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Mortars with addition of local industrial by-products

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Preliminary note

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Mortars with addition of local industrial by-products

Results obtained by studying the possibility of replacing cement in mortars with local industrial by-products, i.e. with silica fume and red mud, are provided in the paper. The results of fresh and hardened mortar properties tests conducted on five mixtures in two series are presented. 10% of cement was replaced with silica fume, and 0%, 5%, 10%, 15% and 20% of cement weight was replaced with red mud. The first series was tested after 28 days of curing, and the second series was subjected to freezing and thawing cycles after proper curing, and then submitted to testing. The results point to satisfactory behaviour of mixtures with a low red mud content.

Key words:
mortar, industrial by-products, silica fume, red mud

Prethodno priopćenje

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Mortovi s dodatkom lokalnih industrijskih nusproizvoda

U radu su predstavljeni rezultati istraživanja mogućnosti pripreme morta zamjenom cementa s lokalnim industrijskim nusproizvodima silicijskom prašinom i crvenim muljem. Prikazani su rezultati istraživanja svojstava svježeg i očvrslog morta na pet mješavina u dvije serije. Cement je zamijenjen silicijskom prašinom u udjelu od 10 posto, a crveni mulj je dodavan u udjelima od 0 %, 5 %, 10 %, 15 % i 20 % s obzirom na masu cementa. Prva serija uzoraka je ispitana nakon 28 dana njegovanja, a druga serija ispitana je nakon propisanog njegovanja uzoraka izloženih ciklusima smrzavanja i odmrzavanja. Rezultati upućuju na zadovoljavajuće ponašanje mješavina s niskim postotkom crvenog mulja.

Ključne riječi:
mort, industrijski nusproizvodi, slicijska prašina, crveni mulj

Vorherige Mitteilung

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Mörtel mit dem Zusatz lokaler industrieller Nebenprodukte

In der Abhandlung werden die Untersuchungsergebnisse der möglichen Mörtelvorbereitung vorgestellt, indem man den Zement durch die lokalen industriellen Nebenprodukte Siliziumstaub und Rotschlamm ersetzt. Dargelegt werden die Untersuchungsergebnisse der Eigenschaften frischen und gehärteten Mörtels an fünf Mischungen in zwei Serien. Der Zement wurde durch Siliziumstaub in einem Verhältnis von 10 Prozent ersetzt, und der Rotschlamm wurde in Anteilen von 0%, 5%, 10%, 15% und 20% unter Berücksichtigung der Zementmasse hinzugefügt. Die erste Serie der Proben wurde nach 28 Tagen Pflege getestet, und die zweite Serie wurde nach der vorgeschriebenen Pflege der Proben, die Gefrier- und Auftauzyklen ausgesetzt waren, getestet. Die Ergebnisse zeigen ein zufriedenstellendes Verhalten bei Mischungen mit einem niedrigen Anteil an Rotschlamm auf.

Schlüsselwörter:
Mörtel, industrielle Nebenprodukte, Siliziumstaub, Rotschlamm

1. Introduction

The demand for alternative materials that can replace natural raw materials for obtaining cement composites is currently on the rise. The production of mortar and concrete, the most widespread cement-based materials, has certainly reached many records figures in recent years. The consumption of natural raw materials in the manufacturing of these composites has increased considerably. Danger to the environment is reflected through the loss of natural resources, as well as in environmental pollution via emission of CO_2 gases during production. One of the first to blame is the cement industry, which is actually one of the largest environmental pollutants. It is directly responsible for about 8 % of total CO_2 emissions in the atmosphere [1]. According to the IMARC group, the total worldwide cement production amounted in 2010 to approximately 3.3 billion tons and, in 2017, this production exceeded 5 billion tons, which is an annual increase of about 7 % [2]. Interestingly, according to some predictions, this volume of cement production was expected to occur no earlier than in 2050 [3].

One of the ideas is the utilization of industrial by-products as raw materials for the production of cement, or for the replacement of cement in composites [4, 5]. Usually, these waste materials are not further used in the industry from which they originate. The use of industrial by-products is justified by an appropriate chemical composition and particle size [6]. For instance, fly ash, silica fume, and limestone have already been successfully used as an addition during cement production. Cement mortar tests involving partial substitution of cement with silica fume and red mud will be presented in this paper.

Red mud is a by-product of the industrial production of aluminium from bauxite ore via the Bayer process. It is estimated that 0.3 to 2.5 tons of red mud are generated from 1 ton of aluminium produced [5]. Bauxite exploitation in the world reached about 270 million tons in 2016, with a total of about 58 million tons of aluminium produced in the same year [7]. This means that over 120 million tons of red mud are generated annually [8]. According to its chemical composition, red mud consists of iron, aluminium, sodium, calcium, and silicon oxides, which is a good basis for research. Chemical composition varies widely depending on the bauxite origin and aluminium production method used. Average values are given in Table 1. Average pH value of red mud is 10 – 12,5, while its particle size is <10 μm [4].

Table 1. Chemical composition of red mud [4]

Component	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	Na_2O
Content [%]	2 - 20	6 - 28	12 - 56	2 - 28	1 - 10

Red mud can not be considered as a full-fledged artificial pozzolan because it does not meet all necessary requirements. However, its index of pozzolanic activity is favourable, and constitutes a good basis for further research [9]. Red mud consists of aluminium and silicon oxides, and can therefore be used as a raw material. Its alkaline character is compatible with cement matrix and, for active formation of CSH gel, pH value should be greater than 11.5 [10, 11]. In other words, hydration will not occur if the pH value is lower than 9.5. The study of hydration of Portland cement to which red mud was added is presented in [12]. It was established that the hydration time was shorten when red mud was added to the sample. Investigations of red mud utilization as clinker have also been reported [13, 14]. In addition, researchers are investigating partial replacement of cement with red mud in mortars and concrete [5, 6, 15]. Durability properties are positively affected by addition of red mud as partial replacement of cement in composites [15]. This is however detrimental to rheological properties of the fresh mix, which must be corrected by adjusting water content [12, 16]. Compressive strength usually decreases with the addition of red mud [9, 16]. On the other hand, some research points to a slight increase in compressive strength when red mud is added [17, 18]. This is dependent on the origin and chemical composition of bauxite [19].

The investigation of cement mortar properties is presented in this paper. Five mixtures in total were prepared with partial replacement of cement with red mud. The latter was added in the quantities of 0 %, 5 %, 10 %, 15 %, and 20 % by mass. Silica fume was added in the constant proportion of 10 % by mass of cement.

2. Materials

The Portland cement CEM II/A-M (S-V) 42,5N, with 20 % of slag and fly ash, was used in this experiment. Crushed and screened limestone was used as the fine-grained aggregate. The grain size distribution of aggregate (sand) is presented in Table 2.

Table 2. Grain size distribution of sand

Aperture [mm]	0.063	0.09	0.125	0.25	0.5	1	2	4	8
Passing [%]	4.46	9.08	13.13	20.62	30.39	44.52	69.26	99.67	100

Table 3. Chemical composition of red mud from Dobro Selo landfill

Component	Al_2O_3	Fe_2O_3	Na_2O	CaO	SiO_2	MnO	TiO_2	Ostatak
Content [%]	16.46	37.85	2.39	10.41	6.79	0.59	1.64	0.516

Table 4. Composition of mortars

Mixture	Cement [kg/m³]	Silica fume		Red mud		Total binder [kg/m³]	Sand [kg/m³]	w/c ratio	w/b ratio
		[%]	[kg/m³]	[%]	[kg/m³]				
CM1	405	10	45	0	0	450	1350	0.711	0.640
CM2	382.5	10	45	5	22.5	450	1350	0.753	0.640
CM3	360	10	45	10	45	450	1350	0.800	0.640
CM4	337.5	10	45	15	67.5	450	1350	0.853	0.640
CM5	315	10	45	20	90	450	1350	0.914	0.640

Red mud was extracted from the Dobro Selo landfill, near Mostar, Bosnia and Herzegovina. It is estimated that approximately 10 million tons of red mud have been deposited in this landfill. The landfill is made of two reinforced concrete pools, in which the red mud is covered with a layer of water. Bauxite processing was stopped in 1991 and, since then, this landfill has not been active [20]. Chemical analysis of the red mud was conducted in 2016, and the corresponding results are presented in Table 3. The presence of sodium makes the red mud potentially harmful to environment, especially if it comes into direct contact with plants [21]. However, radioactivity measurements have shown that there is no direct increased radioactivity of red mud [22], which means that it can be safely used in construction industry. The composition of red mud reveals the presence of aluminium and silicon oxide, as well as of calcium oxide. High iron content gives this material a recognizable red colour. Specific gravity of the red mud was determined by pycnometer method, and it amounts to 3.15 g/cm^3, while density in compacted state is 1.35 g/cm^3. The specific area (fineness) was determined according to EN 196-6 by the air permeability method (Blaine method), and it amounts to 8300 cm^2/g.
The silica fume used in the experiments originates from Jajce, Bosnia and Herzegovina. It is a waste material generated during production of silicon alloys. 9.000 to 10.000 tons of silica fume are generated annually during the production process. Particle size is 0.1 to 0.2 μm, which is approximately 100 times smaller compared to cement. Silica fume was added in liquid state with 44 % of solid matter.

3. Experimental program

The behaviour of fresh and hardened cement mortars, with partial replacement of cement with red mud and silica fume, is analysed in the study. A total of five mortar mixtures were prepared in two series. The composition of these mixtures is presented in Table 4. Silica fume was added in the constant quantity of 10 % by cement mass. Red mud was added as 0 %, 5 %, 10 %, 15 % and 20 % by cement mass. The mixture containing 0 % of red mud was the control (reference) mix. The water binder (w/b) ratio was kept constant of 0,64 for all mixtures, while the water cement (w/c) ratio was variable (Table 4). All samples were prepared according to BAS EN 1015-2:2004.

3.1. Fresh mortar testing

Rheological properties, bulk density, and air content were tested in the scope of fresh mortar testing. As to rheological properties, mortar consistency was tested by flow table according to BAS EN 1015-3:2004. The automatic flow table was used in this experiment, which complies with requirements of the standard. Bulk density was determined according to BAS EN 1015-6:2004. Air content was determined according to BAS EN 1015-7:2004. The standard offers two test methods: Method A "Pressure method" and Method B "Alcohol method". Method A was selected for this experiment.

3.2 Hardened mortar testing

The bulk density, flexural strength, compressive strength, and frost resistance values were tested in the scope of the hardened mortar testing. Flexural and compressive strength were tested according to BAS EN 1015-11:2002. Fresh mortar was prepared in three-piece steel moulds, and so three samples were taken for each mixture. Samples were poured into steel moulds at the vibrating table, and were then cured for no less than 24 hours, i.e. until reaching the strength that enables safe demoulding. After that, samples were cured for 28 days in water. Five mixtures from the first series were tested immediately after curing, and five mixtures from the second series were placed in the freezer and exposed to 25 freeze – thaw cycles. Freezing temperature was -20°C, while thawing temperature was +20°C. Frost resistance was determined by comparing compressive strength results of the two series.

4. Test results

4.1. Fresh mortar test results

To ensure proper fluidity of mortar, the measured spread of mortar on the flow table should be greater than 140 mm [19,

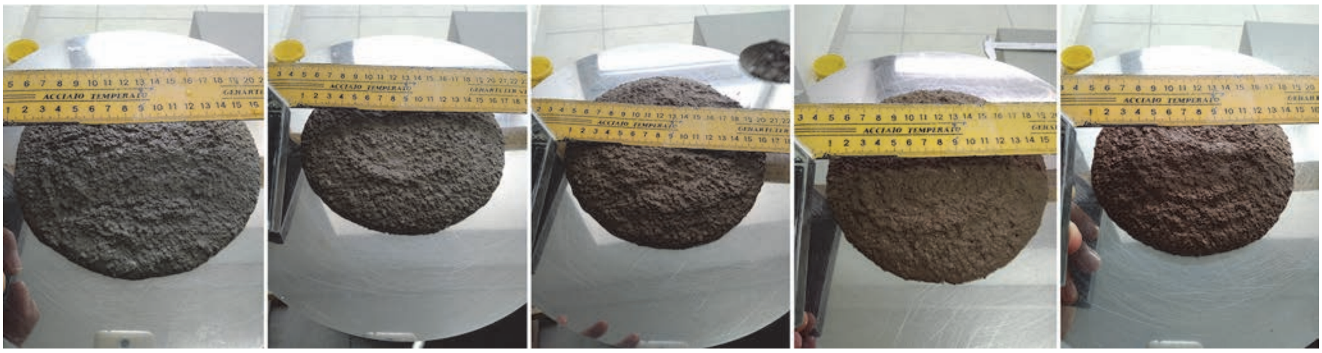

Figure 1. Testing of fresh mortars (from left to right mix CM1, CM2, CM3, CM4 and CM5)

23]. The mortar consistency, i.e. an average measured spread on the flow table is presented in Table 5. The average spread decreases from 179 mm for CM1 mixture, to 160 mm for CM5 mixture. Mortar consistency is significantly affected by the addition of red mud, since red mud particles are considerably finer than cement particles, and they require more water for wetting. It should also be noted that the presence of silica fume only enhances this effect. It is important to emphasize that the water binding ratio is equal for all mixtures, i.e. the amount of water is unchanged. Consistency tests for all mixtures are presented in Figure 1.

Table 5. Average test results for fresh mortar (two series)

No.	Mixture	Spread diameter [mm]	Air content [%]	Bulk density [kg/m³]
1	CM1	179	1.85	2202.0
2	CM2	174	1.75	2199.5
3	CM3	168	1.70	2202.5
4	CM4	167	1.68	2207.5
5	CM5	160	1.60	2211.7

Average test results for air content in fresh mortar are presented in Table 5. Air content in fresh mortar is also affected by particle size. Smaller particles fill the voids and reduce air content. Average values of air content decrease from 1.85 % for CM1 mixture to 1.60 % for CM5 mixture.

Average bulk density values for fresh mortar are also presented in Table 5. An average bulk density value for CM2 mixture is slightly lower compared to CM1. Bulk density for mixtures CM3, CM4, and CM5 is higher compared to CM1. The CM5 mixture has the highest average value of 2211.7 kg/m^3.

4.2. Hardened mortar test results

The samples were weighed and measured after 28 days of curing. Bulk density results of hardened mortars are presented in Table 6. Average values of all test results per mixture were compared to the control mixture CM1 (that does not contain red mud). An increase in bulk density was registered for all mixtures. Only CM3 mixture exhibits a slightly lower increase in bulk density. CM5 mixture shows highest bulk density, which is by 1.30 % higher compared to the control mix CM1.

The flexural and compressive strength of the series 1 hardened mortar was tested immediately after curing. Flexural strength results are presented in Table 7, and average compressive strength values (for 2 results per sample) are presented in Table 8. All mixtures reveal decrease in flexural strength compared to control mix, with the highest decrease registered for CM5 (16.54 %).

Table 6. Bulk density test results for hardened mortar

Mixture	Bulk density [kg/m³]				
	CM1	CM2	CM3	CM4	CM5
Series 1	2237.9	2247.7	2243.0	2268.4	2276.2
	2230.9	2244.1	2238.3	2243.4	2259.8
	2233.6	2263.7	2234.4	2268.8	2282.8
Series 2	2262.0	2246.1	2253.1	2263.7	2274.2
	2241.0	2247.7	2251.2	2250.0	2258.2
	2237.1	2271.9	2255.5	2252.0	2265.6
Average [kg/m³]	2240.4	2253.5	2245.9	2257.7	2269.5
Increase/ Decrease [%]	-	0.58	0.24	0.77	1.30

Table 7. Flexural strength test results, series 1

No.	Mixture	Flexural strength [MPa]			Average [MPa]	Standard deviation	Increase/Decrease [%]
1	CM1	10.08	10.31	10.78	10.39	0.357	-
2	CM2	10.08	9.38	9.61	9.69	0.357	-6.77
3	CM3	9.38	8.91	9.14	9.14	0.235	-12.03
4	CM4	9.61	7.73	8.91	8.75	0.950	-15.79
5	CM5	9.38	7.97	8.67	8.67	0.705	-16.54

Table 8. Average test results for compressive strength, series 1

No.	Mixture	Compressive strength [MPa]			Average [MPa]	Standard deviation	Increase/Decrease [%]
1	CM1	44.28	44.88	44.56	44.57	0.806	-
2	CM2	42.66	42.34	42.25	42.42	0.303	-4.84
3	CM3	40.00	38.22	39.78	39.33	1.194	-11.76
4	CM4	37.91	37.22	37.31	37.48	0.560	-15.91
5	CM5	37.03	35.75	37.28	36.69	0.891	-17.69

Table 9. Flexural strength test results, series 2

No.	Mixture	Flexural strength [MPa]			Average [MPa]	Standard deviation	Increase/Decrease [%]
1	CM1	4.69	4.45	4.69	4.61	0.139	-
2	CM2	4.92	5.16	4.92	5.00	0.139	8.46
3	CM3	4.45	4.45	4.22	4.37	0.133	-5.13
4	CM4	4.45	4.92	3.52	4.30	0.712	-6.80
5	CM5	3.75	3.52	3.75	3.67	0.133	-20.32

Table 10. Average compressive strength results, series 2

No.	Mixture	Compressive strength [MPa]			Average [MPa]	Standard deviation	Increase/Decrease [%]
1	CM1	42.16	35.94	42.72	40.27	3.377	-
2	CM2	40.13	41.16	42.50	41.26	1.622	2.46
3	CM3	37.63	36.16	38.38	37.39	1.467	-7.16
4	CM4	36.25	36.16	36.75	36.39	0.954	-9.65
5	CM5	29.31	30.66	31.50	30.49	1.086	-24.29

Similar to flexural strength, compressive strength also decreased for all mixtures with red mud when compared to control mix. The highest decrease was registered for CM5 mixture (17.69 %). Such decrease is expected, since lower pozzolanic activity is exhibited by red mud in this condition [10, 16].

Just like for series 1, flexural strength for series 2 was tested prior to compressive strength. The corresponding results are presented in Table 9. Samples belonging to series 2 were exposed to 25 freezing and thawing cycles after 28 days of curing. Freezing temperature was -20°C, while thawing temperature was +20°C. Flexural strength results for series 2 are significantly lower compared to those registered for series 1. CM2 mixture shows an increase in flexural strength (8,46 %), while CM3 and CM4 mixtures exhibit slightly lower results compared to control mix. CM5 mix exhibits a notable decrease in strength compared to control mix.

Average compressive strength test results for series 2 are presented in Table 10. The average value for CM2 mix shows a slight increase of 2.46 % compared to control mix. Mixtures CM3 and CM4 show a decrease in strength of up to 10 %, while CM5 mixture shows a substantial decrease in compressive strength. Similar flexural and compressive strength results were registered for series 2. In both cases (flexural and compressive strength) CM2 mix showed slightly better properties compared to control mix. Also, the intensity of decrease in the flexural and compressive strength of the CM3 and CM4 mixtures in similar. This can be explained by the presence of fine red mud particles that seal the pores and reduce porosity (which is evident from the air content test), i.e. they reduce the amount of water in the sample that can be frozen, and the amount of the cement is still large enough for hydration process.

Frost resistance of mortars was tested according to literature recommendations [23]. The resistance was assessed by comparison of compressive strengths for series 1 and 2, as shown in Table 11. Reduction of strength is not uniform for all mixtures. CM1 and CM5 mixtures show a substantially greater decrease in strength compared to CM2, CM3, and CM4 mixtures

Table 11. Comparison of compressive strength test results for series 1 and 2

Mixture	Average compressive strength of series 1 [MPa]	Average compressive strength of series 2 [MPa]	Decrease [%]
CM1	44.57	40.27	-10.68
CM2	42.42	41.26	-2.80
CM3	39.33	37.39	-5.21
CM4	37.48	36.39	-3.01
CM5	36.69	30.49	-20.33

According to recommendations given in [23], a mortar can be considered resistant to frost if the decrease in compressive strength is lower than 20 %. This requirement has been met by all mixtures with the exception of CM5. The decrease in compressive strength is the lowest for CM2 mixture, where it amounts to only 2.80 %. In addition to strength, weight comparisons were also made, but no mass loss was registered for any of the studied mixtures.

5. Conclusion

The paper presents research on the properties of fresh and hardened mortar to which locally available industrial by-products, silica fume and red mud, were added. The following conclusions can be drawn from the presented results:

- Consistency and air content of fresh mortar reduce with an increase in red mud content in the mixture.
- Bulk density increases with an increase in red mud content, but this increase is not significant and amounts to maximum 1.30 % for the mix with 20 % of red mud, compared to the mix without red mud.
- Flexural and compressive strength test results for samples from series 1 decrease with an increase in red mud content. Highest decrease was registered for mixture with 20 % of the red mud (CM5), where it is 16.54 % for flexural strength and 17.69 % for compressive strength.
- Samples tested after exposure to 25 freezing and thawing cycles (series 2) exhibit a considerably different behaviour. Flexural and compressive strength for mix with 5 % of red mud (CM2) shows an increase in both strengths: 8.46 % for flexural strength and 2.46 % for compressive strength compared to control mix. Other mixtures show a decrease in strength compared to control mix.
- Based on comparison of compressive strengths in the first and second series, frost resistance is satisfactory for all mixtures, except for mixture CM5. Much better behaviour was observed for the mixtures containing red mud.

This research has shown that it is possible to prepare mortars by substituting cement with locally available industrial by-products. Samples with higher red mud content (over 15 %) show significant decrease in mechanical properties, while mixtures with low red mud content (5 %) behave satisfactorily. In addition, it should be noted that the water binder ratio was the same for all mixtures in this study. Mortar behaviour should be further examined by retaining the same water cement ratio, and a variable water binder ratio and, optionally, by involving the use of additives. Due to fineness of red mud particles, the samples have proven to be more durable after exposure to freezing and thawing, which is especially true for the mixture with 5 % of red mud. This should however be confirmed by additional tests.

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