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Investigations of Forming Mixture Parameters of Autoclaved Aerated Concrete with Nanoadditives

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Impact of some nanoadditives, such as amorphous SiO_2 (AS) of pozzolanic properties and carbon fibers (CF) reduced to nanosize particles on autoclaved aerated concrete (AAC) forming mixtures, the following parameters were investigated: changes in consistence of binding material during hydration, spreadability, expansion and temperature of mixture, plasticity strength. The investigations were carried out with AAC forming mixture where 10 % lime was replaced by equivalent content of Portland cement. Upon completion of investigations, it was established that the optimal replacement of sand by AS was 1.0 % resulting in increase of temperature of AAC forming mixture by up to 2.1 %, expansion by up to 11.0 %, ultrasonic impulse velocity (UIV) by up to 3.0 % and plasticity strength by up to 271.4 %. Meanwhile the optimal replacement of sand by CF was 0.1 % resulting in decrease in temperature of AAC forming mixture by up to 1.5 %, expansion by up to 16.0 % and increase in UIV by up to 2.0% and plasticity strength by up to 152.9 %.

Keywords: autoclaved aerated concrete, amorphous SiO_2, crushed carbon fiber, nanoadditive, spreadability, expansion, temperature, plasticity strength, UIV.

1. INTRODUCTION

Amorphous SiO_2 (AS) is a very effective pozzolanic material. As an aggregate, AS powder is in particular suitable for modern building industry. It was used at construction since 1994 in New Zealand and with each year its usage grew. AS is a by-product of ferrosilicon and silicon metal production and can be used in shape of very fine powder [1].

In production of concrete with AS, the pozzolanic reaction is running when SiO_2 with specific surface, which can be as high as 200000 cm^2/g, and with high content of amorphous silica (usually about 90 %) [2] links Ca $(OH)_2$ present in solution, in the same way as active pozzolanic additives containing amorphous SiO_2 of opal origin. This additive defines the process of cement hardening and modifies the microstructure of concrete by making it more homogenous and by reducing big pores in number, while the total porosity remains same as that of concretes without additives, as well as decreases conductivity of water and water vapor and increases strength and life of concrete [3].

At the beginning of hardening process of autoclaved aerated concrete (AAC), as in all concretes, hydrosilicates of amorphous state are forming and a nanostructure is building up. Therefore, one can expect that the properties of these concretes can be also changed by using AS [4]. AS can find usage in lightweight concretes because of its low density (630 kg/m^3), as it does not increase the density of AAC; besides, it contains a high content of SiO_2 (90 % – 92 %), which stimulates formation of AAC crystalline structure [4]; is of high thermal insulation, due to its homogenous microporous structure (λ from 0.11 W/m·K to 0.13 W/m·K); is also of high coherence caused by its pozzolanic characteristics. The mentioned properties show that it can be a potential nanoadditive for AAC [1].

Carbon fibers (CF) in concretes is mostly used as a reinforcing additive for improvement of their strength properties, in particular those under flexure and compression. Furthermore, the sources [5, 6] maintain that this additive adheres pore walls and thus ensures the transfer of load through hollows.

As an additive for AAC, CF was used since long and many sources [5 – 8] state that this additive enhances strength properties of concrete. In the work [9] it was established that the liquid phase existent in AAC forming mixture before its hardening wets thoroughly the surface of CF and that in the layer of binding material, which contacts with CF surface and during hardening of which the crystallization of hydrocrystals is going on. Therefore, a very close link appears between CF and binding material. CF filaments seem to be overgrown by needle-like hydrosilicates. In all abovementioned sources [5 – 9] CF was used as a reinforcing additive without considering its possible impact when reduced to nanosize particles.

The production of AAC is rather complicated and the properties of its outcome are subject not only to AAC composition, but also to parameters of forming mixture. The spreadability of AAC forming mixture is preconditioned by the water/solids ratio (W/S), which in most cases decides the properties of forming mixture and product [10]. Other very important properties of AAC forming mixture are the heating of mixture and the expansion height, which are subject not only to W/S ratio, water temperature,

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ambient temperature, composition of binding material, but also to additives used [11]. On such things as expansion of AAC forming mixture, the future AAC macrostructure and porosity depend.

Therefore, the purpose of this work was to estimate the impact of nanoadditives AS and CF on parameters of AAC forming mixture.

2. MATERIALS AND EXPERIMENTAL METHODS

The composition of AAC forming mixture was the following: binding material, lime 90 %, and 10 % lime replaced by equivalent amount of cement according to the formula [12]:

$$K = \frac{44 \cdot C}{A_0}, \qquad (1)$$

where: 44 is a coefficient of recalculation at 22 % activity of mixture, C is amount of Portland cement replaced by lime in kg, A_0 is activity of lime in %.

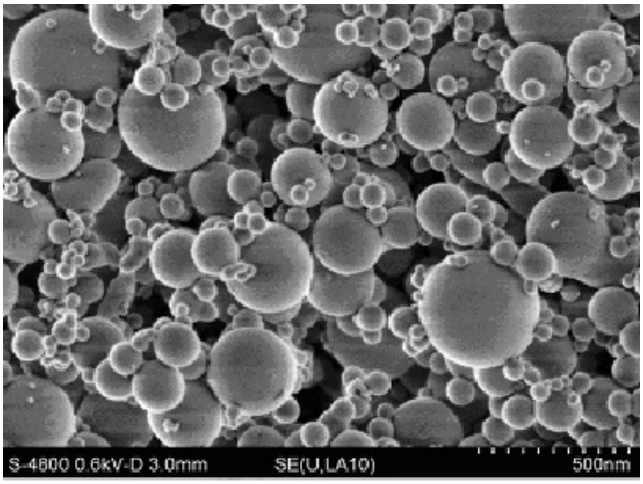
The AAC forming mixtures were prepared using the below mentioned raw materials.

Ground lime from JSC "Naujasis Kalcitas" (Lithuania), corresponding to the requirements of standard LST EN 459-1:2010, activity 90 %, liming duration 5 minutes, liming temperature 55 °C.

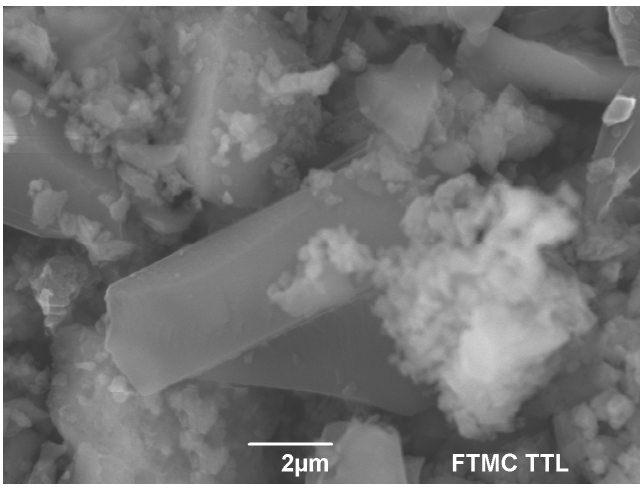
Portland cement CEM I 42.5 R from JSC "Akmenės Cementas" (Lithuania), with specific surface of 3190 cm^2/g, determined by the Blein (Germany) device, and volume stability (expansion) of 0.5 mm, determined by the Le-Chatelier (France) device, corresponding to the requirements of standard LST EN 197-1:2001. Mineral composition of clinker (in %): C_3 – 58.54; C_2S – 15.29; C_3A – 10.40; C_4AF – 10.17. The chemical composition of clinker is provided in Table 1.

Quartz sand from JSC "Anykščių Kvarcas" (Lithuania), ground in ball mill up to fineness of 2766 cm^2/g. The fineness was determined by Blein device according to the requirements of standard LST EN 196-6:2010. The distribution of particles by size is provided in Fig. 2, a. As a gas-generating agent in AAC forming mixtures, aluminum paste "Albo Schlenk Deg 4508/70" (Czech Republic) was used, specific surface 18000 cm^2/g, pure aluminum content in paste 70 %.

The following nanoadditives were used in the work. AS having pozzolanic properties "RW-Füller" (Germany), the chemical composition of which is provided in Table 1,



a



b

Fig. 1. Microscopy images of additives: a – AS; b – CF nanoparticles. Magnification: a – ×60000; b – ×8500

Table 1. Chemical composition of raw materials and additives

Raw materials and additives	Composition (in mass %)										Other, %
	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	R_2O	C	
Lime	4.05	2.21	0.88	89.24	2.38	–	–	–	0.37	–	0.87
Portland cement	20.76	6.12	3.37	63.50	–	1.0	0.3	0.8	–	–	4.15
Sand	97.89	0.60	0.46	0.72	–	–	–	0.06	0.10	–	0.17
CF	3.61	0.13	–	0.26	0.09	–	–	–	–	95.80	0.11
AS	98.00	–	0.05	0.25	0.4	1.2	0.1	0.35	–	0.6	0.00

Table 2. Amount of components AAC forming mixture in grams and in milliliters for 0.01372 m^3 volume

Components	Amount (in grams and milliliters)						
Lime	194.52	194.52	194.52	194.52	194.52	194.52	194.52
Portland cement	19.45	19.45	19.45	19.45	19.45	19.45	19.45
Sand	580.33	577.43	574.53	571.63	579.93	579.53	578.74
AS	–	2.90	5.80	8.70	–	–	–
CF	–	–	–	–	0.40	0.80	1.59
Aluminum paste	1.43	1.43	1.43	1.43	1.43	1.43	1.43
Surfactant	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Water	429.71	429.71	429.71	429.71	429.71	429.71	429.71

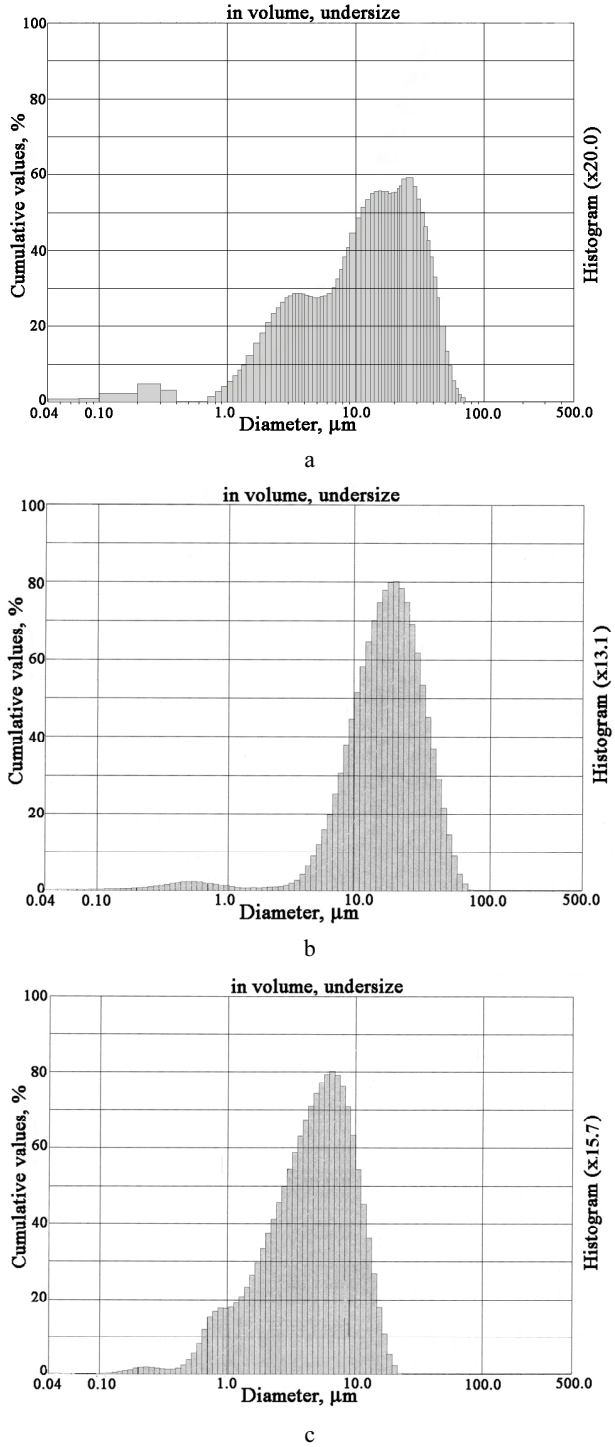


Fig. 2. The distribution of particles by size: a – ground sand; b – AS; c – CF

the shape and size of particles are provided in Fig. 1, a [9]. The distribution of particles by size is provided in Fig. 2, b.

CF ground to nanoparticles, selected in consideration of its resistance to alkaline medium and temperature. The grounding of CF went on for 10 hours in ball mill "Fritsch Pulverisette 7" (Germany) using agate balls of 16 mm diameter, speed 600 rpm.

The connection with matrix of ground CF nanoparticles is provided in Fig. 1, b. The distribution of particles by size is provided in Fig. 2, c. The grounding of CF went on for 10 hours in ball mill "Fritsch Pulverisette 7" (Germany) using agate balls of 16 mm diameter, speed 600 rpm.

For distribution of mentioned additives in AAC forming mixtures, the surfactant "Ufapore TCO" (Norway) was used in content of 0.003 % counting of solids mass.

The compositions of AAC forming mixtures were selected basing on the methodical requirements [13]. The activity of mixture expressed by content of active CaO and MgO was 22 %. The water and solids ratio was 0.54, content of aluminum paste 0.18 % (counting of solids mass).

The components of forming mixtures were mixed in the vertical propeller stirrer of 4 liters by speed of 700 rpm. The duration of component mixing is presented in Table 3.

Table 3. Sequence of components dispensing and duration of mixing for production of AAC

Components	Duration of mixing, min
1. Water + sand	5.0
2. Water + sand + cement	1.0
3. Water + sand + cement + lime	1.0
4. Water + sand + cement + lime + aluminum paste	1.0
5. Water + sand + lime + cement + aluminum paste + AS or CF (mixed with surfactant)	1.0
	Overall: 9.0

For even distribution of nanoadditives and aluminum powder in the mixture, they were dispersed separately by ultrasonic disperser "UZDN-21" (Russia), frequency 22 kHz, capacity 480 W. The dispersion was carried out after mixing nanoadditives with water and surfactant, the duration of process 1 min. Aluminum powder after mixing with water was dispersed for 3 min. For production of AAC, the equal water/solids ratio (0.54) was used. The spreadability was tested by "Suttard" viscosimeter (Russia) according to the methodical requirements [14]. The expansion temperature of AAC forming mixture was measured by a mercury thermometer put into middle of expanding mass. The measurements were carried out each minute and registered according to standard LST 1428.5:1996. The expansion height of mixture was determined as a difference between heights of expanded and initial mixtures, after marking the initial height and registering it each minute. The plastic strength was determined by Rebinder conical plastometer. Ultrasonic impulse velocity (UIV) traveling by AAC forming mixture and showing changes in density of mixture was determined by a device "Pundit 7" (England) according to standard LST EN 12504.4:2004. The investigations of macrostructure of AAC forming paste were carried out by an optical microscope "Motic" (China).

3. RESULTS

3.1. Spreadability

Upon addition of nanoadditive AS or CF into paste, the spreadability of forming paste decreased subject to content of nanoadditives. The impact of additive on spreadability of AAC forming mixture starts with 0.5% AS and 0.05 % CF. Along with increase of AS additive from 0.5 % to 1.5 % mass, the diameter of cake underwent very insignificant changes, i. e. decreased by up to 0.8 %, and in the case of CF addition of from 0.05 % to 0.2 %, the diameter decreased by 1.4 %.

The decrease in spreadability of AAC forming mixture was preconditioned by fineness of nanoparticles, i. e. specific surface of particles.

3.2. Temperature

In comparison of both additives, one can see that they exert a different effect on AAC forming mixture. AS increases the expansion temperature of mixture, while CF decreased it. After consideration separately of the impact of additives on AAC forming mixture, it was established that the AS additive added in content from 0.5 % to 1.5 % increased the expansion temperature of mixture by up to 3.28 % versus reference sample without nanoadditive. In the case of the other nanoadditive, CF, a different situation was observed versus that of AS nanoadditive. The added CF resulted in decrease of temperature. Along with ever increasing addition of this nanoadditive (the highest addition being 0.2 %), the temperature dropped by 2.2 % versus reference sample. It shows that nanoadditive CF does not react with binding material and that its higher content stops the reactions going on in the mixture (Fig. 3).

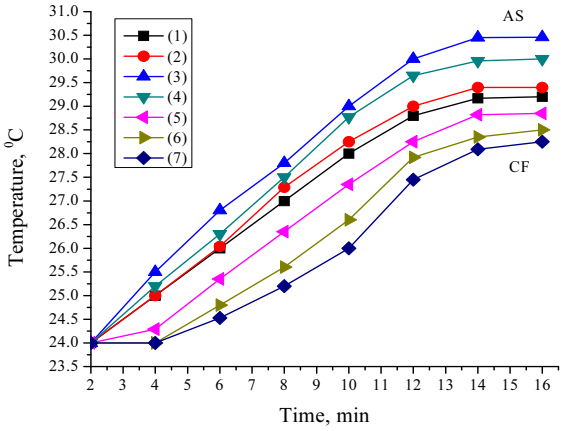


Fig. 3. Expansion temperature of AAC forming mixture at the following content of nanoadditive: 1 – 0.0 %; 2 – 0.5 % AS; 3 – 1.0 % AS; 4 – 1.5 % AS; 5 – 0.05 % CF; 6 – 0.1 % CF; 7 – 0.2 % CF

3.3. Expansion

The obtained results of testing showed that the nanoadditive AS stimulates the reactions running in the mixture, therefore, not only the temperature, but also the expansion is growing. With the nanoadditive AS, the AAC forming mixture expanded higher than the reference sample, namely by 11.0 % with 1.0 % AS and by 17.9 % with 1.5 % AS. The comparison of impact of nanoadditive CF on AAC forming mixture shows the same pattern of changing as in the case of temperatures. The nanoadditive CF decreases the expansion of mixture. Higher addition of CF into AAC forming mixture lowers the expansion, the more of CF, the lesser expansion of AAC forming mixture. Therefore, the optimal added content will be 0.05 %, at which the expansion of mixture changes insignificantly, decreasing by up to 4.6 %, but along with higher addition (0.2 %), the expansion height of mixture decreases by up to 26.1 % versus reference sample (Fig. 4).

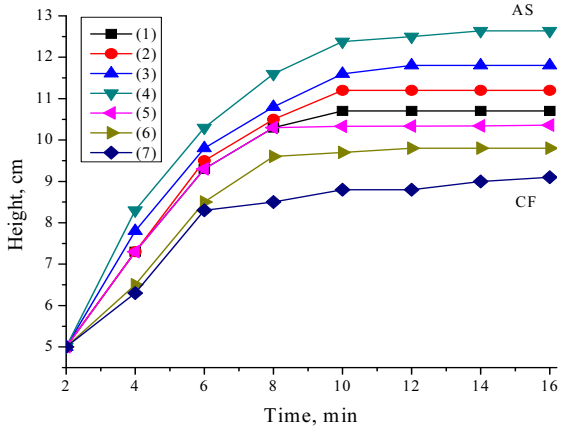


Fig. 4. Expansion of AAC forming mixture at the following content of nanoadditive: 1 – 0.0 %; 2 – 0.5 % AS; 3 – 1.0 % AS; 4 – 1.5 % AS; 5 – 0.05 % CF; 6 – 0.1 % CF; 7 – 0.2 % CF

3.4. Plasticity strength

The test results show that the nanoadditive AS or CF exerts a different effect on plasticity strength of AAC forming mixtures. Since AS is an active nanoadditive, which intensely reacts with binding material and forms a stronger and denser carcass, this ensures a higher plasticity strength to forming mixture. In AAC forming mixtures with nanoadditives AS or CF the plasticity strength is obtained higher than that of reference sample, only the nanoadditive AS results in higher plastic strength than the nanoadditive CF.

Using AS, the plasticity strength of AAC forming mixture increased up to a certain limit. If addition of AS exceeds 1.0 %, the plasticity strength starts decreasing. With AS 1.0 %, the plasticity strength grew by up to 250.0 % and with 1.5 % by up to 285.7 %.

If the inactive nanoadditive CF was used, the plasticity strength increased proportionally to CF content, i. e. upon addition of from 0.05 % to 0.2 %, the plasticity strength increased by from 150.0 % to 157.1 % (Fig. 5).

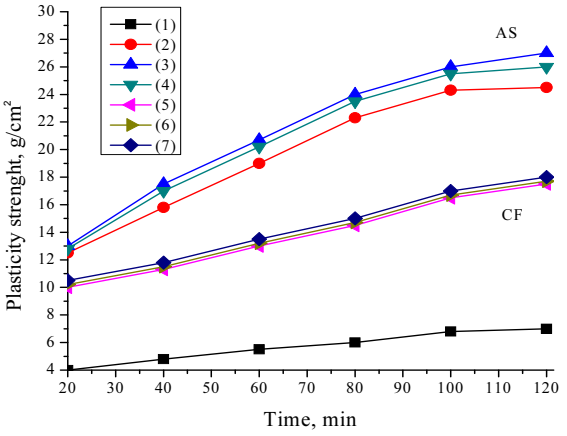


Fig. 5. Plasticity strength of AAC forming mixture at the following content of nanoadditive: 1 – 0.0 %; 2 – 0.5 % AS; 3 – 1.0 % AS; 4 – 1.5 % AS; 5 – 0.05 % CF; 6 – 0.1 % CF; 7 – 0.2 % CF

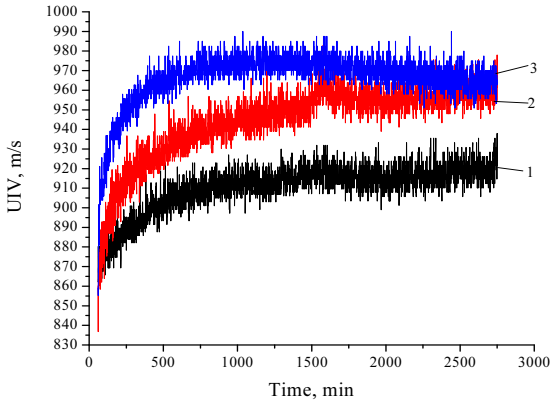


Fig. 6. UIV of AAC forming mixture at the following content of nanoadditive: 1 – 0.0 %; 2 – 1.0 % AS; 3 – 0.1 %

During the tests on plasticity strength, it was observed that the nanoadditives increase the plasticity strength of AAC forming mixture what positively influences the technology of AAC production.

3.5. UIV

UIV was registered only at optimal contents of additives (selected by obtained test results of plastic strength), i. e. at CF 0.1 % and AS 1.0 %. The measurement of UIV in AAC forming mixture was carried out after full expansion of mixture, i. e. after 20 min. since pouring of mixture into vessel.

The nanoadditives AS and CF increase the velocity of ultrasound traveling through forming mixture. The highest UIV was with nanoadditive AS, due to compacting of the material of interpore systems. With nanoadditive AS, UIV increased till 3.0 %, with CF till 2.0 % (Fig. 6).

These investigations confirmed that the nanoadditives increase the plastic strength of AAC forming mixture, since the increase in UIV shows the growing density (or plastic strength) of AAC semifinished product.

3.6. Macrostructure

After the tests of plastic strength, the samples were hard enough to cut off the expanded heap, afterwards the samples were dried and their macrostructure was determined (Fig. 7).

CF into AAC forming mixture, no very remarkable visual changes of macrostructure were observed, only one could see pores distributed more evenly and their diameter more uniform.

The added AS increased the expansion temperature and height of mixture, but when addition of AS exceeded 1.0 %, the plastic strength started decreasing. Basing on these test results, the macrostructural investigations were performed. They demonstrated that a small amount of added AS had no great effect on structure, however, upon addition of more than 1.0 % (in this case 1.5 %), the structure started changing and the contacting pores of various sizes formed. So, a conclusion was drawn to say that the content of AS in excess of 1.0 % in AAC forming mixture increased the temperature of reaction and the expansion height of forming mixture, but all this initiated destruction of generating pores and all the macrostructure.

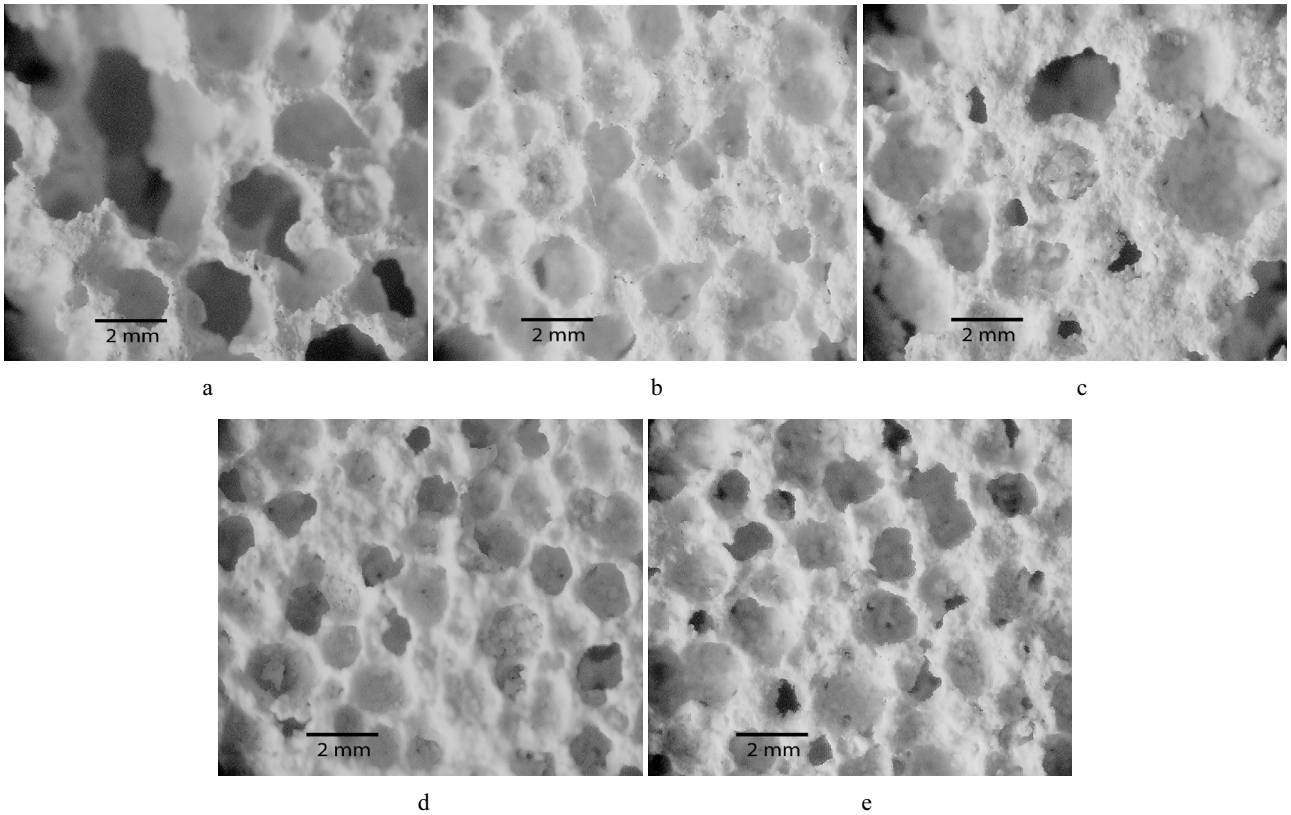


Fig. 7. Pores distributed of AAC forming mixture samples at the following content of nanoadditive: a – 0.0 %; b – 1.0 % AS; c – 1.5 % AS; d – 0.1 % CF; e – 0.2 % CF

4. DISCUSSION

The volume instability of AAC forming mixture limits the abilities of determination of its properties, as selection of unsuitable methods of analysis can lead to the question of reliability of the obtained results. The authors which deal with the impact of various additives, usually emphasize improved properties of AAC products (obtained after treatment in an autoclave) [4, 8 – 10] or choose studies based on a research of forming mixtures with a constant volume, specific to the cementitious materials (hardened in the natural conditions) [2, 3]. The properties of forming mixtures of non-modified AAC studied more widely in [7, 10, 11, 15]. It was found that the spreadability, temperature and plasticity strength depend on water temperature, water and solids ratio and activity of the forming mixture. The influence of fibers of different type and length on the properties of AAC forming mixtures was studied in [7, 8, 12, 16]. It was found that 0.1 % (counting of solids mass) of mechanically untreated CF leads to the decrease of spreadability or temperature of the forming mixtures accordingly by 3.5 % and 10.2 %, or by 1.0 % and 1.5 %, expansion in one case increased by 8.0 % [7], otherwise decreased by 7.1 % [16], while plasticity strength increased accordingly by 116.7 % and 114.3 %. It was also found that fibers of different type and length do not change the macrostructure of AAC. Unfortunately, authors could not find a data on the influence of nano-sized particles, such as AS or CF, on the properties of AAC forming mixtures. In our case, the spreadability, temperature and expansion of AAC forming mixtures with 0.1 % (counting of solids mass) of ground to nanoparticles CF decreased accordingly by 0.8, 2.2 and 4.6 %, while plasticity strength increased by 152.9 %.

5. CONCLUSIONS

1. It was established that the optimal content of nanoadditive AS in AAC forming mixtures is 1.0 % and CF – 0.1 %. They influence positively the properties of forming mixture:

– in AAC forming mixture with AS, the spreadability decreased till 0.5 %; the expansion temperature increased till 2.1 %; the expansion height of mixture increased till 11.0 %; the plastic strength increased till 271.4 %; UIV increased till 3.0 % versus reference sample without additive;

– in AAC forming mixture with CF, the spreadability decreased till 1.1 %; the expansion temperature decreased till 1.5 %; the expansion height of mixture decreased till 16.0 %; the plastic strength increased till 152.9 %; UIV increased till 2.0 % versus reference sample without additive.

2. Upon comparison of impact of both additives on properties of AAC forming mixtures and products, one can state that the additive AS most effectively acts on plastic strength and UIV.

3. The macrostructural investigations showed that AS nanoadditive contributed to the generating of small closed pores of uniform size. Nevertheless, these properties were improving along with increase in AS content to a certain level, i. e. 1.0 %, while in excess of it, more contacting pores generated in the macrostructure. CF nanoadditive has slight effect to macrostructure of AAC.

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