Sp and Gr, strength is the determining factor, so the optimal composition remains RS, 2% Gr, 8% Sp and 6% PaB.

For the acrylic latex component, the error criterion was 2.16%. Significant changes in the water absorption of the samples occur at Lx concentrations of 0.2% and above, amounting to: 18.2%, 29.1% and 30.3%  $\gg$  2.16%. As with Sp, Gr and PaB, strength is the determining factor, so the optimal composition is RS, 2% Gr, 8% Sp, 6% PaB and 0.2% Lx.

#### 4. Water permeability tests

Figure 6 shows the results of water permeability tests on concrete samples. Figure 6a shows the results of the volume of filtrate passing through samples with variable Gr content (the individual values are shown as data points and the averages are connected by a line). Figure 6b shows the same indicators for samples with variable Lx content and a fixed Gr content of 2%. Figures 6c and 6d show the results of filtration coefficient calculations using formula 1. The only difference is that the Gr samples were tested at a pressure of 0.2 MPa and the Lx samples at a pressure of 2.0 MPa for 12 hours according to the wet spot results. Figure 6b uses a logarithmic y-axis for clearer visualisation of the graph and values. Figure 6d shows a fragment of the graph without the RS samples, where the y-axis shows the absolute values of the filtration coefficients, enabling the pattern of change with increasing component concentration to be assessed. Figures 6b and 6d show the corresponding numerical values for the sample type on the x-axis.



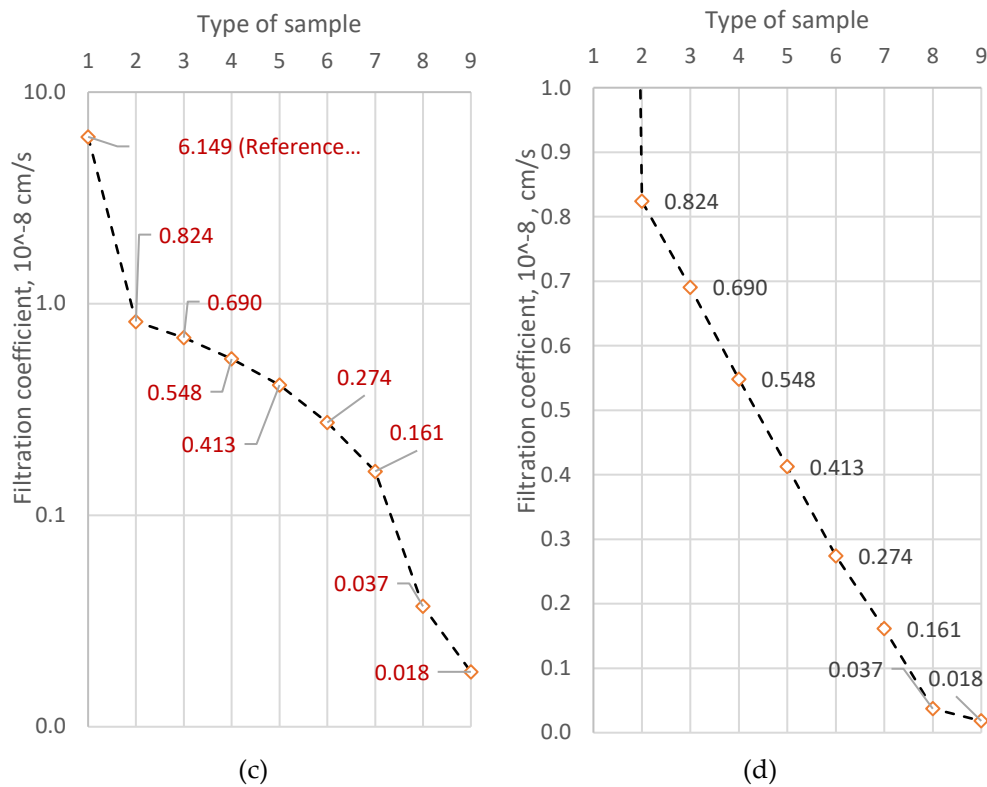


Figure 6. Determination of water permeability.

According to the RS tests, the data points for the volume of filtrate passing through the sample range from 95.06 to 123.32 cm<sup>3</sup>. The average value was 113.34 cm<sup>3</sup> with a standard deviation of 11.01, giving a coefficient of variation of 9.7%. With each subsequent addition of Gr from 1% to 4%, a decrease in the volume of filtrate passing through was observed, amounting to: 15.18 cm<sup>3</sup>, 12.373 cm<sup>3</sup>, 10.10 cm<sup>3</sup> and 7.6 cm<sup>3</sup>. A linear decrease in the volume of filtrate passed was also observed with each subsequent addition of Lx from 0.1 to 0.4%: 50.54, 29.72, 6.83 and 3.35 cm<sup>3</sup>. However, it is incorrect to compare the Gr and Lx values, since the tests were carried out at different pressures and sample exposure times.

Table 4 shows the filtration coefficient results based on the measured filtrate volume, which were calculated using Formula 1.

Table 4. Filtration coefficients.

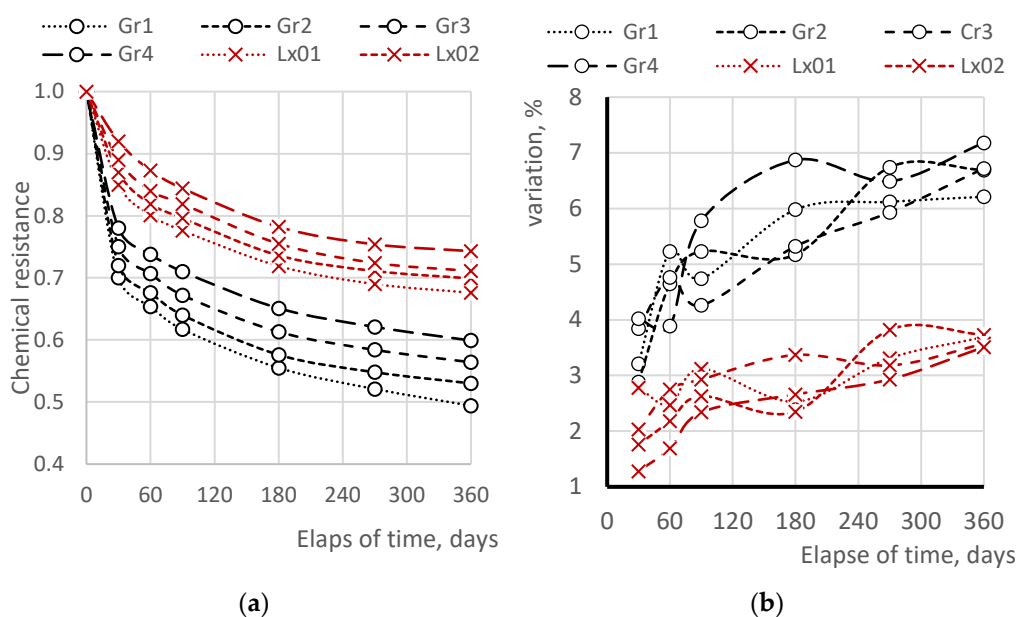
Type	n, GOST12730	Q, cm <sup>3</sup>	δ, cm	S, cm <sup>2</sup>	τ, c	ρ, MPa	K <sub>f</sub> , cm/s	Brand, GOST12730
RS	0.956	113.34	5	100	43200	2039.49	6.14x10 <sup>-8</sup>	W2
Gr1%	0.956	15.18	5	100	43200	2039.49	8.24x10 <sup>-9</sup>	W2
Gr2%	0.956	12.73	5	100	43200	2039.49	6.91x10 <sup>-9</sup>	W2
Gr3%	0.956	10.10	5	100	43200	2039.49	5.48x10 <sup>-9</sup>	W2
Gr4%	0.956	7.61	5	100	43200	2039.49	4.13x10 <sup>-9</sup>	W3
Lx0.1%	0.956	50.54	5	100	43200	2039.49	2.74x10 <sup>-9</sup>	W6
Lx0.2%	0.956	29.72	5	100	43200	2039.49	1.61x10 <sup>-9</sup>	W8
Lx0.3%	0.956	6.83	5	100	43200	2039.49	3.71x10 <sup>-10</sup>	W9
Lx0.4%	0.956	3.35	5	100	43200	2039.49	1.82x10 <sup>-10</sup>	W10

The dynamics of the change in filtration coefficient with variable Gr are linear with respect to the change in filtrate, as there is a proportional relationship between the two. When Gr was included in the composition, the change in concrete grade in terms of water permeability ranged from W2 to

W3; when Lx was added, it ranged from W2 to W10. The partial values obtained can be considered valid, since the coefficients of variation in all test series do not exceed 10%.

### 5. Chemical resistance tests

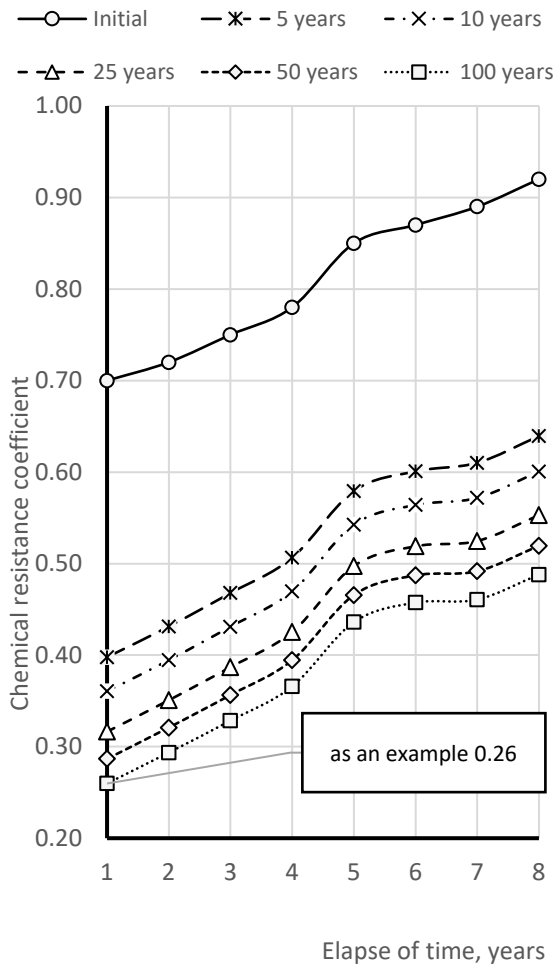
Figure 7 shows how the bending strength (hereafter 'strength') of samples changes depending on how long they are exposed to a 10% sulphuric acid solution (hereafter 'an aggressive environment'). Figure 7a illustrates the calculated chemical resistance coefficients of samples with different granite dust and acrylic latex compositions, while Figure 7b illustrates the corresponding coefficients of variation.



**Figure 7.** Determination of the actual chemical resistance coefficients.

The actual chemical resistance coefficient is determined by the ratio of residual strength (after exposure) to initial strength (before immersion in an aggressive environment):  $k_{(GR)} = \sigma_i / \sigma_I$ . According to the test results, an increase in chemical resistance is observed with each subsequent addition of the additive component. For samples with 4% Gr added, the average resistance after 360 days of exposure is 0.60; for the reference sample, it is 0.41. A similar trend is observed for samples containing Lx: with a maximum Lx concentration of 0.4%, the resistance is 0.74; with an Lx concentration of 0.1%, the resistance is 0.68. Gr samples have residual strength values that exceed RS by 7.3–21.3% (depending on the Gr concentration), whereas for Lx samples, the difference from RS is 36.8–50.4% (depending on the Lx concentration). The coefficients of variation for granite dust addition range from 2.9% to 7.2%, and for acrylic latex from 1.28% to 3.82%. The obtained coefficients of variation indicate high convergence of the data points and therefore high reliability of the results.

Figure 8a shows the results of direct calculations predicting a decrease in the chemical resistance coefficients of concrete structures in aggressive environments after prolonged operation (up to 100 years). The x-axis shows the serial numbers corresponding to the sample type (Gr1-4%, Type 1-4; Lx0.1-0.4%, Type 5-8), and the y-axis shows the chemical resistance coefficients. Figure 8b shows the results of reverse calculations for predicting the service life of concrete structures with a certain reduction in strength (from 10% to 60% of the initial strength). The x-axis shows the percentage loss of strength from the initial value and the logarithmic y-axis shows the service life in years.



Using the example of a Gr1% type sample

$$\begin{aligned} \sum \lg \tau_i &= 12.45 \\ \sum \lg k_{GRi} &= -1.39 \\ \sum \lg \bar{\tau}_i &= 2.08 \\ \sum \lg \bar{k}_{GRi} &= -0.23 \\ \sum (\lg \bar{k}_{GRi} - \lg k_{GRi}) \times (\lg \bar{\tau}_i - \lg \tau_i) &= -0.12 \\ \sum (\lg \bar{\tau}_i - \lg \tau_i)^2 &= 0.85 \end{aligned}$$

$$b = \frac{\sum (\bar{\tau}_i - \tau_i) \times (\bar{k}_{GRi} - k_{GRi})}{\sum (\bar{\tau}_i - \tau_i)^2} = -0.14$$

$$a = \lg \bar{k}_{GRi} - b \cdot \lg \bar{\tau}_i = 0.063$$

Then the equation has the form

$$\lg k_{CR} = 0.063 - 0.14 \lg \tau$$

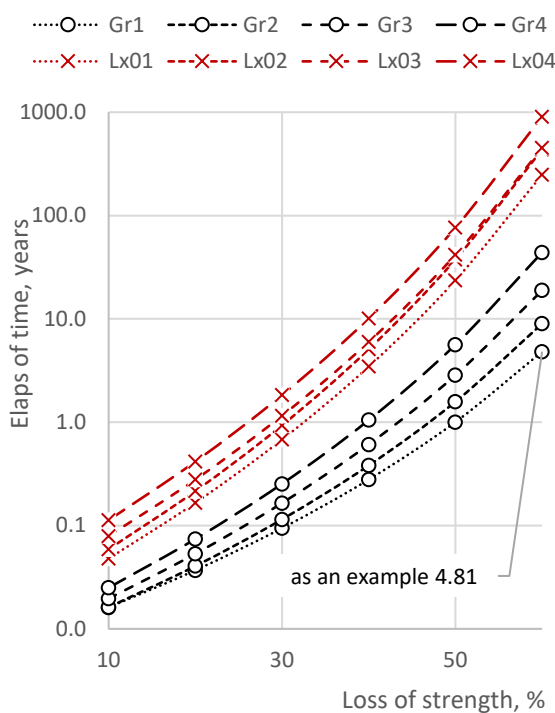
For a duration of 100 years,

$$\tau_i = 36000 \text{ days:}$$

$$\lg k_{CR} = 0.063 - 0.14 \lg 36000 =$$

$$0.063 - 0.14 \times 4.56 = -0.59$$

(a)



Reverse calculation using the example of a Gr1% sample

Let's transform the equation:

$$\lg k_{CR} = 0.063 - 0.14 \lg \tau$$

into the equation:

$$\lg \tau = \frac{\lg k_{CR} - 0.063}{-0.14}$$

Using the example of a loss of strength of 60% of the initial

(at which  $k_{CR} = 0.4$ ):

$$\lg \tau = \frac{\lg 0.4 - 0.063}{-0.14} = \frac{-0.4 - 0.063}{-0.14} = 3.24$$

then:

$$\tau = 1732.89 \text{ days or } = \frac{1732.89}{360} = 4.81 \text{ years}$$

360—the transition from days to years

Thus, for concrete with a 1% addition of granite dust, the duration of operation, at which the strength decreases by 10% from the initial one, is 4.81 years.

(b)

**Figure 8.** Determination of the actual chemical resistance coefficients.

According to the diagrams in Figure 8a, when concrete structures of type 1 are used in aggressive conditions, the residual strength of the concrete will reach 36% of its initial strength after 10 years of use, but the subsequent decrease in strength is not as intense. High intensity of aggressive environment exposure in the initial period of operation is characteristic of all types of samples, as is the decrease in intensity during subsequent operation. Nevertheless, the influence of the additive on the absolute values of the change in strength indicators can be traced: at the maximum concentration of the additive (type 8), the residual strength after 10 years of operation is 60%, and after 100 years of operation, 49%, which significantly exceeds the strength indicator of type 1 samples even after 10 years of operation.

As can be seen from the diagrams in Figure 8b, the life cycle of Type 1 samples is significantly shorter than that of Type 8 samples. Taking a 50% reduction in strength from the design value as an example, the service life of type 1 samples is one year, whereas for type 8 samples it is 76 years. In terms of the standard service life, during which a reduction in strength of 20–30 per cent is permissible depending on the type of structure and whether it is subjected to compression or bending, the service life is up to 13–34 days for type 1 samples and 150–658 days for type 8 samples.

#### 6. Selecting the optimal composition of the modifying additive.

Table 5 summarizes the changes in a particular characteristic according to the concentration of a specific component.

**Table 5.** Summary of test results.

Type	Compression strength, MPa	Banding strength, MPa	Water saturation, %	Filtration coefficient, cm/s	Chemical resistance
1 (RS-Reference sample)	41.89	5.45	5.23	$6.14 \times 10^{-8}$	-
2 (Gr1%)	42.38	5.42	5.04	$8.24 \times 10^{-9}$	0.49
3 (Gr2%)	42.43	5.38	5.02	$6.91 \times 10^{-9}$	0.53
4 (Gr3%)	41.52	5.37	4.97	$5.48 \times 10^{-9}$	0.56
5 (Gr4%)	39.57	5.12	5.08	$4.13 \times 10^{-9}$	0.60
6 (Sp6%)	42.05	5.45	3.53	-	-
7 (Sp8%)	42.35	5.39	3.29	-	-
8 (Sp10%)	42.22	5.42	3.26	-	-
9 (Sp12%)	41.24	5.28	3.24	-	-
10 (Sp14%)	39.56	5.09	3.18	-	-
11 (PaB2%)	43.84	5.64	3.27	-	-
12 (PaB4%)	44.25	5.78	3.24	-	-
13 (PaB6%)	44.47	5.81	3.14	-	-
14 (PaB8%)	44.33	5.73	3.15	-	-
15 (Lx0.1%)	44.53	5.91	3.05	$2.74 \times 10^{-9}$	0.68
16 (Lx0.2%)	44.49	5.88	2.57	$1.61 \times 10^{-9}$	0.70
17 (Lx0.3%)	42.74	5.57	2.23	$3.71 \times 10^{-10}$	0.71
18 (Lx0.4%)	40.96	5.36	2.19	$1.82 \times 10^{-10}$	0.74

According to the data presented in the table, the optimal additive composition in terms of strength indicators is as follows: granite dust (Gr)=2%, soapstone (Sp)=8%, post-alcohol distiller's grains (Pab)=6%, acrylic latex (Lx)=0.2%. Using this composition, the characteristics relative to the reference sample show the following advantages:

- Compressive strength increases by 6.2%.
- Flexural strength increases by 7.9%.

- Water absorption decreases by 50.1%.
- The filtration coefficient decreases by 97.4%.
- Chemical resistance increases by 42.8%.

#### 4. Discussion

According to the conducted studies of the influence of granite filler on the properties of cement and concrete, an increase in the physical and mechanical properties of concrete was determined in [16,17]. Also in [18], issues related to the use of soapstock and sodium hydroxide and their effect on concrete properties were studied. In the paper, soapstock is described as an effective hydrophobizer and additive to reduce concrete salinity. The properties of post-alcohol bard and acrylic latex were studied in [19,20].

The effect of acrylic latex is described in the works, which is confirmed in the process of studying concrete using this component. However, taking into account the characteristics of each component of the additive and after conducting a number of studies, the optimal ratios were determined. The optimal ratios were determined by X-ray diffraction analysis, and the chemical processes occurring during cement hydration at different component ratios were determined, as shown in Table 3 and Figure 2. In the course of the study, the factors influencing the physico-mechanical features of concrete were identified, presented in Table 4.

Figures 1a and 1b show a general positive trend in the increase in concrete strength under compression and bending. The general order curve, constructed along the tangents of each peak curve, illustrates the continuous increase in strength indicators with each subsequent addition of the component. The total increase in compressive strength relative to RF was 6.2% for compression and 7.9% for bending. The increase in strength at low Gr concentrations of up to 2% is due to an increase in concrete density and the pozzolanic effect of including silicon oxide in the composition. The decrease in compressive strength at high granite concentrations may be due to an excess of silicon oxide not participating in the hydration process. The reduction in flexural strength caused by Gr (at any concentration) may be due to the negative effect of increased density (increased mass per unit volume) outweighing the positive pozzolanic effect, which increases the chemical activity of Portlandites. The decrease in compressive strength when adding Sp at any concentration may be due to an excess of fatty acids. The latter has a negative effect on concrete strength, as a large quantity of fatty acids reduces moisture adsorption, thereby limiting the hydration process. Although fatty acids are evenly distributed when they enter the cement mortar due to the use of water-soluble caustic alkalis (NaOH), after setting the alkalis enter the active phase of the cement binder and subsequently form a hydrophobic structure. The increase in flexural strength when Sp is added may be due to a reduction in the concrete's internal stress. For any concentration of PaB, the increase in flexural compressive strength is primarily due to the water-reducing effect resulting from the plasticisation of the concrete mix. The increase in strength at low Lx concentrations can also be explained by the water-reducing effect of surface-active latex. However, the decrease in strength at high concentrations can be explained by the increased concentration of the polymer-containing component in the cement stone. In other words, at the optimum concentration, latex acts as a filler for micropores; at high concentrations, however, its function of replacing other, more durable structural components becomes more prevalent.

According to Figures 1b and 2b, it is necessary to note the reverse trend in the variation coefficients with each subsequent addition of the additive. The latter indicates a decrease in the dispersion of data points and stabilisation of the concrete structure formation.

According to Figure 5a, almost all the component curves have no peak values (except for Gr) and follow the general trend, indicating the continuous influence of components on reducing concrete's water absorption capacity. Therefore, the determining factor in selecting the optimal concentration remains concrete's fundamental property: strength.

The change in water absorption when Gr is added is primarily due to an increase in the concrete's density, which consequently decreases its filtration capacity. When Sp is first introduced

into the concrete composition, there is a noticeable decrease in water absorption, but the percentage increase in the decrease in water absorption with an increase in soap stock cannot be considered significant. With each subsequent addition of Sp, the relative decrease in water absorption decreases rapidly: 6.8% (at 8% soap stock), 1.2% (at 10%), 0.5% (at 14%) and 2.0% (at 14%). This pattern may be associated with the achievement of the maximum (or close to it) value of the hydrophobic effect of Sp. The decrease in water absorption with the addition of PaB is also explained by the plasticising effect, which reduces the number of micropores in the binder structure (at high concentrations). As the area of walls connecting with inert fillers is reduced, the contact zone decreases, resulting in a brittle structure. However, the nominal strength is maintained until the moment of partial destruction due to the dense structure of the material and the plasticising effect of the distiller's grains. Therefore, we consider a ratio of 6% by mass of cement to be optimal for post-alcohol distiller's grains in concrete compositions. A similar pattern to Sp is observed in the case of Lx, with a continuous decrease in water absorption. However, efficiency is lost as the concentration of Lx increases. This can be explained by the approach to the maximum hydrophobic effect, which is achieved by blocking micropores. Therefore, its excess will not significantly affect water absorption capacity.

Upon initial addition of the additive ( $Gr = 1\%$ ), a significant, abrupt decrease in the volume of the filtrate is observed, which is 13.39% of the reference sample; in other words, the volume of the filtrate decreases by 86.6%, or 7.5-fold. Subsequent additions of the additive result in a further decrease in water permeability, which is relatively linear: the volume of filtrate from the reference sample decreases by 88.8%, 91.1% and 93.3% (9 times, 11 times and 15 times, respectively) for  $Gr = 2\%$ ,  $Gr = 3\%$  and  $Gr = 4\%$ . In terms of the percentage effect of the Lx additive, water permeability decreases by 41%, 77% and 51% with each subsequent addition ( $Lx = 0.02\%$ ,  $0.03\%$  and  $0.04\%$ , respectively) and by 41%, 86% and 93% relative to the initial addition of the additive. The dynamics of the change in the filtration coefficient are shown with variable Gr from W2 to W3 and with the addition of Lx from W2 to W10.

As can be seen in Figure 7a, there is a noticeable difference between samples containing a latex additive and those containing a granite additive. This indicates that the hydrophobic effect of Lx has a greater impact on the chemical resistance of concrete than the reduction in water permeability due to increased density when Gr is added. The reasons for this are described above. In all cases, a sharp decrease in strength occurs during the first 30 days of exposure to an aggressive environment; after this period, the rate of decrease in strength slows down. The graphs in Figure 7b show that, in both cases, the spread of individual values increases with exposure to an aggressive environment. This suggests that the aggressive environment has a relatively individual destructive effect on each sample. Nevertheless, this spread remains within acceptable limits and may be associated with the random formation of the structure of each specific sample (including the formation of microcracks and micropores). This affects the rate and nature of aggressive environments penetrating the sample structure, which has a destructive effect when interacting with concrete. It should be noted that the coefficients of variation for samples containing Lx are significantly lower than for samples containing Gr, indicating greater stability in the results for samples containing latex. This can be explained by the hydrophobic effect of latex, which compensates for errors in random structure formation and the formation of microcracks in cement stone structures. In other words, acrylic latex coats the surface of micropores and microcracks, thereby preventing interaction with the aggressive environment, so its penetrating ability is not a minor factor.

The results of the serviceability assessment of concrete structures (with respect to concrete performance in an aggressive environment) subjected to long-term exposure to an aggressive medium (a 10% aqueous solution of sulfuric acid), as shown in Figure 4, are of an individual nature. The extension of the service life is attributed to the combined influence of several factors: the presence of soapstock and acrylic latex promotes volumetric hydrophobization; the presence of post-alcohol distillery stillage increases density by reducing microporosity; and the presence of granite powder reduces the contact area with the binder. Thus, the combined effects led to an increase in the chemical resistance of the concrete, which is quantitatively confirmed by laboratory studies. Although life cycle

calculations relative to standardised strength reduction indicators (20-30%) may seem low (with a service life of up to 13–34 days for type 1 samples and 150–658 days for type 8), the service life may be longer in practice for two reasons. Firstly, the aggressiveness of the environment may be lower than in the tests. Secondly, increasing the design strength relative to the degree of aggressiveness of the environment (rather than relative to loads) will significantly increase the service life, since the intensity of the impact decreases significantly over time, as mentioned earlier. For example, with a design strength of 25 MPa, the permissible strength reduction is 17.5 MPa, which is 70% of the design strength. If a strength of 35 MPa is assumed, then 17.5 MPa is 50% of the design strength. As mentioned earlier, a 50% reduction in strength results in a service life of 76 years.

According to the summary results presented in Table 4, the optimal composition in terms of strength indicators is as follows:

Granite dust: Gr = 3%

Soapstone: Sp = 8%

Post-alcohol distiller's grains: Pab = 6%

Acrylic latex: Lx = 0.2% These indicators show a significant improvement in physical and mechanical characteristics (see below). However, if the main indicator is not strength but water absorption, for example, then another composition can be selected as optimal, in which the best water absorption characteristics are manifested. In this case, a different composition can be accepted, but with an adjustment for the cement grade.

## 5. Conclusions

1. Energy-dispersive X-ray spectrometry revealed that adding up to 4% by weight of granite powder to sand results in a significant increase in silicon content and its corresponding oxides. This explains why cement stone becomes more resistant to aggressive environments. Adding soapstock and post-alcohol distiller's grains decreases the proportion of free calcium and increases the proportion of carbon and oxygen. This results in the formation of a greater number of carbonate phases, which improves the compaction of the structure and increases the hydrophobicity and durability of concrete. Small doses of acrylic latex (0.1–0.4%) alter the distribution of silicon and aluminium through electrostatic interactions. At 0.4%, the most balanced chemical composition is formed without compromising the structure, confirming the optimality of this dosage. Overall, the observed changes in elemental composition align with the enhanced density, crack resistance, and corrosion resistance of the modified concretes. Based on the data obtained from the analysis, the hypothesis that chemical processes influence the change in the physical and mechanical properties of concrete is confirmed. The influence of electrostatics on the quality of chemical processes was also revealed.

2. A series of laboratory tests were carried out on concrete samples with varying compositions of the following components in the following proportions, in order to assess changes in their physical and mechanical properties:

Granite (Gr): 1%, 2%, 3% and 4% by weight relative to sand

Soapstock (Sp): 6%, 8%, 10%, 12% and 14% by weight relative to cement

Post-alcohol distiller's grains: 2%, 4%, 6% and 8% by weight relative to water

Acrylic latex: 0.1%, 0.2%, 0.3% and 0.4% by weight relative to soapstone and water

3. Research into the influence of granite dust (Gr) on the physical and mechanical properties of concrete indicates that the optimal Gr concentration for demonstrating the best concrete properties is 2%. This results in an increase in compressive strength of 1.3% compared to the reference sample (RS), a decrease in flexural strength of 1.3%, a decrease in water absorption of 4.0%, a decrease in the filtration coefficient of 88.7% and an increase in chemical resistance of 8.2%. The changes in compressive and flexural strength of 1.3% compared to the reference sample (RS) can be attributed to statistical error since this value does not exceed the coefficient of variation of the RS. However, a significant decrease in both compressive and flexural strength is observed with a subsequent increase

in the concentration of Gr, which exceeds the statistical error and therefore has a negative effect on the strength properties of concrete.

4. Studies on the effect of soapstock (Sp) on the physical and mechanical properties of concrete have shown that the optimal Sp concentration for achieving the best concrete properties is 8%. This results in an increase in compressive strength of 1.1% relative to the reference sample, a 1.1% decrease in flexural strength, and a 37.1% decrease in water absorption. While each subsequent addition of Sp leads to a positive trend of decreasing water absorption, the strength properties show an opposite downward trend. Thus, as with granite dust, strength characteristics are decisive in selecting the optimal concentration. At Sp = 8%, changes in strength do not exceed the statistical error RS (i.e., there is no decrease in strength), and the transformation of water absorption capacity indicators is quite noticeable.

5. Studies examining the effect of post-alcohol stillage (PaB) on the physical and mechanical properties of concrete have found that the optimal PaB concentration for achieving the best concrete performance is 6%. This results in a 6.2% increase in compressive strength, a 6.6% increase in flexural strength and a 40.0% decrease in water absorption relative to the reference sample. A positive trend towards increasing concrete strength is observed with PaB, which can be attributed to the additive's influence, as the obtained values significantly exceed the statistical error. The optimal PaB concentration of 6% was chosen because further increasing PaB results in a negative trend of reducing concrete strength and increasing water permeability.

6. Studies on the effect of acrylic latex Lx on the physical and mechanical properties of concrete indicate that the optimal Lx concentration for demonstrating the best concrete properties is Lx = 0.2%. This results in an increase in compressive strength of 6.2% compared to the reference sample, a decrease in flexural strength of 7.9%, a decrease in water absorption of 50.1%, a decrease in the filtration coefficient of 97.4% and an increase in chemical resistance of 42.9%. Although changes in strength properties are insignificant in relation to PaB, there is still a positive trend. However, a decrease in water absorption capacity and water permeability is quite noticeable. The most significant finding, however, is the sharp increase in the chemical resistance of concrete, which rises from 8.2 (at Gr2%) to 42.9%. Opting for an Lx concentration of 0.2% is associated with a sharp decrease in concrete strength with subsequent increases in concentration, despite continuous positive growth in other indicators.

7. According to the research, the optimal concentrations of the modifying additive components are as follows: granite dust (Gr) = 2%, soapstock (Sp) = 8%, distillation waste (Pab) = 6%, and acrylic latex (Lx) = 0.2%. There are significant changes in the physical and mechanical properties relative to RS, with all evaluation criteria showing positive improvement, particularly in terms of resistance to aggressive environments. The proposed additive helps to extend the lifespan of concrete structures in aggressive environments. Using the additive can increase the service life of concrete structures from 1–2 years to 76 years, as presented in the article as an example in the discussion. With proper design, the service life can be extended significantly further. The use of the additive makes it possible to increase the service life of concrete structures in aggressive environments (10% sulfuric acid solution) from 1-2 years to 76 years (presented in the article as an example in the discussion), and with proper design by much more.

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