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Iryna Kirakevych, Bohdan Rusyn, Yurii Bobetskyi

CEMENTITIOUS SYSTEMS FOR HIGH-PERFORMANCE CONCRETES WITH IMPROVED CORROSION RESISTANCE

Department of Building Production,
Lviv Polytechnic National University
iryna.i.kirakevych@lpnu.ua

The article presents the results of research on cementitious systems "Portland cement CEM I 42,5 R – active mineral additives – microfillers – superplasticizer – hardening accelerators" for high-performance concrete with improved corrosion resistance. The resistance of concrete to corrosion caused by the influence of chemical substances was investigated – sulfate corrosion (class XA), which combines the processes of formation and accumulation of sparingly soluble salts in concrete, which are accompanied by internal stresses and destructive phenomena in concrete. The increase in corrosion resistance of high-performance concretes based on modified cementitious systems is explained mainly by the creation of a fine-crystalline microstructure with the formation of C-S-H phases, which contribute to the pores colmatation with age of hardening.

Key words: high-performance concretes; polycarboxylates; highly dispersed mineral additives; cementitious systems; carbonization; corrosion resistance.

Introduction

Durability is the ability to last a long time without significant deterioration. A durable material helps the environment by conserving resources and reducing wastes and the environmental impacts of repair and replacement. The production of replacement building materials depletes natural resources and can produce air and water pollution. Concrete resists weathering action, chemical attack, and abrasion while maintaining its desired engineering properties (Sanytsky, Kropyvnytska, Heviuk, Sikora & Braichenko, 2021). Different concretes require different degrees of durability depending on the exposure environment and the properties desired. Concrete ingredients, their proportioning, interactions between them, placing and curing practices, and the service environment determine the ultimate durability and life of the concrete (Sohail, Kahraman, Nuaimi, Gencturk & Alnahhal, 2021).

Modern high-tech concrete is a composite building material made using high-quality components, characterized by improved rheological, mechanical and technical properties, applied for new types of concrete structures, with high durability. P. Aicin (Aicin, 2003) admitts that in all developed countries, the production of "high-performance concrete" (HPC) is constantly growing. The term "high-performance concrete" refers to concrete that meets a set of special interconnected requirements for composition, technology, properties, specific purpose and operating conditions. Such concretes are characterized by high rheological (class according to specified slump test S4-S5) construction and technical properties: compressive strength class C60/70 and higher, frost resistance grade F300 and higher, high values of corrosion resistance, waterproofing, low porosity and abrasion (Sanytskyi, Rusyn, Kirakevych and Kaminskyi, 2023).

J. Jasiczak and others (Jasiczak, Wdowska & Rudnicki, 2008) showed that the idea of creating high-performance concrete involves optimization of their micro- and mesostructure due to high packing density at the level of finely dispersed particles (physical optimization); hydraulic and pozzolanic reactions at the level of supplementary cementitious materials (chemical optimization); improvement of the transition zone between the matrix of cement stone and aggregate (optimization of adhesion). Therefore, the modification of Portland cements and concretes with highly dispersed mineral additives is a relevant direction for obtaining concretes of a new generation (Switonski, Mrozik & Piekarski, 2004).

The research of the results in the field of construction material science shows that the directed formation of the necessary construction and technical properties of high-performance concrete is achieved due to the carefully controlled distribution of individual components in different particle size ranges of composite binders, which are obtained by modifying Portland cements with highly dispersed mineral additives of various types with increased values of surface energy and pozzolanic activity (modified composite portland cements) (Sanytsky, Kropyvnytska, Vakhula & Bobetsky, 2024). The complexity of designing high-performance concrete consists in ensuring both high technological properties of the concrete mixture and the necessary operational characteristics of hardened concrete (Runova, Gots, Rudenko, Konstantynovskyi & Lastivka, 2018). At the same time, concrete during it's life cycle is subject to the process of destruction (corrosion) as a result of chemical or physical influence. Electrochemical, chemical and biological corrosion of concrete are observed. The corrosion resistance of concrete depends on the phase composition of cement stone, since the solubility and reactivity of its individual phases differ significantly. The properties of aggressive environments that affect building structures made of concrete and reinforced concrete are extremely diverse, that is why the study of the corrosion resistance of concrete, as well as the study of corrosion processes and the resistance of cementitious systems to various negative influences, is an urgent task (Nivin, Jędrzejewska, Varughese & James, 2022).

Corrosion processes in concrete structures under the influence of seawater, aggressive underground and industrial wastewater containing chlorides, sulfates, and magnesium ions are based on the interaction of these ions with calcium hydroxide and hydroaluminates of cement stone. At the same time, the number of reaction products formed from a unit volume of the reacting cement stone component can increase by 2–5 times. This causes the occurrence of internal stresses, the formation of cracks and leads to corrosion of concrete. That is why, in order to prevent this type of corrosion during construction, it is necessary to use concretes of a new generation based on cementitious systems with a reduced content of portlandite, which essentially enters into exchange reactions in aggressive environments (Valcuende, Lliso-Ferrando, Ramón-Zamora & Soto, 2021). Improved corrosion resistance of high-performance concrete is possible by increasing the potential of the cementitious system due to a complex combination of effective highly dispersed pozzolanic additives and polycarboxylate-based superplasticizers. The consequence of this is the directed regulation of the hydration mechanism of such systems and the formation of a fine crystalline microstructure in high-performance concretes (Kropyvnytska, Sanytsky, Rucińska & Rykhlitska, 2019).

The analysis of data in the field of monolithic concreting technology, as well as the known regularities of the formation of the structure of artificial stone with the necessary properties, shows the feasibility of developing cementitious systems by combining complex chemical additives with a plasticizing-accelerating effect and rational selection of finely dispersed mineral additives of various types (Krivenko, Petropavlovskyi & Kovalchuk, 2018). Therefore, the production of high-performance concrete with improved corrosion resistance requires the study of cementitious systems “Portland cement CEM I-42,5 R – active mineral additives – microfillers – superplasticizer – hardening accelerators”, which allows to control the production and kinetics of structure formation, intensify the initial stages of hardening and create a particularly dense structure, to obtain high indicators of the early strength of hardened concrete, providing high corrosion resistance (Kirakevych, Sanytsky, Shyiko & Kagarlitskiy, 2021). Minerals of the cementitious matrix after hydration have different solubility in water, therefore most concrete corrosion processes in inorganic salt solutions are determined by exchange chemical reactions between cement hydration products and substances dissolved in water (Borziak, Plugin, Chepurna, Zavalniy & Dudin, 2019).

According to EN 206-1:2000 the aggressive environment is divided into classes. The work studied the resistance of concrete to corrosion caused by the influence of chemicals – sulfate corrosion (class XA) (Chousidis, Rakanta, Ioannou & Batis, 2015). This type of corrosion combines all the processes of interaction of concrete, which are associated with the formation and accumulation of sparingly soluble salts in concrete, which are characterized by an increase in volume during the transition to the solid phase, the formation of crystals, which in turn are accompanied by internal stresses and destructive phenomena in concrete (Gots, Berdnyk, Lastivka, Maystrenko & Amelina, 2023). Such phenomena occur as a result of crystallization not only of reaction products, but also of salts in the pores of concrete that come from the outside in the form of solutions (Valcuende, Parra, Marco, Garrido, Martínez & Cánoves, 2012).

The increase in corrosion resistance of high-performance concretes based on cementitious systems is explained by the creation of a fine crystalline microstructure (Ting, Wong, Rahman & Meheron, 2021). Highly dispersed mineral additives of pozzolanic action ensure binding of portlandite in C-S-H phases, which contributes to the colmatations of pores with age of hardening, such a structure of cement stone in concrete ensures an increase in its corrosion resistance (Sun & Wu, 2022).

The aim of this study is the research of cementitious systems "Portland cement CEM I-42.5 R – active mineral additives – microfillers – superplasticizer – hardening accelerators" for high-performance concretes with improved corrosion resistance.

Materials and methods

For the production of modified cementitious systems and high-performance concretes on their base, portland cement of general construction purpose CEM I-42.5 R of PJSC Ivano-Frankivskcement was used. In order to obtain the necessary flowability and strength of cementitious systems, along with polycarboxylate additive (PC), highly dispersed fly ash (5 % mass.) and highly dispersed quartz sand (5 % mass.), thus the modified composite portland cement (MCPC) (Ivashchyshyn, Sanytsky, Kropyvnytska & Rusyn, 2019). The cementitious systems CEM I-42.5 R and 5 % mass. limestone microfiller (LM), CEM I-42.5 R and 5 % mass. metakaolin (MK) were used for further studies of corrosion resistance.

Corrosion resistance of cementitious systems (beam samples 40×40×160 mm) was determined according to DSTU B 27677:2011 by changes in the strength of the samples after 6 months of their curing in a sodium sulfate solution (concentration $[SO_4^{2-}]$=10 g/l). This makes it possible to visually observe the destruction process and, in combination with strength tests, characterizes the behavior of concrete in aggressive water. Samples of cementitious systems were stored in water, after 28 days part of the samples was placed in water and the other part – in a solution of salts for further hardening. After 6 months, the coefficient of corrosion resistance (KR_6) was determined as the ratio between the strength of the samples cured in water to the strength of the samples cured in the sulfate environment. The binder is considered unstable in relation to an aggressive environment if the coefficient of stability $KR_6<0.90$.

Corrosion resistance of cementitious systems ($C:S$=1:2) was also determined according to the accelerated method by changing the strength of samples in a sulfate environment (concentration $[SO_4^{2-}]$=30 g/l). After one day of hardening, a part of the samples was placed into water with a temperature of +20 ºC, and the second part – in a Na_2SO_4 solution. Corrosion resistance was evaluated by the coefficient of corrosion resistance by flexural strength (CR), which is equal to the ratio of flexural strength after 8 weeks in an aggressive environment (CR_{agr}^8) to the flexural strength of samples hardened for 8 weeks in water (CR_{wat}^8) (Haufe, Vollpracht & Matschei, 2021).

Results and discussions

When carbon dioxide acts on concrete under normal conditions, the process of carbonization occurs. The concentration of CO_2 in the air is usually 0.02–0.03 %, and inside residential premises it can reach 0.1 %. Carbon dioxide carbonizes not only $Ca(OH)_2$, but can interact with about 85 % of the entire solid phase of concrete, including calcium hydrosilicates and hydroaluminates, magnesium hydroxide, etc (Shi, Zhao & Wan, 2015). Determination of the presence of unbound $Ca(OH)_2$ in concrete was determined by the colorimetric method (Looney, Leggs, Volz & Floyd, 2022), which consists in splitting the concrete in a given place and wetting the freshly formed surface of the chip with a 0.1 % alcoholic solution of phenolphthalein. In places where $Ca(OH)_2$ is bound, the concrete surface does not change color (Fig. 1, *a*), and in places where $Ca(OH)_2$ is not bound, the surface turns bright crimson (Fig. 1, *b*).

The research results of the resistance of cementitious systems to sulfate corrosion after 6 months are presented in the Table 1. The compressive strength of samples of cementitious systems based on highly dispersed fly ash and quartz sand, which were stored in a sulfate environment for 6 months, practically does not change, while the coefficient of corrosion resistance, calculated from the value of flexural strength, is CR^6_{flex}=1.1. The coefficient of corrosion resistance of the cementitious system based on CEM I-42.5 R without additives (*F*=125 mm) is 1.03. Highly dispersed active mineral additives and PC in the modified composite portland cement system (MCPC) ensures its increased density, which leads to an increase in corrosion resistance (CR^6_{flex}=1.1). The value of the coefficient CR^6_{flex}=1.1 for samples based on cementitious systems with modified composite portland cement (MCPC) and limestone microfiller (LM) indicates an increase in its resistance to aggressive environments.

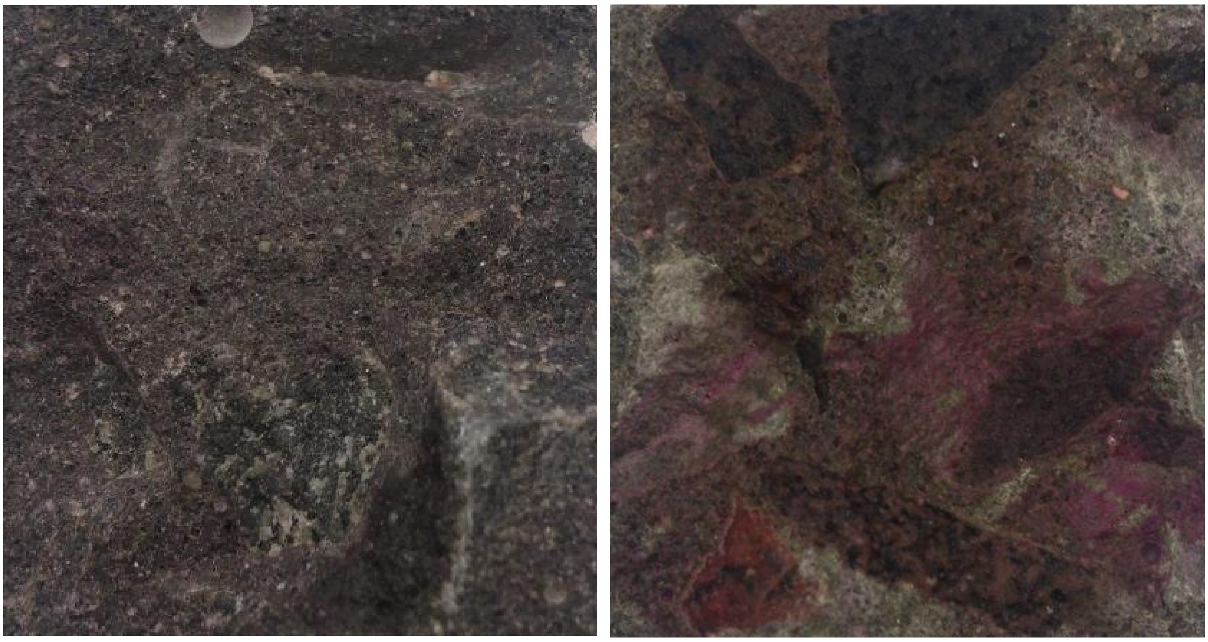
a *b*

Fig. 1. Determination of the presence of unbound Ca(OH)₂ in high-performance concrete (a) and plasticized concrete (b)

Table 1

Corrosion resistance of cementitious systems (C:S=1:2, 6 months)

Cementitous systems	*W/C*	Flow-ability, mm	Strenght, MPa				CR^6_{flex}	CR^6_{compr}
			H_2O		Na_2SO_4			
			flexural	comp-ressive	flexural	comp-ressive		
CEM I-42.5 R	0.39	125	3.7	62.8	3.8	64.6	1.03	1.03
CEM I-42.5 R + LM	0.39	235	11.4	78.4	10.5	70.0	1.10	1.11
MCPC	0.39	290	4.2	62.0	4.6	66.9	1.10	1.08

Corrosion resistance of cementitious systems (*C:S*=1:2) was also determined according to the accelerated method by changing the strength of samples in a sulfate environment (concentration $[SO_4^{2-}]$=30 g/l). After one day of hardening, a part of the samples was placed into water with a temperature of +20 ºC, and the second part – in a Na_2SO_4 solution. Corrosion resistance was evaluated by the coefficient of corrosion resistance by flexural strength (*CR*), which is equal to the ratio of flexural strength after 8 weeks in an aggressive environment ($CR_{Na_2SO_4}{}^8$) to the flexural strength of samples hardened for 8 weeks in water ($CR_{H_2O}{}^8$). As can be seen from the test results (Table 2), samples based on cementitious systems after storage for 8 weeks in an aggressive environment are characterized by an increase in compressive strength by 8–12 % and flexural strength by 3–9 % compared to the control samples. The coefficient of stability of samples without additives (*W/C*=0.39, *F*=125 mm) is CR^8_{flex}=0.63.

Table 2

Influence of mineral additives on corrosion resistance of cementitious systems (C:S=1:2, 8 weeks)

Cementitous systems	*W/C*	Flow-ability, mm	Strenght, MPa				CR^8_{flex}	CR^8_{compr}
			Na_2SO_4		H_2O			
			flexural	compressive	flexural	compressive		
CEM I-42.5 R	0.39	125	5.5	53.1	8.8	57.1	0.63	0.93
CEM I-42.5 R	0.48	200	4.0	29.6	6.9	32.5	0.58	0.91
CEM I-42.5 R + LM	0.39	235	11.4	78.4	10.5	70.0	1.09	1.12
CEM I-42.5 R + MK	0.39	210	7.7	62.5	9.0	65.1	0.86	0.96
MCPC	0.39	290	4.1	62.7	3.7	61.6	1.10	1.02

The increasing amount of mixing water (*W/C*=0.48, *F*=200 mm) provides increased porosity and a decrease in corrosion resistance (CR^8_{flex}=0.58) of concrete without additives when it was stored in a Na_2SO_4 solution, this can be explained by the formation of ettringite, an increasing in volume system and the appearance of internal stresses. The value of the coefficient of corrosion resistance of cementitious systems CR^8_{flex}=1.09–1.10 for high-performance concrete indicates an increase in the resistance of concrete to the influence of aggressive environments (CR^8_{flex}>0.70). High-performance concretes under normal hardening conditions are characterized by increased corrosion resistance (CR_6=1.1), and according to the value of compressive strength, they are classified as high-strength (class C 45/55).

The set of conducted studies allowed to substantiate the possibility of obtaining cementitious systems through a systematic combination of mineral and complex chemical additives with a plasticizing-accelerating effect, which ensures the directed formation of a dense structure of high-strength concrete with increased corrosion resistance.

Conclusion

Cementitious systems "Portland cement CEM I-42.5 R – active mineral additives – microfillers – superplasticizer – hardening accelerators" have been researched for its further application in high-performance concrete. Highly dispersed mineral additives of pozzolanic action ensure binding of portlandite in C-S-H phases, which contributes to the colmatations of pores with age of hardening, such a structure of cement stone in concrete ensures an increase in its corrosion resistance. The use of modified cementitious systems ensures the production of concretes which are characterized by increased compressive strength (class C45/55) and high corrosion resistance (CR_6=1.1).

Prospects for further research

The use of cementitious systems for high-performance concrete with increased corrosion resistance will contribute to the design and construction of buildings and structures which are under the influence of aggressive environments, such as: underground and industrial wastewater containing chlorides and sulfates. To increase the efficiency and durability of high-performance concretes based on modified cementitious systems, it is advisable to continue research of cementitious systems with highly dispersed additives and concretes on their base according to EN 206-1 exposure classes XD, XS, XF.

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І. І. Кіракевич, Б. Г. Русин, Ю. Б. Бобецький
Національний університет “Львівська політехніка”,
кафедра будівельного виробництва

ЦЕМЕНТУЮЧІ СИСТЕМИ ДЛЯ ВИСОКОФУНКЦІОНАЛЬНИХ БЕТОНІВ З ПІДВИЩЕНОЮ КОРОЗІЙНОЮ СТІЙКІСТЮ

Наведено результати дослідження цементуючих систем “портландцемент ПЦ I-500 Р-Н – активні мінеральні добавки – мікронаповнювачі – суперпластифікатор – прискорювачі тверднення” для одержання високофункціональних бетонів підвищеної корозійної стійкості. Досліджено стійкість цементуючих систем до корозії, спричиненої впливом хімічних речовин – сульфатної корозії (клас ХА), що об’єднує процеси утворення та накопичення малорозчинних солей та зумовлює збільшення об’єму із переходом у тверду фазу з утворенням кристалів і супроводжується внутрішніми напруженнями та деструктивними явищами у бетоні.

Корозійну стійкість цементуючих систем визначали згідно з ДСТУ Б 27677:2011 за зміною міцності зразків після шести місяців їх зберігання у середовищі натрію сульфату (концентрація $[SO_4^{2-}]$=10 г/л). Значення коефіцієнта KC_6=1,1 для високофункціональних бетонів на основі цементуючих систем свідчить про підвищення його стійкості до агресивних середовищ. Корозійну стійкість також визначали згідно з прискореною методикою за коефіцієнтом міцності при згині, що дорівнює відношенню міцності на згин зразків після витримування вісім тижнів у сульфатному середовищі (концентрація $[SO_4^{2-}]$ = 30 г/л) до міцності на згин зразків, що тверднули вісім тижнів у воді. Зростання корозійної стійкості зразків на основі цементуючих систем на 2–12 % пояснюється створенням щільної та дрібнопористої мікроструктури. Високодисперсні добавки пуцоланової дії забезпечують зв’язування портландиту в C-S-H фази, що сприяє кольматації пор із віком тверднення та забезпечує підвищення корозійної стійкості цементного каменю у бетоні. Застосування цементуючих систем для високофункціональних бетонів із підвищеною корозійною стійкістю сприятиме будівництву споруд, що перебувають під впливом агресивних середовищ, зокрема підземних та промислових стічних вод, які містять хлориди та сульфати.

Ключові слова: високофункціональні бетони; полікарбоксилати; високодисперсні мінеральні добавки; цементуючі системи; карбонізація; корозійна стійкість.