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ВПЛИВ ВИСОКОВОЛЬТНОЇ ЕЛЕКТРОСТАТИЧНОЇ ОБРОБКИ НА ПУЦОЛАНОВУ АКТИВНІСТЬ ЗОЛИ-ВИНЕСЕННЯ ТА ВЛАСТИВОСТІ ЦЕМЕНТНО-ЗОЛЬНИХ ПАСТ

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Анотація. Розглянуто вплив високовольтної електростатичної обробки на пуцоланову активність золи-винесення та властивості цементно-зольних паст. Встановлено, що активована електрофізично зола-винесення характеризується більш високим ступенем гідратації та пуцоланової активності. Це впливає на реологічні властивості цементно-зольних паст – частинки золи-винесення виконують роль пластифікуючого агенту в системі. З підвищенням ступеня гідратації золи-винесення підвищується ступінь гідратації цементу, що забезпечує зростання міцності каменю в'яжучого при стиску. Модифікування адгезійних контактів на межі цементна паста – мінеральна добавка (електрогетерогенні контакти) впливатиме на формування більш однорідної та щільної мікроструктури цементного каменю та бетону.

Ключові слова: зола-винесення, портландцемент, електростатична обробка в полі коронного розряду, ступінь гідратації, сферичні частинки, міцність при стиску.

ВЛИЯНИЕ ВЫСОКОВОЛЬТНОЙ ЭЛЕКТРОСТАТИЧЕСКОЙ ОБРАБОТКИ НА ПУЦЦОЛАНОВУЮ АКТИВНОСТЬ ЗОЛЫ-УНОСА И СВОЙСТВА ЦЕМЕНТНО-ЗОЛЬНЫХ ПАСТ

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Аннотация. Рассмотрено влияние высоковольтной электростатической обработки на пуццолановую активность золы-уноса и свойства цементно-зольных паст. Установлено, что активированная электрофизически зола-унос характеризуется более высокой степенью гидратации и пуццолановой активности. Это оказывает влияние на реологические свойства цементно-зольных паст – частички золы-уноса играют роль пластифицирующего агента в системе. С повышением степени гидратации золы-уноса повышается степень гидратации цемента, что обеспечивает рост прочности камня вяжущего при сжатии. Модифицирование адгезионных контактов на границе цементная паста – минеральная добавка (электрогетерогенные контакты) будет оказывать влияние на формирование более однородной и плотной микроструктуры цементного камня и бетона.

Ключевые слова: зола-унос, портландцемент, электростатическая обработка в поле коронного разряда, степень гидратации, сферические частицы, прочность при сжатии.

AN INFLUENCE OF A HIGH-VOLTAGE ELECTROSTATIC TREATMENT ON POZZOLANIC ACTIVITY OF FLY ASH AND PROPERTIES OF CEMENT-FLY ASH PASTES

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Abstract. An influence of a high-voltage electrostatic treatment on pozzolanic activity of fly ash and properties of fly ash-cement pastes is considered. An electro-physically activated fly-ash powder is characterized by a higher hydration and pozzolanic activity. This affects the rheology of cement fly-ash pastes, fly-ash particles playing the role of a plasticizing agent in the system. The higher degree of fly-ash hydration, the higher degree of cement hydration, that provides an increase of ash-cement paste compressive strength. A retrofitting of adhesive contacts on the boundary of cement paste–mineral additive (electro-heterogeneous contacts) will effect the formation of a more homogeneous and dense microstructure of a cement paste and concrete.

Keywords: fly-ash, Portland cement, corona electrostatic treatment, degree of hydration, spherical particles, compressive strength.

1.Introduction

High Performance Concrete is a complex mixture often containing 5-10 different materials [1]. Among these materials mineral additives with high degree of pozzolanic activity play an important role in providing the concrete strength and durability. Silica fume appears to be the most performing siliceous product among the pozzolanic materials for high performance concretes, however it is not available in large amounts and it is also the most expensive mineral additive (about 0.25-0.50 ^/kg in Europe). On the other hand, fly ash is available in large amounts and is relatively cheap (0.02-0.03 ^/kg in Europe) [2].

Coal fly ash is a supplementary cementitious material generated by thermal eclectic power stations and widely used in the composition of concretes. The power stations generally use either pulverized coal-fired or cyclone furnaces. Fly ash constitutes a major component (75-80 %) of by-product material at pulverized coal-fired power plants [3].

It's a matter of common knowledge that fly ash is usually used to replace the part of Portland cement by improving some properties of concrete such as reduction in heat of hydration, increasing durability and impermeability of the concrete in service [4]. The active effect of fly ash is composed of two parts: the pozzolanic activity of fly ash itself and the promoting role of fly ash to hydration of Portland cement [5]. However, with increased portion of cement replacement the concrete strength decreases. So in order to get resemblance in properties with ordinary Portland cement fly ash needs special treatment like mechanical grinding, accelerated thermal curing, and alkali or sulfate activation. On the other hand, mechanical grinding is connected with additional energy consumption, alkali activation can lead to alkali-silica reaction and sulfate activation can decrease the durability of concrete due to the large ettringite content [6].

Different electrophysics activation methods are used in a less measure. It is established earlier that high-voltage electrostatic polarization of Portland cement causes acceleration of its hydration and intensifies cement paste structuring [7, 8]. Bipolar or unipolar activation of cement and mineral powder (fly ash or limestone filler) in corona electrostatic field influences interparticle interactions in cement paste stipulating its rheological properties and strength characteristics of the concrete [9]. *The purpose of this investigation* is to determine the influence of high-voltage electric treatment on the degree of pozzolanic activity of fly ash and the properties of fly ash-cement pastes.

2. Materials and methods

2.1. Materials

Class F fly ash (FA) of Uglegorsk power station (Donetsk region), Ordinary Portland Cement (OPC) CEM I 42.5 N and tap water were used in this investigation. The Blaine fineness of dispersed powders were 345 (OPC) and 360 (FA) m^2/kg. The specific gravities of OPC and FA were 3.11 and 2.34, respectively.

Fly ash replacement ratio was 30 and 50 % by mass of cementitious powder (OPC + FA). The fly ash–OPC pastes were prepared with a water/cementitious (W/C) ratio which correspond to the normal consistency of Vicat apparatus. The composition of cement pastes is given in Table 1. Fly ash was previously activated in the installation for treatment of dispersed powders in corona high-voltage electric field (indexes of cement pastes – 1A-FA, 2A-FA). With the purpose of determination of the influence of high-voltage electric activation on the degree of hydration of fly ash the control compositions of cement pastes were prepared also (indexes – 1C-FA, 2C-FA).

2.2. The installation for treatment of fly ash in corona high-voltage electric field

Dispersed powder of fly ash was treated on the specially constructed installation which functions on the principle of a corona electrofilter (Fig. 1). The corona electrode – a copper wire with a diameter 0.6 mm is coaxial with the grounded metallic tube [10]. Material was treated at the electric field strength E = 20-25 kV/cm and the current intensity I =25–40 jA. The charge of corona electrode was negative. These parameters allow to create the sufficient concentration of ions in all volume of the activation chamber – space charge density near the wire was 170 $\mu C/m^3$ (10^6 single-charge ions in 1 mm^3) [9].

Table 1. The composition of cement pastes.

Index	Composition of cement paste		
	OPC, g	FA, g	W/C
1C-FA	294	126	0.224
1A-FA	294	126	0.218
2C-FA	210	210	0.214
2A-FA	210	210	0.202

2.3. Determination of degree of hydration of fly ash

To stop the hydration reaction the cement-fly ash paste samples were cracked and soaked in acetone for 1 day. These samples were dried at room temperature (20°C) for 2 h and further dried at 40°C for 3 h. Then the samples were grinded until the powder passed through 80-µm sieve. After that the samples were dried again at 110°C for 12 h.

The degree of hydration of fly ash was determined by a selective dissolution method. This method is consisted in the determination of the quantity of unreacted fly ash in hardened fly ash–cement paste which is successfully separated using 2 N HCl and 5 % Na_2CO_3 solutions. To dissolve the component including the Ca ions from the unhydrated OPC and the hydrated products, 2 N HCl is added. Gel composed of SiO_2, Al_2O_3, Fe_2O_3 and unhydrated fly ash is left. After that, 5 % solution of Na_2CO_3 is added to dissolve the SiO_2, Al_2O_3 and Fe_2O_3 gel. Finally, only unreacted fly ash is left.

The degree of hydration of fly ash was calculated by the following equations:

$$\alpha = 1 - \left[\frac{x_s (1 - Ig_s')}{a_1 \cdot a_2} \right] \tag{1}$$

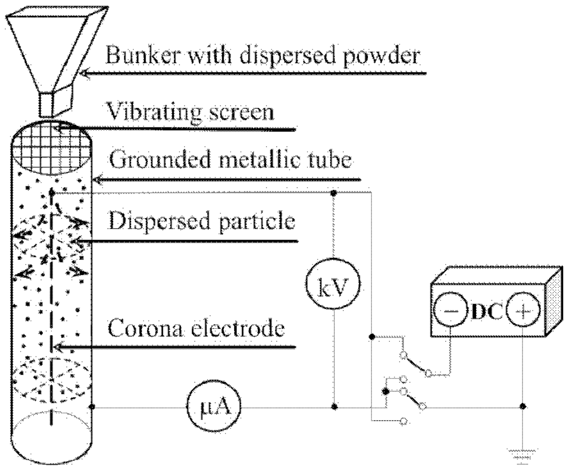

Fig 1. Installation for dispersed materials activation in the corona-electrostatic field.

$$a_1 = \frac{(1-Ig_F)R}{(1-Ig_f)R+(1-Ig_0)(1-R)} \qquad (2)$$

$$a_2 = \frac{x_f - Ig_F}{1-Ig_f} \qquad (3)$$

In the above, α is the degree of hydration of fly ash; a_1 is the ignitial fraction of fly ash before the dissolution process; a_2 is the residual ignition weight of 100 % fly ash after the dissolution process; R is the replacement ratio of fly ash by weight; x_s and x_f are the weights after the dissolution process of the hydrated sample and unhydrated fly ash, respectively. Ig_f, Ig_F, Ig_o and Ig_s' refer to the ignition loss of unhydrated fly ash, unhydrated fly ash after the dissolution process, unhydrated OPC and the hydrated sample after the dissolution process, respectively. In Eq. (1), $x_s(1-Ig_s')$ shows the ignition weight of unhydrated fly ash in a hydrated sample after the dissolution process and a_1a_2 shows the ignition weight of unhydrated fly ash in an unhydrated sample after the dissolution process [11].

The consistency of investigation process is the following:

1) The initial portion of FA m=1 g is ignited, after that the loss of ignition (LOI) is measured – Ig_f.
2) The initial portion of FA m=1 g is treated with 2 N HCl solution, after that is dried and weighed – x_f.
3) The portion of FA m=x_f is ignited, after that the loss of ignition (LOI) is measured – Ig_F.
4) The portion of unhydrated OPC with addition of FA (replacement mass fraction R) m=1 g is treated successively with 2 N HCl and 5 % Na_2CO_3 solutions, after that is dried and weighed. Then the residual specimen is ignited, after that the loss of ignition (LOI) is measured – Ig_0.
5) The portion of hydrated OPC with addition of FA (replacement mass fraction R) m=1 g is treated successively with 2 N HCl and 5 % Na_2CO_3 solutions, after that is dried and weighed – x_s.
6) The portion of hydrated OPC m=x_s is ignited, after that the loss of ignition (LOI) is measured – Ig_s.

The details of these procedures are given in [11]. The LOI of materials were detected using differential thermal analysis (DTA) and thermogravimetric (TG) analysis.

3. Results and discussion

3.1. Degree of hydration of fly ash

The results of calculations the degree of hydration of fly ash under the data of differential thermal analysis (DTA) and thermogravimetric (TG) analysis are given in Table 2. It can be seen from the table that fly ash treated in the corona electrostatic field has the degree of hydration in 1.4 times higher in comparison with untreated sample. There are several considerations to explain this phenomenon. Firstly, the properties of solid surface in a great deal are determined by concentration and polarity of active centers, i.e. by its electric relief. According to [12] the active centers (functional groups) of surface of mineral additives (fillers) influence on the processes of Portland cement hydration from the first seconds of interaction of cement minerals with mixing water. So, cancellation of surface charges results in diminishment of activity of matter to chemical interactions - there is the "aging" of surface and increase of its reactionlessness. It has been established earlier [13] that mineral additives treated in the high-voltage corona electric field with different polarity of discharge electrode showed substantial differences in distributing on the surface of acid-basic centers. The acid (pK_a =-4,4), moderately acid (pK_a = +2,1) and basic (pK_a = +8,8 and +12,9) centers are the main adsorptive sites for an

Table 2. The calculated data of the degree of hydration of fly ash.

Index	The calculated data								
	LOI, %	x_f, g	I_{g_F}, g	I_{g_f}, g	R	I_{g_0}, g	x_s, g	I_{g_S}, g	α
FA (initial) – 1.0 g*	6.0	-	0.060	-	-	-	-	-	-
FA (dissolution in 2N HCl) – 1.0 g	3.5	0.9	0.032	-	-	-	-	-	-
OPC+FA – unhydrated sample (dissolution in 2N HCl, 5% Na_2CO_3) – 1.0 g	9.0	-	-	-	0.3	0.038	-	-	-
2C-FA – hydrated sample (dissolution in 2N HCl, 5% Na_2CO_3) – 1.0 g	5.5	-	-	-	0.3	-	0.240	0.013	**13.6**
2A-FA – hydrated sample (dissolution in 2N HCl, 5% Na_2CO_3) – 1.0 g	6.2	-	-	-	0.3	-	0.225	0.014	**19.1**

1.0 g* - the initial probe of material before the dissolution process

initial fly ash powder. After the treatment of fly ash in electrostatic field with a negative discharge electrode amount of acid and moderately acid centers substantial increased but amount of centers with pK_a = +8,8 diminished. In accordance to [14] the strengthening of acid properties of quartz surface filler resulted in activation of acid-basic reactions with cementing phases. Besides, the long-range field of electrically active relief of solids surface will affect the processes of structure formation in cement paste.

3.2. Fly ash-cement paste rheology

It is usually reported that, if the volume concentration of a solid is held constant, the addition of mineral fillers improves concrete performance but reduces workability because the fine mineral powder will increase the water demand due to the increase in surface area. However in certain cases the use of fine mineral additives can reduce the water demand or increase the slump, especially fly ash [1]. The most common reason for increasing the workability of cement paste is that the spherical fly ash particles easily roll over one another, reducing interparticle friction [15, 16]. Besides, according to [17] clinker powder has a positive integral surface charge, thus the extent of potential energy barrier between particles is small that is not prevent their flocculation with decreasing the volume of "free water" in dispersed system [18]. The fine particles of fly ash with negative surface charge [17] get adsorbed on the oppositely charged surfaces of cement particles and prevent them from flocculation (Fig. 2). The cement particles are thus effectively dispersed and will trap large amounts of water that means that the system will have a reduced water demand to achieve a given consistency [15]. Secondly, the fine fly ash particles attached to the surface of the larger cement particles will form 3-d spherical shape agglomerates – "spherical cement" that provide high fluidity of cement pastes and concrete mixtures [17].

This effect is very strong in the systems with high volume of fly ash, i.g. High- Performance, High-Volume Fly Ash Concretes [15]. In the case of electric activation of fly ash in corona high-voltage electric field the value of negative zeta-potential of particles will increase to a great extent, thus the water demand of fly ash-cement paste must decrease. These phenomena are illustrated by the data given in table 1. We can see that the system with electrically treated fly ash powder 1A-FA has the water-cementitious ratio on 2.7% smaller in comparison with the control sample 1C-FA (the Vicat normal consistencies of both systems are similar). When the cement replacement is higher (2A-FA, 2C-FA) this effect rises up to 5.6%.

3.3. Fly-ash-cement paste compressive strength

Table 3 illustrates the results of compressive strength testing of hydrated fly ash – cement pastes

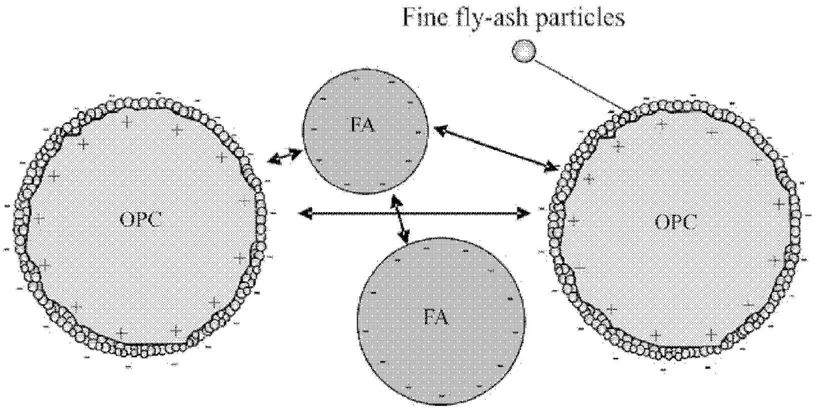

Fig. 2. The repulsion forces between unipolar charged fly ash – cement particles.

Table 3. The compressive strength of fly ash-cement pastes.

Index	Compressive strength, MPa		Relative compressive strength, %	
	3-day	28-day	3-day	28-day
1C-FA	22.4	67.8	100	100
1A-FA	26.2	76.3	117	112.5
2C-FA	13.8	44.7	100	100
2A-FA	17.0	53.2	123	119

with different replacement ratio of Portland cement by fly ash. It can be seen clearly that systems with high content of fly ash (50 % replacement ratio) have considerably less compressive strength on comparison with the systems whereas the replacement ratio is only 30%. These differences are essential at the early period of cement paste hydration. On the other hand the index of strength activity (relative compressive strength) of fly ash treated in electrostatic field is stronger when the replacement ratio is higher. It has been reported [5] that when the content of fly ash is less, its pozzolanic activity can exert well, but its promoting role to the hydration of cement is weaker. When the content of fly ash is more, it is less than its pozzolanic activity can be used, but its promoting role to the hydration of cement is stronger.

It is necessary to mark also that strength increasing of the systems with activated fly ash is connected with plasticizing effect of fine fly ash particles attached to the oppositely charged surfaces of Portland cement particles.

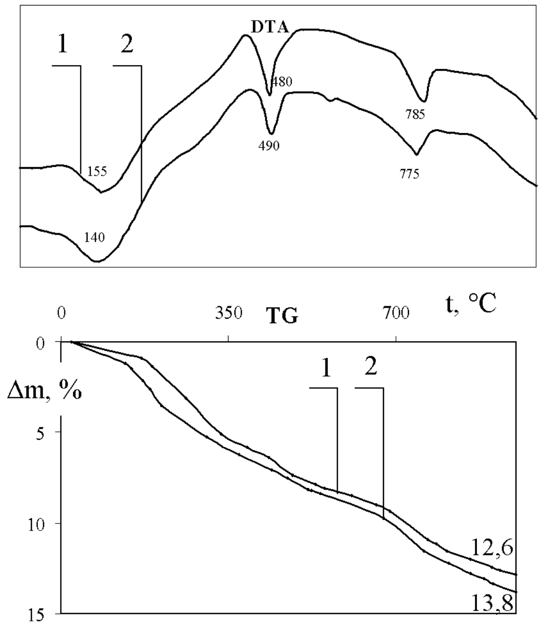

Fig. 3. DTA and TG-curves of hydrated cement pastes with treated (2) and untreated (1) fly ash powder.

Fig. 3 shows that the total loss of ignition of fly-ash cement paste with activated in electric field fly ash powder is higher on 9.5 % in comparison with control sample. The endothermic peaks on DTA curves of activated and control samples are similar. However the depth of endothermic peak in the thermal interval with extreme point 140-155°C which is associated with dehydration of CSH-gel is well above for the activated sample. While the endothermic peak with extreme point 480-490°C (dehydration of calcium hydroxide) for activated fly ash – cement paste is not so mach as for control sample. This is due to the activating effect of high-voltage corona electrostatic treatment on the degree of hydration of fly ash.

Conclusion

1. With the help of a selective dissolution method the degree of hydration of fly ash was determined. Fly ash powder treated in the electrostatic corona field has the degree of hydration in 1.4 times higher in comparison with untreated sample.
2. The system with electrically treated fly ash powder 1A-FA has the water-cementitious ratio on 2.7% smaller in comparison with the control sample 1C-FA (the Vicat normal consistencies of both systems are similar). When the cement replacement is higher (2A-FA, 2C-FA) this effect rises up to 5.6%. This is due mainly to the repulsion forces between unipolar charged fly ash – cement particles.
3. The systems with high content of fly ash (50 % replacement ratio) have considerably less compressive strength on comparison with the systems whereas the replacement ratio is only 30 %. These differences are essential at the early period of cement paste hydration. On the other hand the index of strength activity (relative compressive strength) of fly ash treated in electrostatic field is stronger when the replacement ratio is higher.

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Зайченко Микола Михайлович – кандидат технічних наук, доцент, працює доцентом кафедри технологій будівельних матеріалів, виробів і автомобільних доріг Донбаської національної академії будівництва і архітектури. Наукові інтереси: високоміцні і особливо високоміцні бетони на основі фізико-хімічно модифікованих дисперсних компонентів бетону.

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