Досліджено вплив співвідношення основних оксидів лужних гідроалюмосилікатів складу (0,7÷1Na_2O+ +0÷0,3K_2O)·Al_2O_3·(2÷7)SiO_2·nH_2O та температури твердіння 20÷80 °С на процеси їх структуроутворення. При твердінні за нормальних умов оптимальною структурною формулою лужного гідроалюмосилікату є (0,2K_2O+0,8Na_2O)·4,5SiO_2·Al_2O_3·nH_2O, що дозволяє отримувати водостійкий штучний камінь за рахунок синтезу гідратних новоутворень цеолітоподібних мінералів типу: цеоліту Na–A; натрієвого та калієвого гейландиту, а також калій-натрієвого філліпситу

Ключові слова: лужний гідроалюмосилікат, співвідношення основних оксидів, температура твердіння, фазовий склад, структуроутворення

Исследовано влияние соотношения основных оксидов щелочных гидроалюмосиликатов состава (0,7÷ ÷1Na_2O+0÷0,3K_2O)·Al_2O_3·(2÷7)SiO_2·nH_2O и температуры твердения 20÷80 °С на процессы их структурообразования. При твердении в нормальных условиях оптимальной структурной формулой щелочного гидроалюмосиликата является (0,2K_2O+0,8Na_2O)·4,5SiO_2× ×Al_2O_3·nH_2O, что позволяет получать водостойкий искусственный камень за счет синтеза гидратных новообразований цеолитоподобных минералов типа: цеолита Na–A; натриевого и калиевого гейландита, а также калий-натриевого филлипсита

Ключевые слова: щелочной гидроалюмосиликат, соотношение основных оксидов, температура твердения, фазовый состав, структурообразование

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INFLUENCE OF THE RATIO OF OXIDES AND TEMPERATURE ON THE STRUCTURE FORMATION OF ALKALINE HYDRO-ALUMINOSILICATES

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1. Introduction

It is known that properties of building composite materials depend on the efficiency of fulfilling functions of each of the components of a composite, matrix and its fillers [1]. Control over these properties is possible through optimization of formulation of a composite matrix, correct choice of fillers and technology of creation of a composite material.

Compositional matrix at the stage of formation a composite is a binding substance and at the stage of operation – artificial conglomerate, which it forms. Properties of cement composites depend first of all on the type of binding substance and operational properties of artificial stone that formed in the process of structure formation and is characterized by its phase composition.

For the first time in paper [2], the possibilities to obtain artificial stone based on the compounds of alkaline metals and aluminosilicate components were theoretically substantiated and theoretically proven. By nature, the processes of their condensation and hydration do not differ from silicate systems of the Earth's crust. The difference is in terms of activation of original waterless mineral substances. In the first case, the activation is achieved by artificial treatment or the action of active substances, in the second – due to the action of solar energy and energy of atmospheric reagents.

The research conducted by scientific school of the V. D. Glukhovsky NDIVM (Kiev, Ukraine) [3] proved that not only the processes of structure formation of alkaline aluminosilicates are close to natural, but also the products of their hydration that are the analogues of natural zeolites. Scientific papers of international groups of authors [4, 5] contain a study of properties of alkaline aluminosilicates such as high resistance to aggressive environment and effect of high temperatures. It is the phase composition of the matrix that is the basis for providing high performance and special properties of alkaline aluminosilicates.

Therefore, a study of the influence of temperature of structure formation and the ratio of basic oxides of alkaline aluminosilicates on the properties and phase composition of artificial stone is a relevant issue of modern materials science in the direction of expanding scope of their use.

2. Literature review and problem statement

As a result of research and study of the processes of interaction of clay minerals with alkaline reagents, it was determined that under normal conditions of hardening, artificial stone in softened in water medium, that is, it is not waterproof. To provide for its water resistance, long temperature processing is required [6].

Usually, to obtain artificial stone based on alkaline aluminosilicates at low temperatures (<100 °C) and atmospheric pressure, they use aqueous silicate solutions of sodium or potassium and metakaolin. Alkaline aluminosilicates at early stages of hardening are presented by the N(K)–A–S–H gel, as well as active anionic groups in the form of tetrahedrons $[AlO_4]^{5-}$ and $[SiO_4]^{4-}$. Tetrahedrons $[AlO_4]^{5-}$ and $[SiO_4]^{4-}$ in the presence of alkaline solution form more complex systems with a common apex – atom of oxygen in the course of condensation compensate for its valence with transition into the state with minimum reserve of free energy [7]. The final products of new formations are zeolite-like alkaline hydro-aluminosilicates of the $N(K)AS_xH_y$ type.

Control over both the process of structure formation and operational properties of alkaline hydro-aluminosilicates is possible by using variation of the ratio of basic oxides of a binding substance, dispersity of mineral particles and hardening conditions. Papers [8, 9] consider influence of the ratio of basic oxides of a binder on the properties and phase composition under conditions of hardening of artificial stone at temperatures 20 °C, 85 °C and autoclave treatment, but the authors did not examine the effect of temperatures in the range of 20÷85 °C on the properties and phase composition of alkaline hydro-aluminosilicates.

Research into influence of composition of the alkaline component and temperature of structure formation of alkaline hydro-aluminosilicates based on the ash burning is represented in articles [10, 11]. Improvement in physical and mechanical properties of artificial stone was noted when using mixed sodium-potassium alkaline component, as well as at elevated temperature of its hardening.

Study of influence of temperature processing of alkaline hydro-aluminosilicate on the physical-mechanical properties of artificial stone is presented in paper [12]. Optimum processing temperature of alkaline aluminosilicate was determined without changing its structural formula; effect of phase composition of stone is not considered.

Article [13] presented results of research into influence of temperature of the initial hardening (20÷100 °C) on mechanical properties and porous structure of artificial stone, obtained on the base of burnt kaolin. Study of influence of phase composition of alkaline hydro-aluminosilicate on the properties of stone was not given.

Examination of influence of the ratio of oxides of alkaline hydro-aluminosilicates and temperature of their hardening on the properties of artificial stone is presented in the papers [14–17]. It is indicated that for high operation characteristics of stone from alkaline aluminosilicate there is a need for increased temperatures of hardening.

In order to expand areas of use alkaline aluminosilicate binders, are promising research of phases artificial stone alkaline hydro-aluminosilicate depending on the ratio of oxides at normal and increased temperatures 20÷80 °C of hardening, as well their influence on its properties.

3. Goal and tasks of the study

The goal of this work is to study influence the ratio of oxides of alkaline hydro-aluminosilicate and temperature of hardening on the phase composition of artificial stone.

To accomplish the goal, the following tasks were solved:

– to determine effect of the ratio of oxides of alkaline hydro-aluminosilicate on the process of structure formation and the properties of artificial stone in the temperature range of 20÷80 °C;

– to determine the optimal ratio oxides of alkaline hydro-aluminosilicate with the possibility of forming the water-resistant structure of stone after hardening at normal temperatures.

4. Materials and methods of the study

To obtain alkaline aluminosilicate of basic structural formula $(0{,}7÷1Na_2O+0÷0{,}3K_2O)\cdot Al_2O_3\cdot(2÷7)SiO_2\cdot nH_2O$, we used: metakaolin, microsilica, and sodium liquid glass; if necessary, potassium or additional sodium component was introduced with the help of aqueous solutions of their hydroxides.

The ratio of basic oxides was considered within the range: K_2O/Al_2O_3=0÷0,3, Na_2O/Al_2O_3=0,7÷1 (under condition R_2O/Al_2O_3=1) and SiO_2/Al_2O_3=2÷7. The value of the ratio H_2O/Al_2O_3 was adopted as permanent to ensure identical viscosity of each composition of hydro-aluminosilicates (18 cm by the Suttard's viscometer). Calculation of the ratio of basic oxides was carried out by taking into account recommendations [18, 19].

Structure formation of alkaline aluminosilicates occurred in two stages. The first – initial hardening at temperature of 20 °C, 40 °C, 60 °C and 80 °C lasted for 12 hours. The second stage is the continuation of hardening for 27,5 days at temperature of 20 °C.

As the criteria for evaluation of the properties of artificial stone based on alkaline hydro-aluminosilicates we selected its strength at compression (R_c) and water resistance (softening coefficient, C_s). Softening coefficient describes the ratio of strength of artificial stone at compression, kept in water (full water saturation), to the strength of the stone under dry conditions.

Determining phase composition of the products of hydration of alkaline hydro-aluminosilicates was studied by using X-ray diffraction, differential thermal analysis and electron microscopy.

X-ray diffraction analysis was carried out at the diffractometer DRON-4-07 with a copper tube at voltage of kV, current 10...20 mA and the range of angles 2θ=10...60°. Identification of the new formations was carried out based on the data, as well as using the database PDF-2 Data Base (Sets 1–50 plus 70–88) with the software module JCPDFWIN 2.1 (JCPDS-ICDD, 2000). Comprehensive differential thermal analysis was carried out at the derivatograph (Paulik-Paulik-Erdey) made by MOM (Budapest). To identify results of thermal methods of research, we used data of ref. [18, 19].

5. Results of the study

Fig. 1, 2 demonstrate radiographs and electronic photographs of microstructure of cleaved fragment of artificial stone depending on the ratio SiO_2/Al_2O_3=2÷7 in the original composition of alkaline hydro-aluminosilicate (under condition K_2O/R_2O=0,15 та $(Na_2O+K_2O)/Al_2O_3$=1) after hardening temperature of 80 °C.

Phase composition of hydrated new formations at low ratios SiO_2/Al_2O_3=2÷3 after hardening at temperature 80 °C is characterized by zeolite-like new formations of the type: Na–A of zeolite (d/n=0,699; 0,365; 0,336; 0,293 nm), natrolite (d/n=0,287; 0,243; 0,138 nm), usingite (d/n=0,492; 0,347; 0,295 nm). Electronic photographs of microstructure of the cleaved fragment of stone display existence of amorphous phases of alkaline hydro-aluminosilicates and particles of non-reacted metakaolin (Fig. 2).

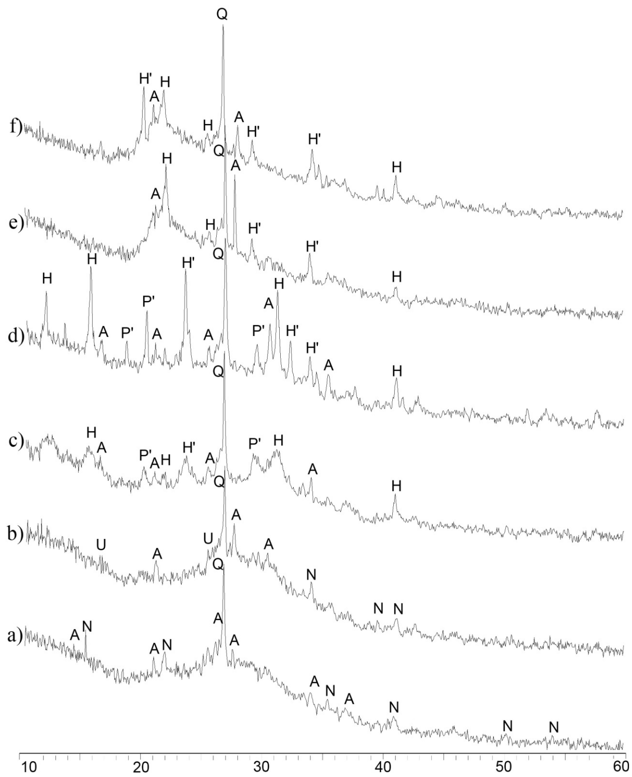


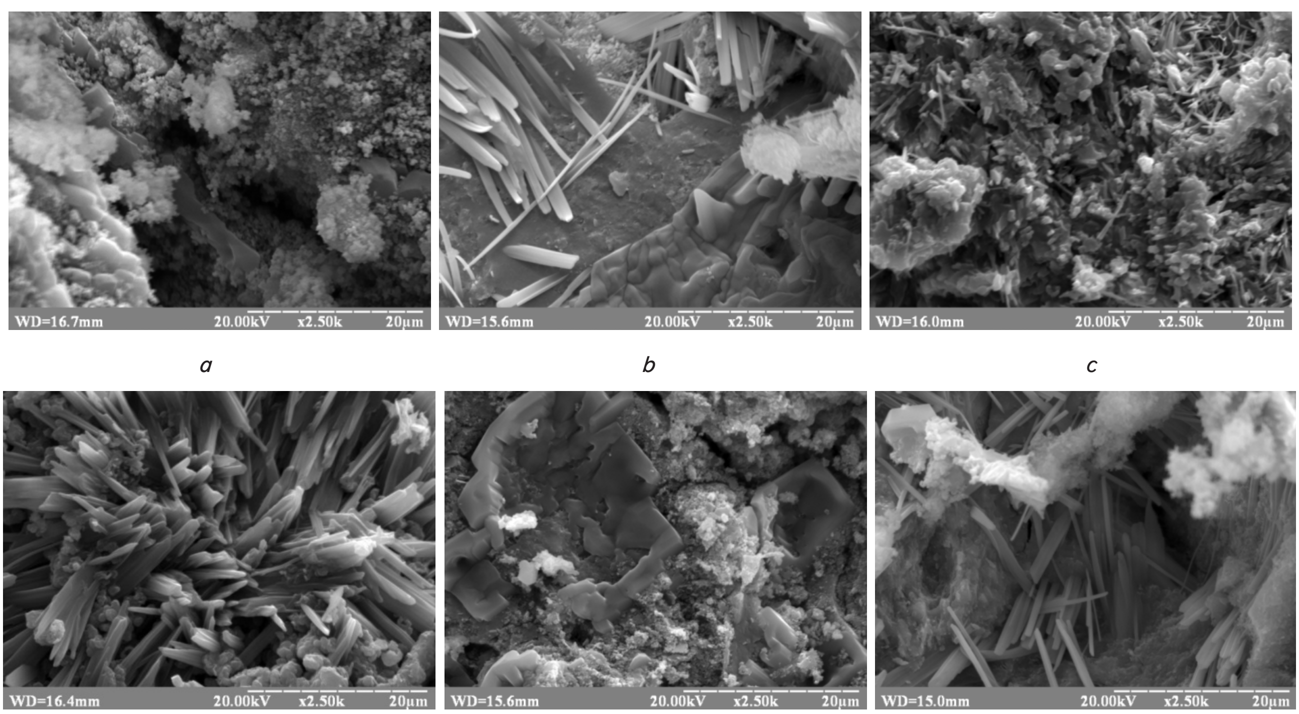
Fig. 1. X-ray of artificial stone after hardening at temperature 80°C, obtained at the ratio K_2O/R_2O=0,15 and SiO_2/Al_2O_3, respectively: *a* – 2; *b* – 3; *c* – 4; *d* – 5; *e* – 6; *f* – 7. Legend: Q – quartz; N – natrolite; A – zeolite Na–A; U – usingite; P' – Na–K phillipsite; H – Na heulandite; H' – K heulandite

Alkaline hydro-aluminosilicates with the ratio SiO_2/Al_2O_3=4÷5 are characterized by the presence of zeolite-like new formations of the type: Na–A zeolite (d/n=0,699; 0,365; 0,336; 0,293 nm), sodium heulandite (d/n=0,509; 0,392; 0,296 nm), potassium heulandite (d/n=0,342; 0,281; 0,273 nm) and sodium-potassium phillipsite (d/n=0,498; 0,408; 0,269 nm). Crystallinity of the structure is rather high, which is indicated by the largest intensity of diffraction bursts in the radiographs (Fig. 1) and electronic photographs of the microstructure of artificial stone (Fig. 2).

Fig. 3, 4 demonstrate radiographs and electronic photographs of the microstructure of the cleaved fragment of artificial stone after hardening at a temperature of hardening 80 °C, obtained at the ratio K_2O/R_2O=0÷0,3, $(Na_2O+K_2O)/Al_2O_3$=1 and SiO_2/Al_2O_3=5. At the ratio SiO_2/Al_2O_3=5, phase composition of artificial stone is characterized by zeolite–like new formations of the heulandite and phillipsite type (Fig. 3), and the microstructure of artificial stone displays a large number of subcrystal phases (Fig. 4).

Alkaline hydro-aluminosilicates (at K_2O/R_2O=0) is characterized by the presence in the phase composition of the following zeolite-like new formations of the type: natrolite, zeolite Na–A and Na heulandite. When potassium ions replace sodium ions, there are additional zeolite-like new formations created of the type of potassium heulandite and sodium-potassium phillipsite, the presence in new formations of the natrolite phase is not observed, crystallinity of the products of hydration increases.

Fig. 5 displays diagrams of influence of the ratio of oxides of alkaline hydro-aluminosilicates on the strength and water resistance of artificial stone after hardening at temperature 80 °C.



a *b* *c*

d *e* *f*

Fig. 2. Microphotographs of the surface of the cleaved fragment of artificial stone after hardening at temperature 80 °C at the ratio K_2O/R_2O=0,15 and SiO_2/Al_2O_3, respectively: *a* – 2; *b* – 3; *c* – 4; *d* –5; *e* – 6; *f* – 7 (x2500 times)

The highest indicators of strength and water resistance characterize hydro-aluminosilicates with the ratio SiO_2/Al_2O_3=4,5÷6,5 and K_2O/R_2O=0,15÷0,25. At the ratio SiO_2/Al_2O_3>5, strength of the stone is slightly reduced. Introduction of potassium ions to the composition of hydro-aluminosilicate improves strength characteristics and water resistance of stone.

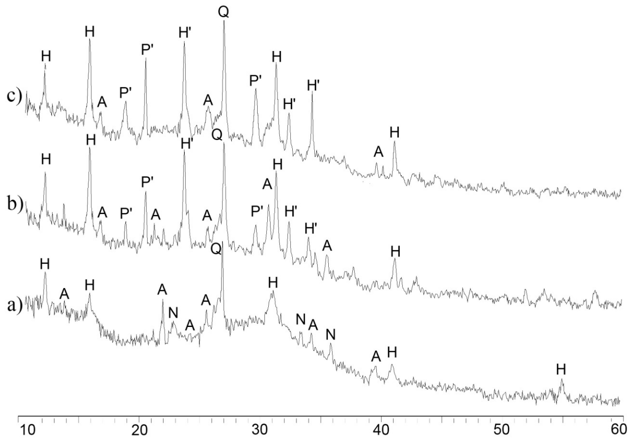
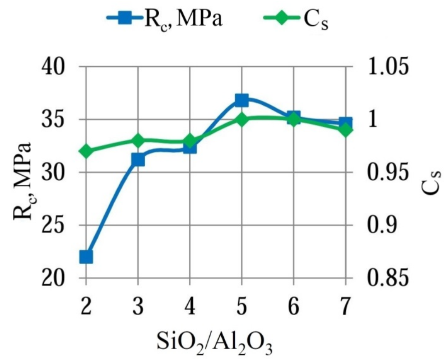
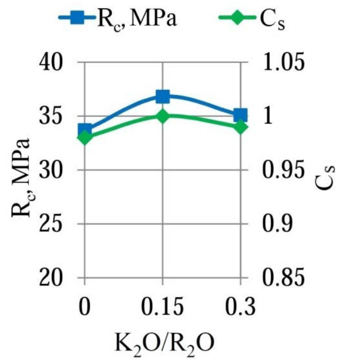


Fig. 3. X-ray of artificial stone after hardening at temperature 80 °C, obtained at the ratio SiO_2/Al_2O_3=5 and K_2O/R_2O, respectively: *a* – 0; *b* – 0.15; *c* – 0.3. Legend: Q – quartz; N – natrolite; A – zeolite Na–A; P' – Na–K of phillipsite; H – Na heulandite; H' – K heulandite



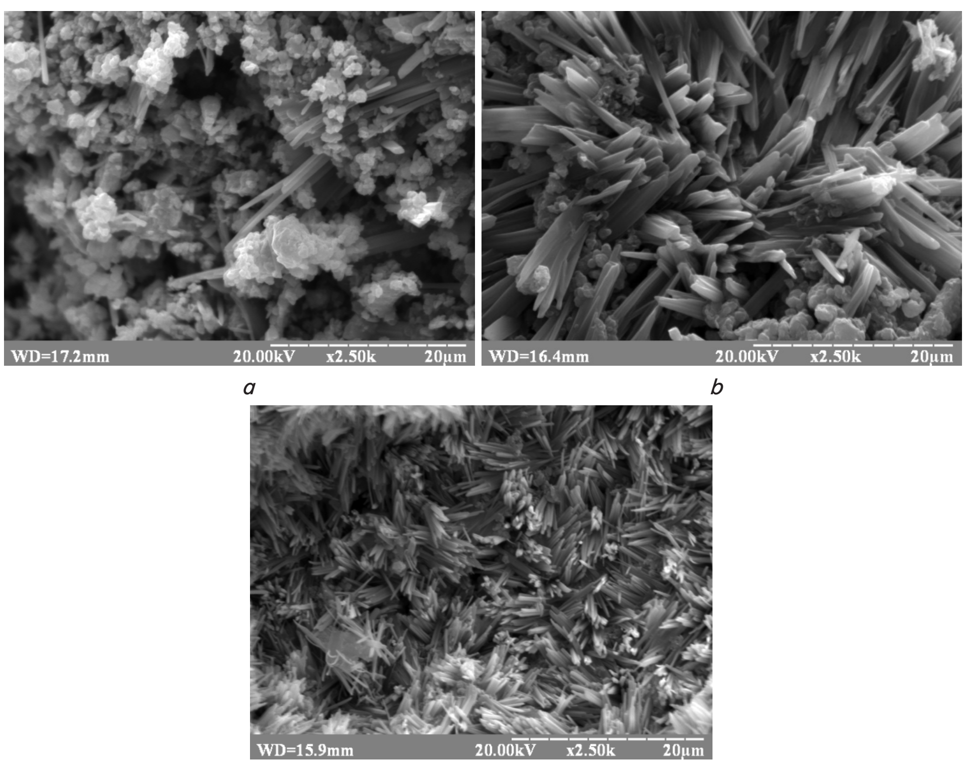
a



b

Fig. 5. Influence of the ratio of hydro-aluminosilicate oxides on strength at compression and water resistance of artificial stone after hardening at temperature 80 °C, respectively: *a* – SiO_2/Al_2O_3, at K_2O/R_2O=0,15; *b* – K_2O/R_2O, at SiO_2/Al_2O_3=5

Radiographs and electronic photographs of microstructure of the cleaved fragment of artificial stone depending on the ratio SiO_2/Al_2O_3=2÷7 (under condition K_2O/R_2O=0,15 and $(Na_2O+K_2O)/Al_2O_3$=1) after hardening at temperature 20 °C are displayed in Fig. 6, 7.



a *b*

c

Fig. 4. Electronic photographs of the microstructure of the cleaved fragment of artificial stone after hardening at temperature 80 °C, obtained at the ratio SiO_2/Al_2O_3=5 and K_2O/R_2O, respectively: *a* – 0; *b* – 0.15; *c* – 0.3 (x2500 times)

According to X-ray diffraction analysis, phase composition of artificial stone after hardening at temperature 20 °C, obtained at the ratio SiO_2/Al_2O_3=2÷3, is characterized by zeolite-like new formations of the type: Na–A of zeolite, natrolite and usingite (Fig. 6).

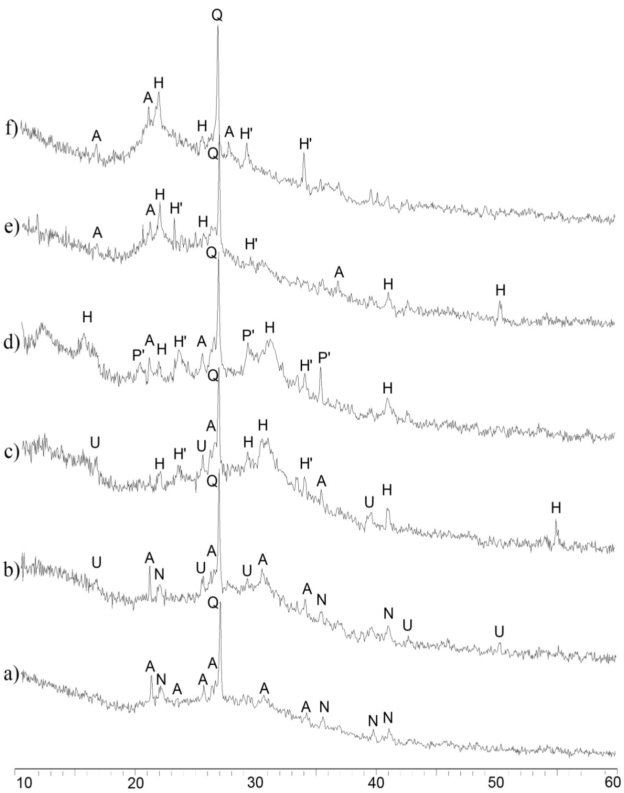


Fig. 6. X-ray of artificial stone after hardening at temperature 20 °C, obtained at the ratio K_2O/R_2O=0,15 and SiO_2/Al_2O_3, respectively: *a* – 2; *b* – 3; *c* – 4; *d* – 5; *e* – 6; *f* – 7. Legend: Q – quartz; N – natrolite; A – zeolite Na–A; U – usingite; P' –Na–K of phillipsite; H – Na heulandite; H' – K heulandite

Electronic photographs of microstructure of the cleaved fragment of artificial stone display noticeable presence of a large number of particles of the non-reacted metakaolin (Fig. 7).

Alkaline aluminosilicates with the ratio of oxides SiO_2/Al_2O_3=4 form zeolite-like new formations of the type: Na–A of zeolite, sodium heulandite, potassium heulandite and usingite (Fig. 6). When increasing the ratio of SiO_2/Al_2O_3 to 5, there are zeolite-like new formations of the type Na–K of phillipsite. At SiO_2/Al_2O_3>5, phase composition is characterized by zeolite-like formations of the heulandite type (Fig. 6). Registered new formations are confirmed by the data of electronic microscopy (Fig. 7).

Fig. 8, 9 present radiographs and electronic photographs of microstructure of the cleaved fragment of artificial stone depending on the ratio K_2O/R_2O=0÷0,3 in the original composition of alkaline hydro-aluminosilicate (under condition $(Na_2O+K_2O)/Al_2O_3$=1 and SiO_2/Al_2O_3=5) after normal conditions of hardening (W=65 % and T=20 °C).

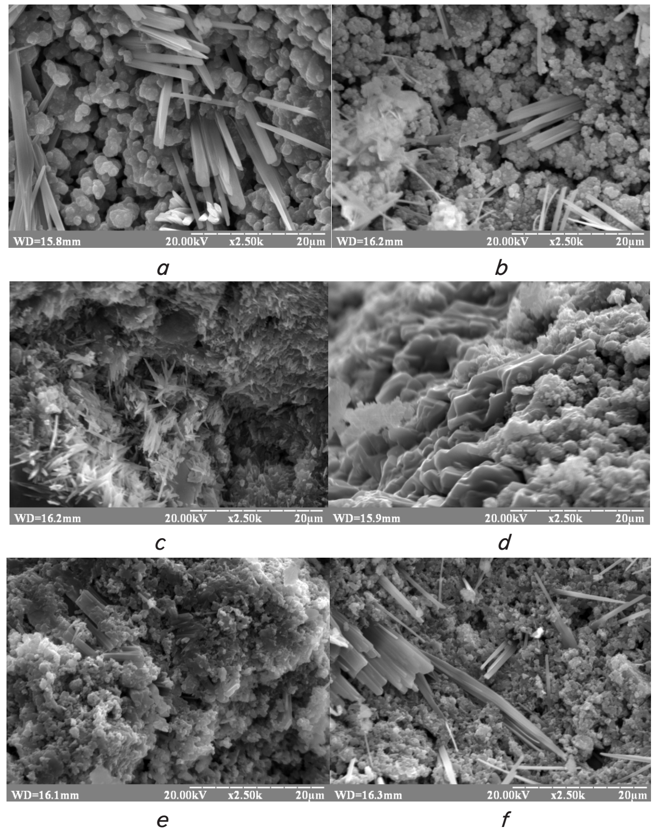


Fig. 7. Electronic photographs of microstructure of artificial stone after hardening at temperature 20 °C, obtai ned at the ratio K_2O/R_2O=0,15 та SiO_2/Al_2O_3, respectively: *a* – 2; *b* – 3; *c* – 4; *d* – 5; *e* – 6; *f* – 7 (×2500 times)

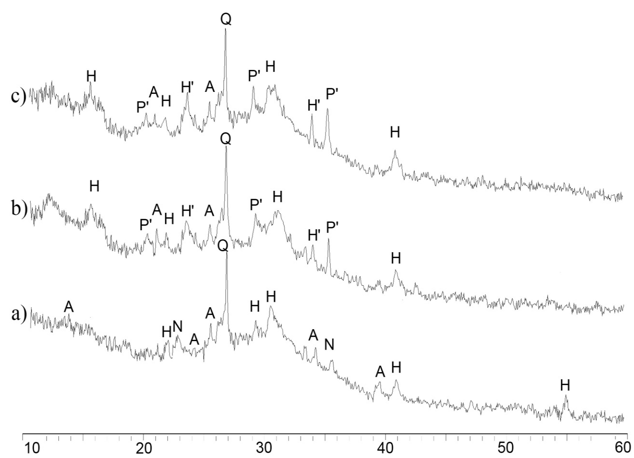
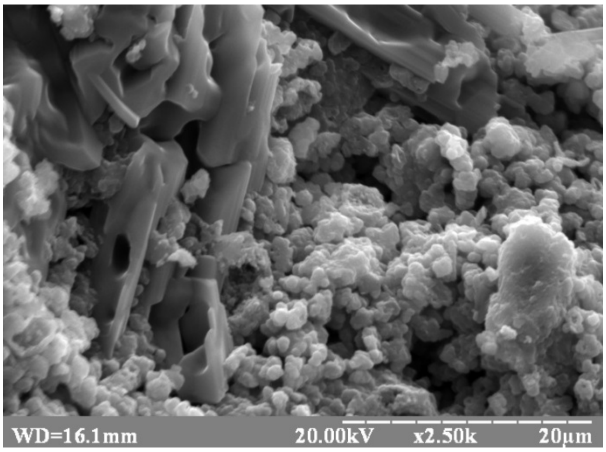
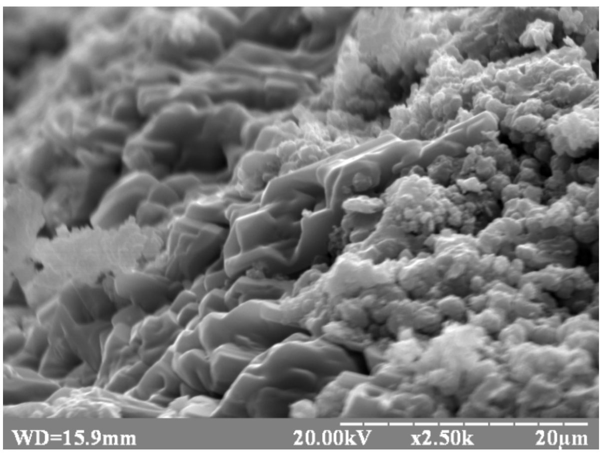


Fig. 8. X-ray of artificial stone after hardening at temperature 20 °C, obtained at the ratio SiO_2/Al_2O_3=5 and K_2O/R_2O, respectively: *a* – 0; *b* – 0.15; *c* – 0.3. Legend: Q – quartz; N – natrolite; A – zeolite Na–A; P' – Na–K of phillipsite; H – Na heulandite; H'– K heulandite

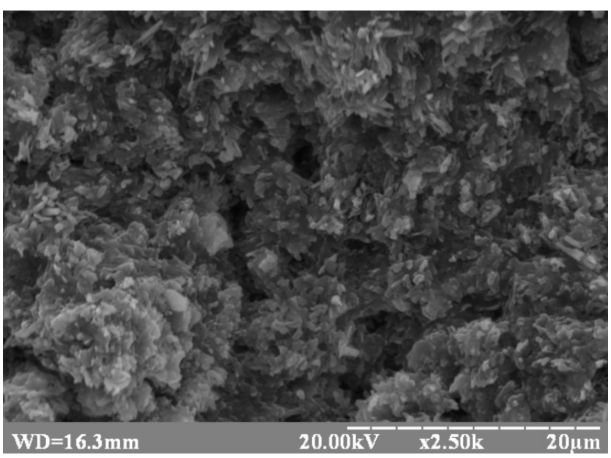
When introducing potassium ions into the composition of alkaline component of hydro-aluminosilicate, the phase composition of artificial stone is characterized by the presence of zeolite-like new formations of the type: zeolite Na–A, sodium and potassium heulandite, Na–K of phillipsite, crystallinity of the products of hydration is increased, which is confirmed by electronic photographs of microstructure of stone (Fig. 9) and the intensity of diffraction bursts in radiographs (Fig. 8).



a



b



c

Fig. 9. Electronic photographs of microstructure of the cleaved fragment of artificial stone after hardening at temperature 20 °C, obtained at the ratio SiO_2/Al_2O_3=5 and K_2O/R_2O, respectively: *a* – 0; *b* – 0.15; *c* – 0.3 (×2500 times)

Influence of the ratio of hydro-aluminosilicate oxides on the properties of artificial stone after hardening at temperature 20 °C is displayed in Fig. 10. The highest compression strength indices characterize hydro-aluminosilicates with the ratio SiO_2/Al_2O_3=3,5÷5,0 and K_2O/R_2O=0,15÷0,25. Water resistance of artificial stone increases at SiO_2/Al_2O_3= =4,0÷5,5 and when introducing potassium ions into the composition of alkaline hydro-aluminosilicate in the amount of K_2O/R_2O=0,15÷0,3.

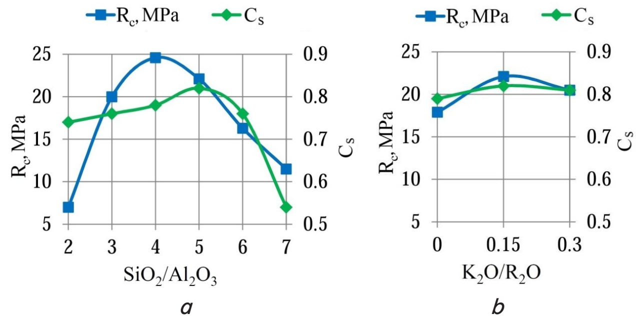


Fig. 10. Influence of the ratio of hydro-aluminosilicate oxides on strength at compression and water resistance of artificial stone after hardening at temperature 20 °C, respectively: *a* – SiO_2/Al_2O_3, at K_2O/R_2O=0,15; *b* – K_2O/R_2O, at SiO_2/Al_2O_3=5

Results of the study of influence of the temperature of hardening on the phase composition of artificial stone based on alkaline hydro-aluminosilicate are shown in Fig. 11, 12.

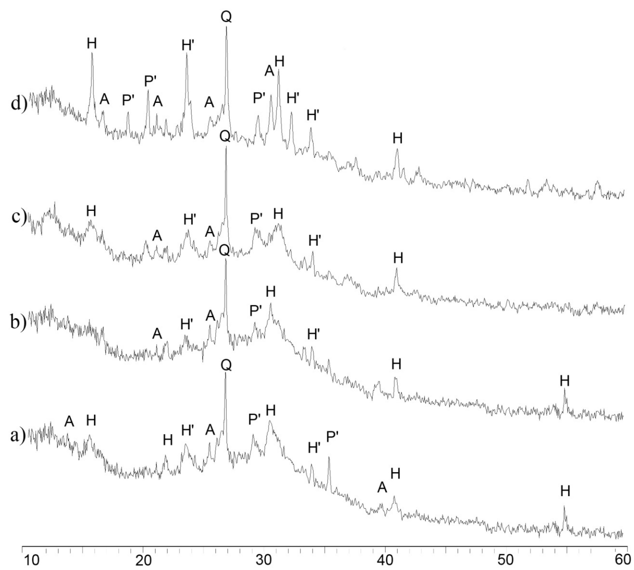
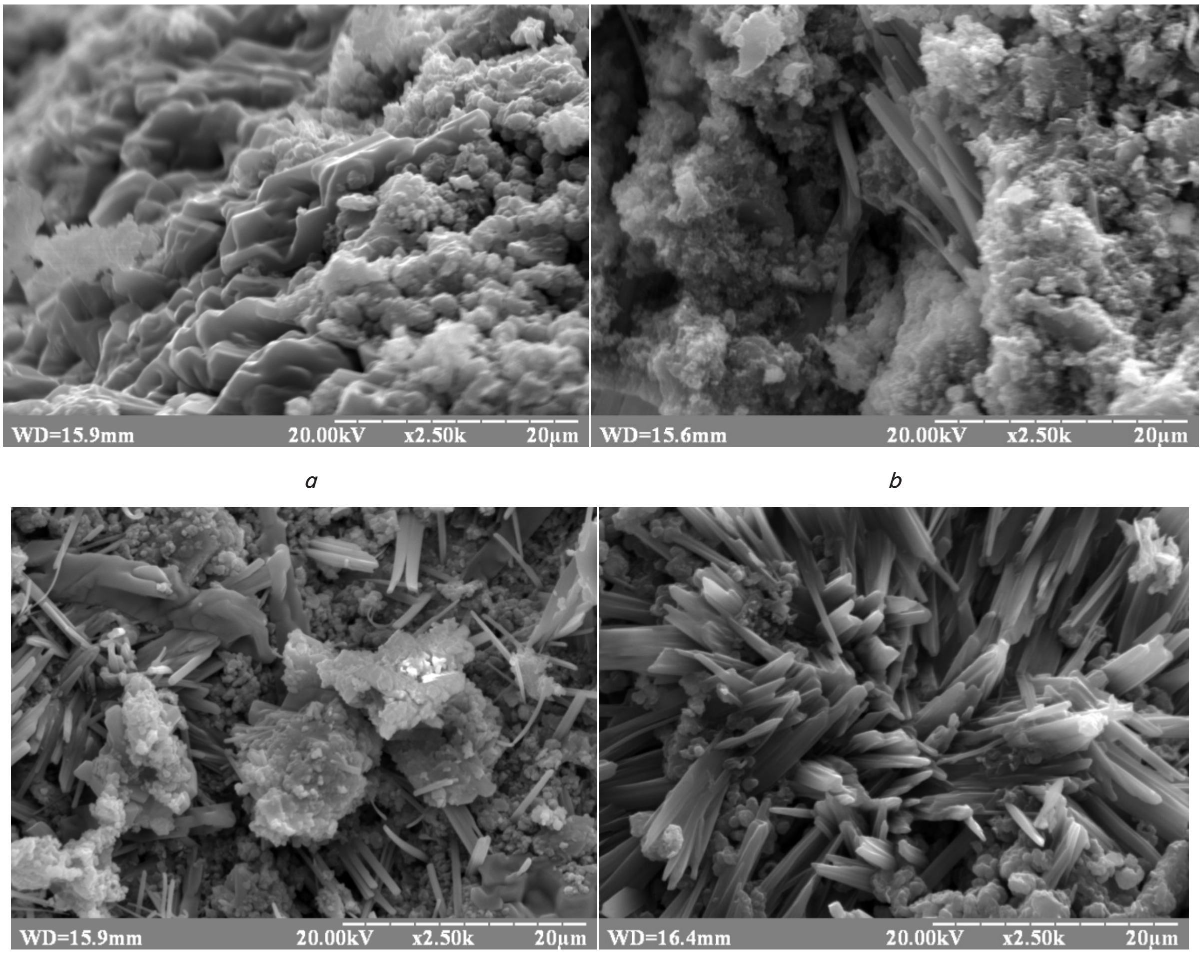


Fig. 11. X-ray of artificial stone obtained at the ratio K_2O/R_2O=0,15 and SiO_2/Al_2O_3=5, after hardening at temperatures, °C: *a* – 20; *b* – 40; *c* – 60; *d* – 80. Legend: Q – quartz; A – zeolite Na–A; P'– Na–K phillipsite; H – Na heulandite; H' – K heulandite

It was defined that phase composition of alkaline hydro-aluminosilicate at the ratio of basic oxides K_2O/R_2O= =0,15 and SiO_2/Al_2O_3=5 is characterized by the presence of zeolite-like new formations of the type: Na–A zeolite, potassium and sodium heulandite, as well as sodium-potassium phillipsite (Fig. 11). The process of structure formation of artificial stone accelerates with rising temperature without affecting the phase composition, but increasing their degree of crystallinity (Fig. 12).



a *b*

c *d*

Fig. 12. Electronic photographs of microstructure of artificial stone, obtained at the ratio K_2O/R_2O=0,15 and SiO_2/Al_2O_3=5, after hardening at temperatures, °C: *a* – 20; *b* – 40; *c* – 60; *d* – 80 (×2500 times)

Fig. 13 displays diagram of the influence of temperature of hardening of alkaline hydro-aluminosilicate on the properties of artificial stone based on it. At the ratio SiO_2/Al_2O_3=5 and K_2O/R_2O=0,15, a substantial increase in strength occurs at temperature of hardening 40÷80 °C. The highest water resistance characterizes artificial stone after thermal treatment at temperature 60÷80 °C.

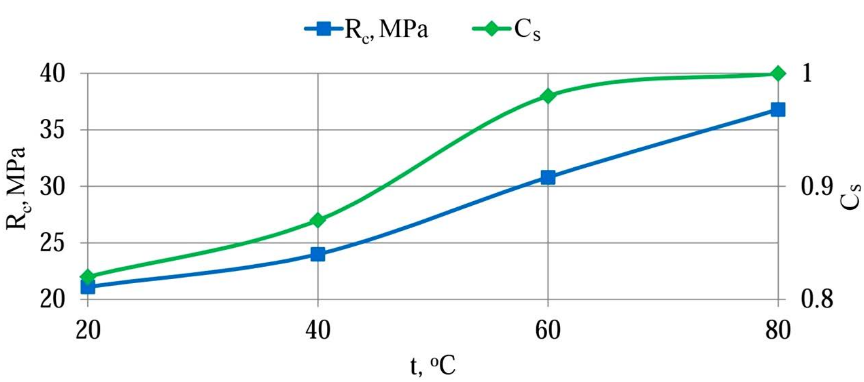


Fig. 13. Influence of temperature of hardening of artificial stone on strength at compression and water resistance of artificial stone after hardening at temperature in the range 20÷80°C at SiO_2/Al_2O_3=5 and K_2O/R_2O=0,15

6. Discussion of results of research into effect of the ratio oxides of alkaline hydro-aluminosilicates on phase composition and properties of artificial stone based on them

In the course of optimization of compositions of alkaline hydro-aluminosilicates of general structural formula – $(0,7÷1Na_2O+0÷0,3K_2O)·Al_2O_3·(2÷7)SiO_2·nH_2O$, it was revealed that phase composition of artificial stone depends mainly on the ratio of oxides, temperature of hardening in the range of 20÷80 °C increases the rate of structure formation of zeolite-like hydro-aluminosilicates.

A determining factor for the type of hydrated new formations is the ratio SiO_2/Al_2O_3; when it is increased, zeolite-like phases are formed with a larger amount of silica in the crystal lattice. At hardening of alkaline hydro-aluminosilicates of the above mentioned structural types under normal conditions of hardening, providing the highest degree of structure crystallinity, the ratio SiO_2/Al_2O_3=4÷5 is optimal. Hydro-aluminosilicates with such a ratio of oxides are characterized also by the highest indicators of strength and water resistance of artificial stone based on them.

Introduction of potassium ions into the composition of hydro-aluminosilicate helps to obtain potassium and sodium-potassium zeolite-like new formations and to increase degree of crystallinity of the specified phases. To accelerate structure formation of alkaline hydro-aluminosilicate under normal conditions of hardening, the introduction of potassium oxide is required at K_2O/R_2O=0,15÷0,3. Introduction of potassium ions into the composition of alkaline hydro-aluminosilicates also contributes to the increase in water resistance and strength characteristics of artificial stone, regardless of the temperature of its hardening.

When the temperature of hardening of alkaline hydro-aluminosilicate rises from 20 to 80 °C, phase composition of artificial stone does not practically change, however, this leads to the increased speed of structure formation and the degree of crystallinity of artificial stone.

As a result of analysis of influence of the ratio of oxides of alkaline hydro-aluminosilicate on the properties and composition of artificial stone, we determined its optimum structural formula $(0,2K_2O+0,8Na_2O)\cdot 4,5SiO_2\cdot Al_2O_3\cdot nH_2O$, which makes it possible under normal conditions of hardening to obtain water-resistant artificial stone by synthesis in the composition of hydrated new formations of zeolite-like minerals of the type: zeolite Na–A; sodium and potassium heulandite, as well as potassium-sodium phillipsite (Fig. 14).

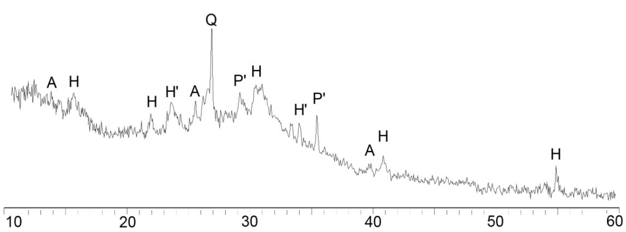


Fig. 14. X-ray of artificial stone based on alkaline hydro-aluminosilicate of the form $(0,2K_2O+0,8Na_2O)\cdot 4,5SiO_2\cdot Al_2O_3\cdot nH_2O$ after hardening at temperature 20 °C for 90 days. Legend: Q — quartz; A — zeolite Na—A; P'— Na—K of phillipsite; H — Na heulandite; H' — K heulandite

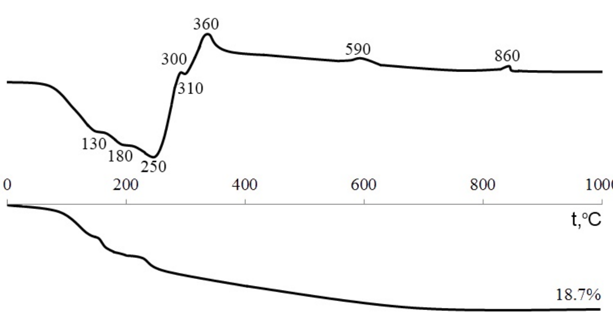


Fig. 15. Derivatogram of artificial stone based on alkaline hydro-aluminosilicate of the form $(0,2K_2O+0,8Na_2O)\cdot 4,5SiO_2\cdot Al_2O_3\cdot nH_2O$ after hardening at temperature 20 °C for 90 days

Results of X-ray phase analysis are confirmed by the data of DTA. The DTA curve recorded stepped dehydration of artificial stone to 250 °C that is typical for zeolite-like new formations of the heulandite and phillipsite type, which is confirmed also by endo-effect at 310 °C. Exo-effects (+) at temperatures 360, 590 and 860 °C indicate the presence of zeolite-like new formations of the type Na–A in the phase composition of stone (Fig. 15).

Electronic photographs of microstructure demonstrate dense structure of artificial stone; there are subcrystal phases and a large number of crystalline new formations. Heulandite-like new formations are characterized by crystals in the form of prisms-plates, phillipsite-like – by cruciform crystals (Fig. 16).

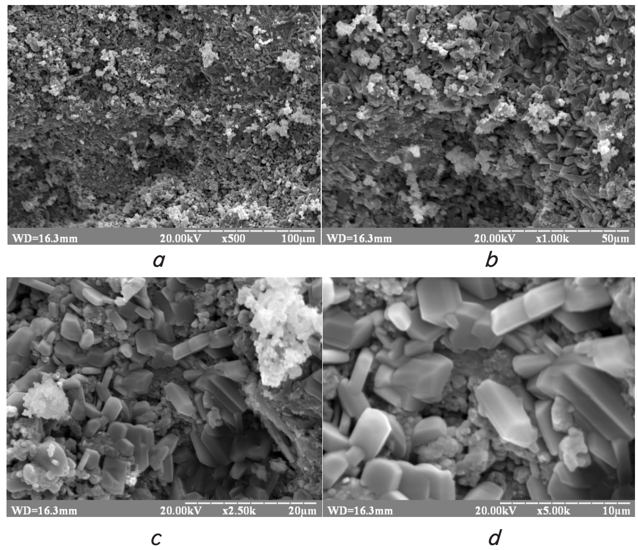


Fig. 16. Electronic photographs of microstructure of the cleaved fragment of artificial stone based on alkaline hydro-aluminosilicate of the form $(0,2K_2O+0,8Na_2O)\cdot 4,5SiO_2\cdot Al_2O_3\cdot nH_2O$ after hardening at temperature 20 °C for 90 days with magnification: *a* — ×500; *b* — ×1000; *c* — ×2500; *d* — ×5000

Thus, it is established that the process of structure formation of alkaline hydro-aluminosilicate of optimal composition $(0,2K_2O+0,8Na_2O)\cdot 4,5SiO_2\cdot Al_2O_3\cdot nH_2O$ in the direction of synthesis of zeolite-like new formations is possible under normal conditions. However, this process proceeds slowly and hence the need for its acceleration.

7. Conclusions

As a result of the conducted research the influence of oxide ratio and hardening temperature of alkaline hydro-aluminosilicate on structure formation and phase composition of artificial stone, in particular:

– at the ratio of SiO_2/Al_2O_3>5 alkaline hydro-aluminosilicate for formation of stone with high performance properties there is a need for high temperature hardening;

– the presence in the alkali hydro-aluminosilicate of potassium ions, at the ratio of $K_2O/R_2O \geq 0,15$, increases the degree of crystallinity and density of structure of artificial stone and contributes to the formation of high silica zeolite-like hydrated phases;

– the temperature of hardening (range 20÷80 °C) for the first 12 hours of structure formation stone, with the same ratio of oxides of alkaline hydro-aluminosilicate, determine the degree of crystallinity of the structure without changing products of hydration;

At hardening of alkaline hydro-aluminosilicate under conditions at normal temperatures, the optimal ratio oxides is $(0,2K_2O+0,8Na_2O)\cdot 4,5SiO_2\cdot Al_2O_3\cdot nH_2O$, which makes it possible to obtain the water-resistant of artificial stone by synthesis, in the composition of hydrated new formations, of zeolite-like minerals of the type: zeolite Na–A; sodium and potassium heulandite, as well as potassium-sodium phillipsite.

Further research may be directed to searching for chemical modifiers of alkaline hydro-aluminosilicate to accelerate the processes of structure formation under normal conditions of hardening.

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