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MINIMISATION OF ENVIRONMENTAL EFFECTS RELATED WITH STORING FLY ASH FROM COMBUSTION OF HARD COAL

The tests have been carried out on granulation of fly ash that may significantly reduce the consequences of its storage and transport harmful to the environment, mainly secondary dusting. Granulation was conducted using a disk granulator. The experiments involved the evaluation of a loose additive (bentonite) impact, the analysis of moisture content and water and sodium glass impact, the optimisation of the added bentonite amount and the granulated product breaking resistance test. The were examined such parameters as the ratio of ash to fillers, the amount of supplied binding liquid (distilled water or waterglass) and the place of its feeding. The addition of bentonite to bed (ash) made it possible to completely granulate the material. The introduction of waterglass in place of demineralised water produced the desired effect.

1. INTRODUCTION

In view of the European Union's circular economy strategy, there is a need to develop treatments that may allow to improve the management of industrial waste [1]. Different types of solid waste generated in these processes should ultimately be treated as by-products and be used in various industries (such as fly ash from coal combustion, for example). The regulations in Europe pertaining to the protection of air provide that

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the permissible levels of fly ash emissions from hard coal combustion in power plants generating less pr more than 500 MW are 100 and 50 mg·m^{-3}, respectively [2]. Coal fly ash is a by-product of the combustion of coal in electric power generating plants. It consists of powdery particles that are spherical in shape, and mostly glassy in nature [3]. Reference data shows that ash with finer graining is characterised by a larger content of the vitreous phase, while coarser grades are dominated by unburned coal particles [4]. Fly ash is a major waste of coal-power generation and its management is a major environmental and economic challenge [5]. Recently, a growing interest in the utilization of fly ashes has been observed such as ashes collected directly under electrostatic precipitators and ashes deposited in open locations for temporary storage as well [6].

Fly ash is found in flue gas removed from furnaces and stopped in the process of flue gas purification (usually using electrostatic precipitators), as well as mechanically, using cyclones or, less often, bag filters. The main components of fly ash include silicon, aluminium and calcium oxides. The chemical composition of ash depends on the type and quality of the burned coal. The so-called furnace waste is reused or stored in dumps, depending on the quality of the coal that it derived from, the flue gas contamination level and the purification method. The approach to the issue of furnace waste has significantly changed in the last twenty years.

The advantage of fly ash is its pozzolanic properties. It means that it reacts with lime in the presence of water and at room temperature, giving rise to a material that does not dissolve in water. Its pozzolanic properties result from the presence of silicon dioxide (SiO_2) and aluminum trioxide (Al_2O_3) in their amorphous forms. Those qualities make it a valuable ingredient of cement and concrete, equivalent to mineral aggregates and often even outperforming them. Fly ash, meeting the requirements of standards PN-EN 450-1:2012 *Fly ash for concrete. Part 1. Definitions, specifications and conformity criteria*, and PN-EN 450-2:2006 PN-EN 450-2:2006. *Part 2. Conformity evaluation* has the widest application in the cement and concrete technology. Fly ash with high fineness exhibits high pozzolanic activity and can be used to make high-strength concrete. Evaluation of the pozzolanic activity of ash falls into three categories: mechanical, physical and chemical. The pozzolanic properties of a coal combustion bottom ash were investigated in study [7]. Payá et al. [8] as well as Kurama and Kaya [9] pointed out an increase in pozzolanic activity with grinding of fly ash. From theoretical analyses and experimental results, Chenghi et al. [10] concluded that the addition of pozzolanic material influences the packing state and decreases the amount of filling water. This role depends on the fineness of pozzolanic materials. Each fly ash particle has its own conditions for the pozzolanic reaction [11]. The research showed that pozzolanic activity of bottom ash increases only slightly if the specific surface area of particles expands [12]. The addition of fly ashes improves the mechanical characteristics of concretes and mortars as a consequence of their pozzolanic activity and the spherical form of their particles [13]. The use of coal fly ash for soil amelioration may provide soil enrichment [14]. In some cases, depending on the composition of the original waste, the combustion

conditions and the combustion technology adopted bottom ash may also exhibit pozzolanic activity as a consequence of high contents of amorphous phase. High Si and Al contents suggested possible pozzolanic activity of the material [15].

A major problem with the reuse of fly ash in civil engineering is the mass of loss on ignition which should be lower than 9%. Ash that due to its properties cannot be reused or whose use is economically unjustified is disposed to waste dumps.

In contrast to large energy producers, local producers are still facing the challenge of managing fly ash resulting from hard coal combustion. Local heat generating plants burn coal from various sources and of varied quality, which is why the parameters of the resultant fly ash vary as well [16]. The applied technological solutions also pose a problem. The boiler plants of most municipal and local heat generating plants are equipped with stoker-fired boilers, steam boilers or water boilers. Fly ash from such boilers contains more unburned coal particles than ash from pulverized-fuel boilers or fluidised boilers used in professional power generation by large producers. Furthermore, the particle size of the generated waste is relatively small.

Özer et al. [17] present an extensive review of the coal-agglomeration process, characterization methods of coal hydrophobicity, and factors affecting the agglomeration-process in terms of energy recovery and ash recycling. Granulation may be a solution to the problem of local heat generating plants with the storage and reuse of fly ash [18]. The processing of dust into a granulated product solves several issues important in terms of waste management. A granulated product, in contrary to ash, may be stored in dumps, as it does not scatter dust. For the same reason, no special means of transport are necessary for transporting granulated products, while the storage of ash in the form of dust requires silos and its transport – tankers. Furthermore, granulated fly ash, in contrast to dust, provides greater possibilities of reuse, especially because of the fact that it is easy to store and transport.

In order to distinguish the individual fractions, a granulated product undergoes a screening process using sets of sieves [19]. It must be noted that the mentioned ash cannot be used for the manufacture of building materials due to its unsuitable parameters, such as low pozzolanic properties or high ignition losses (high content of unburned coal).

The aim of the tests presented in this article is to process fly ash (dust) into a form that would significantly reduce the consequences of its storage and transport harmful to the environment, mainly secondary dusting. It was found that granulation would be the right method of processing the ash [20, 21]. Furthermore, it was assumed that the developed method of rendering fly ash harmless should be as simple and an inexpensive as possible. It should not require the use of specialized nor dedicated equipment, nor significantly change the composition of the granulated material and the costs of its use should remain economically justified.

The main aims of the granulation tests include: minimisation of environmental impact related with storing fly ash, mainly secondary dusting, reduction of fly ash storage

and transport costs and keeping the potential possibilities of reusing the processed waste.

2. TESTING METHOD AND MATERIALS

Fly ash granulation tests were done at the laboratory of the Department of Process Equipment of the Lodz University of Technology, Poland. The fly ash undergoing tests was obtained from the Regional Heat Generating Plant No. 1 (RHGP1) of the Heat Supply Company PEC S.A. located in Kalisz, Poland. The ash was derived from the combustion of fine hard coal in WR-10 11.63 MW water boilers with 83% nominal efficiency (PEC 2012). Fly ash was removed from flue gas in a cyclone battery. The ash was sampled for the tests during the normal operation of the heat generating plant, directly from the cyclones in the flue gas purification installation. The tested ash sample was typical of the given dust extraction systems and technology – an average of several days. Grain-size composition of the fly ash used for the test is shown in Fig. 1. Its chemical composition is given in Table 1.
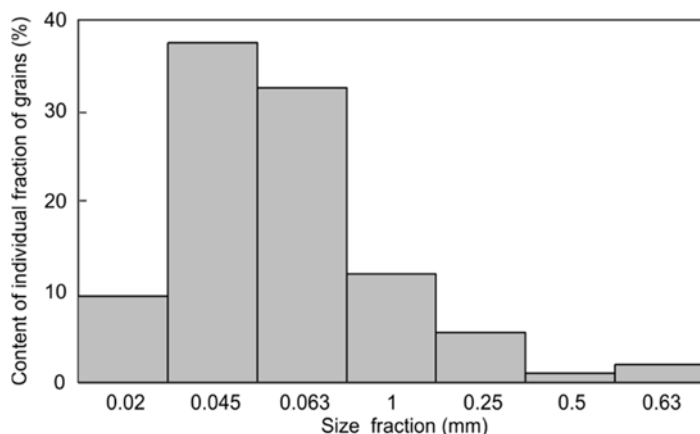

Fig. 1. A particulate composition in the form of a histogram for fly ash

Table 1

Chemical composition of fly ashes from the combustion of fine hard coal

Component	SiO_2	Al_2O_3	Fe_2O_3	CaO	CaO free	MgO	SO_3	Na_2O	K_2O	Cl^-
Content, wt. %	42.1	22.0	5.8	3.5	0.5	2.1	0.5	1.0	2.4	0.017

The samples were granulated in a disk granulator (Fig. 2) equipped with a granulator disk, an electric motor with regulated rotational speed and a granulated deposit moistening

system. A binding liquid was fed directly onto the granulated material using spraying nozzles. The liquid flow rate was controlled using a floating flow meter – a rotameter.
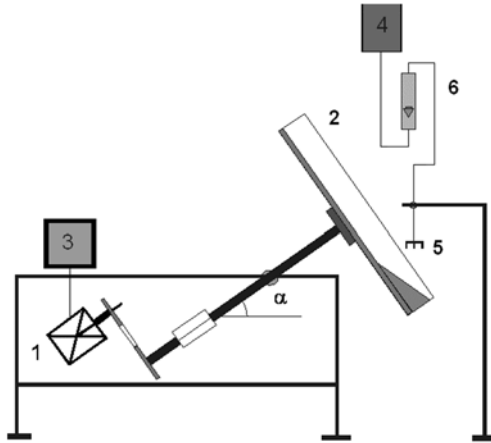

Fig. 2. Test station diagram: 1– motoreductor, 2– granulator disk,
3 – inverter, 4 –liquid tank, 5 – sprinkler, 6 – rotameter

The fly ash from RHGP1 used in the tests was characterized by a very dark color (Fig. 3) suggesting a high content of unburned coal particles. This fact formed the basis for doing an ignition losses test (weight method). During preliminary tests the parameters characterizing the processed ash were determined: density 2.27 g/cm^3, bulk density 0.364 g/cm^3, angle of natural build-up 32°. In test No. 13, fly ash derived from brown coal (source: Bełchatów power plant) combustion was granulated (Fig. 3) (composition of this fly ash was as follows: SiO_2 – 41.0 wt. %, Al_2O_3 – 19.2 wt. %, Fe_2O_3 – 6.6 wt. %, CaO – 23.4 wt. %, SO_3 – 2.1 wt. % [22]). The Bełchatów sample is the ash of brown coal burned in a fluidised boiler. Then, preliminary tests and 19 basic granulation tests were done using different ratios of ash, fillers and binding liquid for the purpose of establishing an optimum method of obtaining a granulated product.

The samples of the ash before the treatment underwent drying in a laboratory drier at 105 °C. The variable process parameters for all of the tests are given in Table 2. In the first series (tests Nos. 1–3), tests were conducted using 500 g samples of fly ash, without fillers and demineralized water was used as the binding liquid (tests Nos. 1–13). In the subsequent series, bentonite was added to the granulate (tests Nos. 4–19), and in the tests Nos. 14–19 waterglass (R140 CAS: 1344-09-8) was used as the binding liquid. The binding liquid was fed at the top of the deposit (tests Nos. 1, 2) and at the bottom of the deposit (tests Nos. 3–19). Binding liquid flow rate Q was 0.83 m^3/s (test No. 1) and 1.25 m^3/s (tests Nos. 2–19). Granulator disk angle of inclination was equal to 35°. As a result of the tests 19 granulated products were obtained in disk agglomeration processes (Fig. 3) 100 granules were selected from the granulated product obtained in each of the tests Nos. 4–19,

that were then dropped onto a concrete floor from a height of 1 m, counting the number
of non-broken granules [23] for the purpose of establishing their breaking resistance.
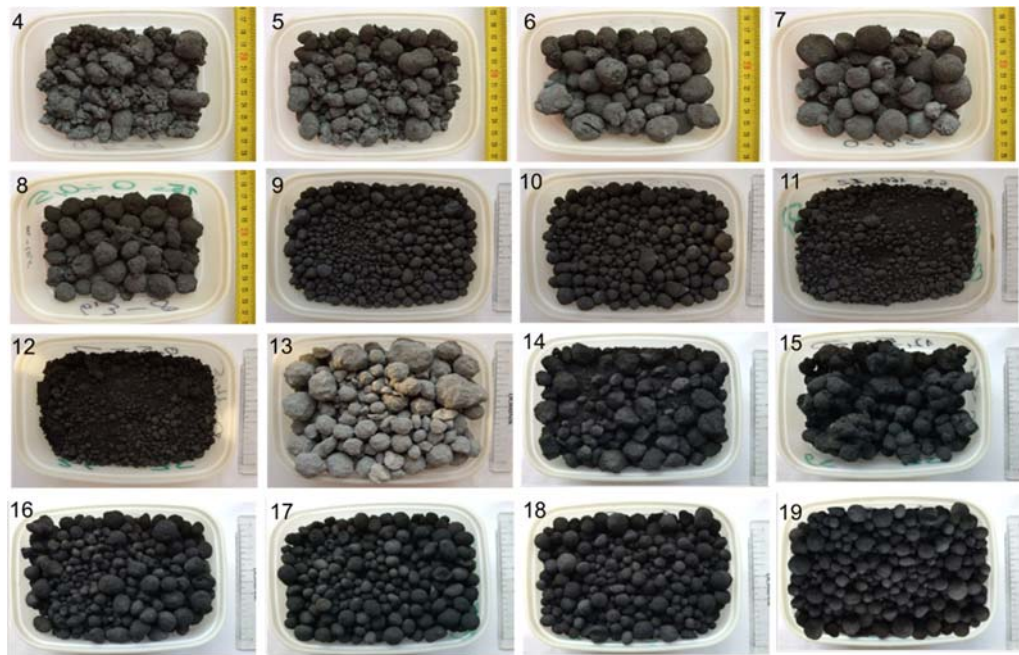

Fig. 3. The granulates obtained in trials Nos. 4–19

3. RESULTS

3.1. PRELIMINARY TESTS

The preliminary research (tests Nos. 1–3) made it possible to determine the opti-
mum parameters of the granulation process, such as:

Binding liquid feeding place. In the first two tests the binding liquid was fed at the
top of the deposit and starting with test No. 3 it was being fed at the bottom of the
deposit. A change of the place of the liquid feeding caused a 10% increase in the amount
of the granulated material. The liquid feeding from the bottom of the deposit was thus
considered optimal.

• *Binding liquid flow rate*. An increase in the flow rate made it possible to obtain a bet-
ter result in a shorter period of time. 20% of ash was granulated in 45 min in the first test
($Q = 0.83$ m^3/s), while 25% was granulated in 30 min in the second test ($Q = 1.25$ m^3/s).

Agglomerates exceeding 1 mm in size were considered granulated material (no secondary dusting). The fraction 0–1 mm was constituted of abraded or non-granulated material [24]. The non-pressure granulation process in the disc equipment consists in the formation of fine-grained (ca. 0–100 μm) material of agglomerates with sizes over 1 mm.

• *Amount of fed binding liquid*. The results of tests 1–3 proved that the best results were obtained with a high liquid flow rate 1.25 m³/s and moisture content 100 wt. % defined as the ratio of the weight of water to the weight of the dry material. Optimum values of the process parameters were determined by comparing the obtained granulation results (particles size) for different tests (PN-ISO 565:2000 *Test sieves. Metal wire cloth, perforated metal plate electroformed sheet. Nominal size of openings*).

• *Disk rotational speed*. Observations of the deposit behavior in the granulator disk and the results of granulation proved that the highest grade of the material granulation were obtained with the rotational speed of 14 rpm.

The preliminary tests revealed that in order to obtain satisfactory granulation degree use of an additional filler and waterglass solution as a binding liquid can lead to better results.

T a b l e 2

Variable parameters of the granulation process

Test No.	Binding liquid	t [min]	t_n [min]	m_p [g]	m_b [g]	W [wt. %]	N [rpm]
1		45	10	500	0		12
2		30		500	0		
3		30		500	0		
4		7		250	250	100	
5		7	6.40	300	200		
6		7		350	150		
7	water	10		400	100		
8		12		450	50		
9		20	6.00	450	50	90	
10		45	5.20	450	50	85	
11		60	5.00	450	50	75	14
12		70	4.20	450	50	70	
13		12	5.20	450	50	80	
14		60	5.20	350	150	80	
15		13	6.00	350	150	90	
16	waterglass solution	9	6.40	350	150		
17		15		400	100	100	
18		25		450	50		
19		35		475	25		

t – process duration, t_n – moistening time, m_p – ash weight, m_b – bentonite weight, W – moisture content, n – rotational speed.

3.2. ANALYSIS OF IMPACT LOOSE ADDITIVE AND BINDING LIQUID

In order to improve the obtained results bentonite was added (tests Nos. 1–3), first in 1:1 ratio. The entire tested sample was granulated. Irregularly shaped granules, 3–15 mm in diameter, were obtained. A very wet granulated product was sticking to the disk at the last stage of the process, making the free flow of the deposit impossible. After the granules were dried, their durability (determined on the basis of organoleptic assessment) was much higher than that of granules from the previous test (without bentonite). This made the pouring of a dry granulated product, and an additional breaking resistance test possible. What is interesting, a dripping wet granulated product was obtained, while bentonite is known for its absorption properties. This suggests that the water absorption of the tested fly ash is greater than that of bentonite itself.

In order to eliminate the problem of the wet granulated product sticking to the granulator disk, in the subsequent tests, starting with test 5, the content of bentonite in the samples was reduced. Furthermore, the tests were aimed at determining the minimal content of bentonite that enables the processing of the sample of the tested ash into a granulated product. Sample No. 5 contained 40 wt. % of bentonite, each subsequent sample – 10 wt. % less. The diminishing of the amount of bentonite in sample No. 8 to 10 wt. % prevented the granulated product from sticking to the disk, thus making it possible for the entire sample to be successfully granulated.
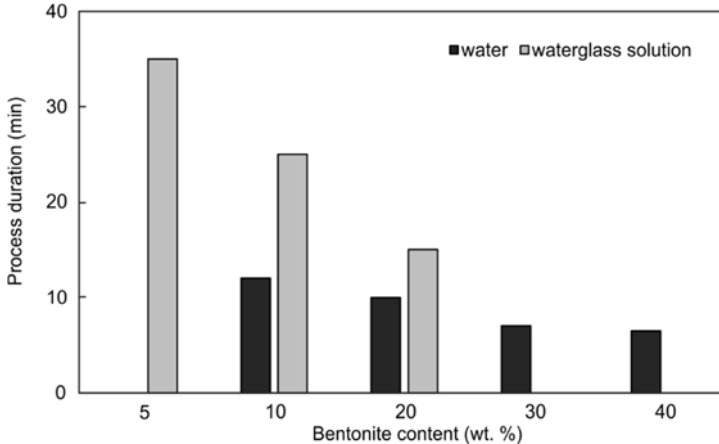

Fig. 4. Granulation time in the function of bentonite share for two types of binding liquids: water and waterglass solution (flow rate $Q = 1.25$ m^3/s, moisture content $W = 100$ wt. %)

In tests Nos. 14–16 30 wt. % addition of bentonite was used while the amount of the waterglass increased in each subsequent test. An increase in the mass of the added binding liquid significantly shortened the duration of the granulation process. In test

No. 14 the addition of the binding liquid equal to 80% of the sample mass made it possible to granulate 1/2 of the sample in 60 min. In test No. 16, the addition of the binding liquid equal to 100% of the sample mass made it possible to granulate the entire sample in as few as 9 min (Fig. 4).

Bentonite is often used as a binding agent in granulation processes being considered a model material. Montmorillonite is the main ingredient of bentonite. It is a silty material that establishes permanent, durable material bonds with the binding agent. The properties of bentonite result from its crystalline structure. It has a surface available for water and exchangeable ions in interpacket spaces. Because of its optimum sorptive properties bentonite is commonly used as an adsorbent. In each subsequent test, starting with test No. 16, the content of bentonite in the overall mass of the tested sample was reduced, i.e., from 30 wt. % in sample No. 16 to 5 wt. % in sample No. 19. The addition of the binding liquid – waterglass – equal to 100% by weight remained unchanged. The content of bentonite in the tested samples affects the duration of the granulation process and the shape of the resultant granules. A high content of bentonite caused fast granulation of the material (small number of fractions below 1 mm), however, the obtained granules had an irregular shape (Fig. 3). A reduction in the bentonite content increased the time necessary for the granulation of the sample, however, the obtained granules had a regular, compact shape. Most of all, restricting the share of additives to the necessary minimum provides a more economical product.

After the minimum content of bentonite in the sample was determined, the next step was to specify the minimum amount of the added binding liquid. Distilled water was used as the binding liquid in the discussed tests. In order to determine this value, 4 tests were done, 9–2, reducing the mass of the added water with each subsequent test – starting with 90 wt. % in test 9 to 70 wt. % in test 12, where 100% is the mass of the liquid equal to the mass of the tested sample. Reducing the mass of the added liquid forces an extension of the process duration, and their shaping and compacting takes more time. The binding agent facilitates the bonding of the agglomerated material particles as a result of the effect of capillary pressure forces caused by interfacial surface tension. The granulation process is related to the deposit humidity, which is a sum of the humidity absorbed from the environment and amount of fed binding liquid. The higher the deposit humidity, the higher the granulation rate is. The presence of water facilitates the material particles binding as a result of interparticle attraction (establishment of strong enough adhesive joints). Furthermore, it may be assumed that the supplied water may react with calcium oxide contained in ash, and thus accelerate the formation of granules.

In test No. 9 (90 wt. % of liquid), 20 min were needed to granulate the sample, while as many as 60 min were necessary for the complete granulation of the sample in test No. 11 (75 wt. % of liquid) (Fig. 5). 70 wt. % of the liquid was added to sample 12, while this amount proved to be insufficient for the complete granulation of the tested material. The granulation of the entire sample requires the addition of the liquid equal to at least 75% of the granulated sample weight.
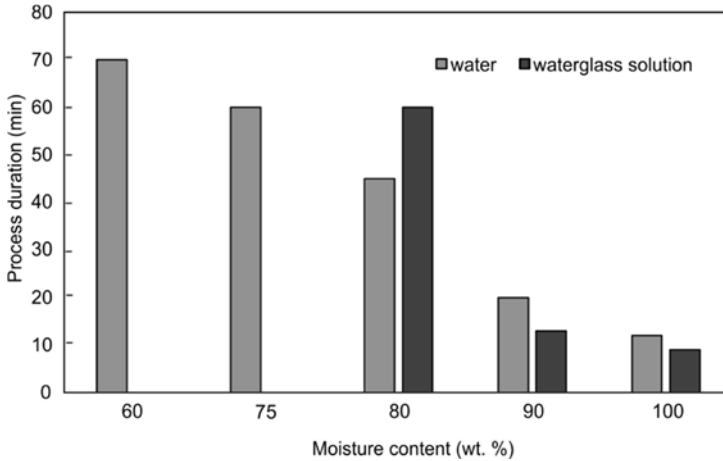
Fig. 5. Effect of moisture content on the duration of a granulation process
for two types of binding liquids: water and waterglass solution

One test using fly ash from another source was done at the end of the series using demineralised water. For reference purposes, in test No. 13 fly ash derived from brown coal combustion from the Bełchatów power plant was granulated. The process parameters are given below. The addition of bentonite amounted to 10 wt. %, while the moisture content of the granulated material equaled 80%. The granulation process was relatively quick, approximately 1/2 of the sample was granulated after the end of moistening (5 min 20 s), and after the next 5 min the entire sample was processed. With the same parameters and 10 wt. % bentonite content as many as 45 minutes were necessary to granulate sample No. 10 (hard coal ash, Kalisz heat generating plant). This results from a significantly lower content of unburned coal particles in the ash from Bełchatów. Loss on ignition (LOI) amounts to 2.4% in the Bełchatów ash and 25.7% in the Kalisz ash. The share of unburned coal is directly linked to the combustion process performance. It increases when technologies reducing nitrogen oxides content in exhaust gases are used in combustion systems.

3.3. GRANULATED PRODUCT BREAKING RESISTANCE TEST

One of the aims of the conducted experiments was to obtain a product that can be transported using non-specialised means of transport. That is why the obtained granulated product was supposed to be durable enough so that it could be poured during loading and unloading. Sufficient breaking resistance would be proven for those samples in which the ratio of non-broken granules is equal to or greater than 98%. Such durability is shown by the granulated product obtained in tests Nos. 15–18. In these samples, the moisture content amounted to 90 wt. % in sample No. 15, and 100 wt. % in the other

samples. The addition of bentonite was variable as well, amounting to 30 wt. % in samples 15 and 16, 20 wt. % in sample 17 and 10 wt. % in sample 18. The obtained results are shown in Fig. 6.
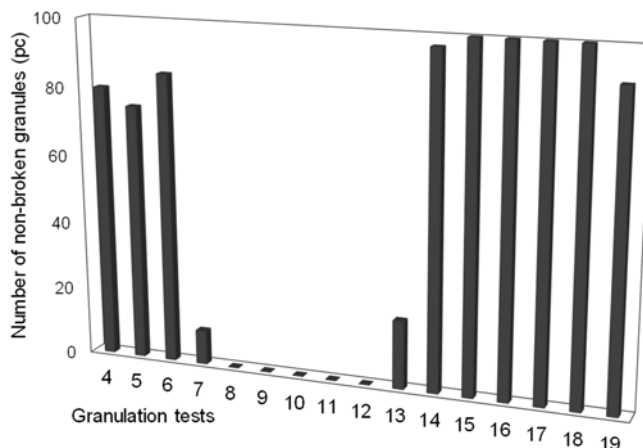

Fig. 6. The results of breaking resistance test

4. CONCLUSIONS

Granulation may be a solution to the problem of local heat generating plants with the storage and reuse of fly ash with ignition losses of over 20%. Efficient granulation of fly ash with a high content of unburned coal requires the use of a viscous binding liquid (waterglass) and a few percent addition of a filler (bentonite). Factors such as the place of the binding liquid feeding, the amount and type of the added binding liquid, the amount of the added filler are of importance for the quality of the obtained granulated product and the rate of the process. The attempts at obtaining a granulated product using demineralised water without the addition of bentonite did not produce the expected results. The samples were not granulated in their entirety.

The addition of bentonite made it possible to completely granulate the material. Granules with a high bentonite content had an irregular shape, while those with a low filler content did not show breaking resistance. Only the introduction of waterglass in place of demineralised water produced the desired effect. The resultant granulated product was characterised by a regular shape and good breaking resistance.

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