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Towards the Cleaner Production of Cementitious Materials with the Synergistic Addition of Granite Powder Waste and Fly Ash

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The production of building materials is one of the most polluting industries for the natural environment. To change this, solutions are being sought that can reduce the environmental impact of the construction industry. One such solution is the modification of cement mixes with the addition of supplementary cementitious materials (SCMs). Such materials include granite powder waste and fly ash. Granite powder waste is generated when processing granite rocks, and fly ash is produced by burning fossil fuels. Both materials cause significant environmental problems. Their addition to cement-based materials may not only improve the condition of the natural environment, but also reduce the amount of cement used by the construction industry. The possibilities of reducing the amount of cement in cement mixes by using these materials were determined. It was concluded that the combined use of these materials in cement mixes enables the achievement of better mechanical properties, a lower amount of consumed embodied carbon dioxide (ECO_2), and a decreased price of the mix. In conclusion, the most favourable proportions of both materials in cement composites were identified with regards to mechanical and environmental aspects.

1. Introduction

Granite powder waste is a co-product obtained when processing granite rocks (Chajec, 2021). In the last few decades, the use of rock products as tombstones, overlays of floors, and rock plates has significantly increased (Sadowski et al., 2022). This process is connected with the increased production of granite powder waste (GPW). GPW contributes to environmental pollution. This kind of waste, due to it not having any industrial use, is mainly stored in heaps (Jeyaprabha et al., 2017). Nevertheless, due to the size of grains of granite powder, which is similar to that of cement, GPW is often used as a filler in cementitious mixes (Belebchouche et al., 2021). The filler fills the air voids in a mix with small particles, and in turn the mix has a higher density and better mechanical parameters (Chu and Kwan, 2019). Chu (2019) described the importance of a filler in cementitious mixes, and observed that the technology of its production has a significant impact on the properties of fresh mixes – an appropriately designed mix may allow a more desirable consistency, air content, and workability of the mix to be achieved. Chen et al. (2021) investigated the effect of fine grains on the packing density of cement mixes. They concluded that it is possible to calculate the optimal addition of fine grains of aggregate in order to increase the mechanical properties of composites. Another way to use granite powder in cementitious materials is to replace it with fine aggregate. In some countries, there is a problem with regards to accessing fine aggregate that can be used to produce concrete. Attempts have been made to replace fine aggregate with the addition of granite powder waste. Manikandan and Felixkala (2017) studied the effect of replacing fine aggregate with the addition of granite powder waste on the properties of self-compacting concrete (SCC). They observed that granite powder has a great potential to partially replace fine aggregate in SCC. They did not observe significant changes in the workability of a mix, or a decrease in the mechanical properties of SCC modified with granite powder. However, there are no descriptions in literature of attempts to replace cement with granite powder. It is particularly important to emphasise that cement is responsible for almost 8 % of the production of carbon dioxide (CO_2) related to human activity (Dobiszewska, 2017). Therefore, it is necessary to

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seek the so-called Supplementary Cementitious Materials (SCM), which will enable the amount of used cement to be reduced. The best way to test the potential of using granite powder waste as SCM is to compare its effect with the most popular SCM - fly ash (FA). Fly ash is a waste generated during the combustion of hard coal. Due to its good reactivity and pozzolanic properties, the use of fly ash has increased so significantly in recent years that the material is no longer seen a waste, but instead as a desirable product. Figure 1 shows the production process of granite powder and fly ash, and also tests that were conducted to analyse the properties of mixes modified with the addition of SCMs.

Figure 1: Production of cementitious materials with the synergistic addition of granite powder waste and fly ash

The key aim of this paper is to compare the effects of granite powder waste and fly ash with regard to the environmental and mechanical aspects of cementitious composites produced from these materials. To achieve this goal, the mechanical, economical, and ecological properties of composites were investigated.

2. Materials and Methods

Nine test series were performed as part of the research. Table 1 shows the chemical composition of the ingredients used in the research. Table 2 presents the compositions of the tested series (OPC – reference series with 100 % of Ordinary Portland cement CEM I 42.5R (OPC), GPx/Fax – series modified with the replacement of OPC with x% addition of granite powder (GP) or fly ash (FA), GpxFAx – series modified with the replacement of OPC with simultaneous addition of x% of GP and FA). The compositions of the mixes used in the research series are similar to those that are currently used and described in the literature (see Table 5).

Table 1: Chemical compositions of the raw materials used in the tests

	CaO	SiO_2	Al_2O_3	K_2O	SO_3	MgO	FeO	NaO	Fe_2O_3
OPC	56.62 %	16.03 %	6.10 %	7.32 %	6.62 %	2.61 %	2.26 %	0.00 %	2.44 %
FA	3.77 %	43.40 %	33.96 %	7.55 %	0.00 %	3.77 %	3.02 %	2.64 %	1.89 %
GP	8.30 %	53.63 %	19.90 %	3.29 %	0.00 %	5.54 %	3.46 %	3.46 %	2.42 %

Ordinary Portland cement CEM I 42.5R (Górażdże, Poland), fly ash (Kogeneracja, Wroclaw, Poland) and granite powder waste (Strzegom, Poland) were used in the tests. To mix the ingredients, the quantity of raw materials was precisely measured. They were then placed in a mixer and mixed for 3 min. After this time, the amount of ingredients was measured, and then water and plasticiser were added and stirred for 5 min. The samples prepared in this way were stored for 24 h, after which they were demolded. Six samples were prepared for each test. The prepared samples were then placed in a container full of water to mature. The compressive strength of the samples was determined using a testing machine after 28 days of curing. For this purpose, six samples of each test series, with dimensions of 40x40x80 mm, were subjected to compression in a testing machine (Figure 2) until they were destroyed. The destructive force for each of the samples was recorded, and then the compressive strength was calculated as the mean value of the obtained results. Afterwards, the results were statistically processed.

Table 2: Compositions of the tested series

Test series	Binders (kg/m³)			Ingredients (kg/m³)		
	OPC	FA	GP	Aggregate	Superplasticizer (SP)	Water
OPC	380	0	0			
GP10	342	0	38			
GP20	304	0	76			
GP30	266	0	114			
FA10	342	38	0	1520	1.44	152
FA20	304	76	0			
FA30	266	114	0			
GP20FA10	266	76	38			
GP10FA20	266	38	76			

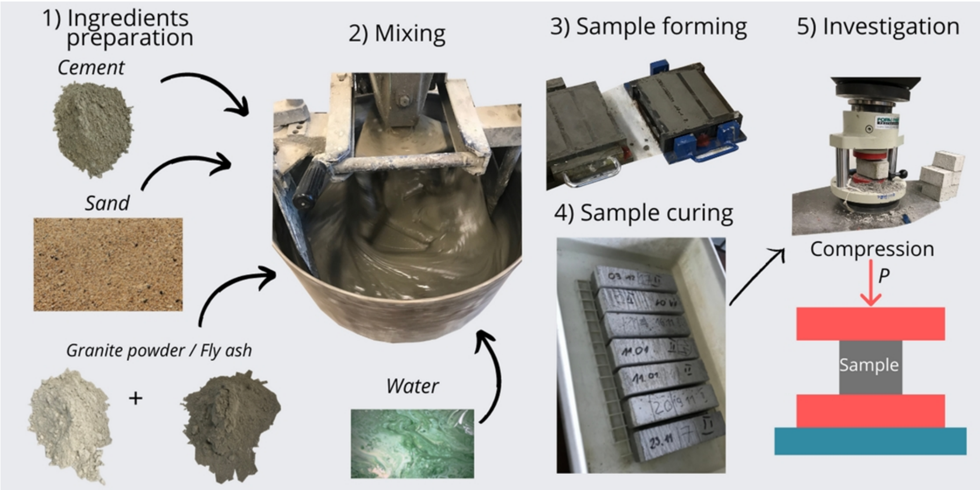

Figure 2: Production and testing of the samples

3. Results and Analysis

Currently, possibilities of reducing the amount of cement in cement mixes are being actively sought. As noted above, it is possible to substitute 30 % of the amount of cement in the mix with the addition of siliceous fly ash or granite powder waste. The addition of FA and GP enables the achievement of improved mechanical properties and durability of cementitious materials.

Table 3 presents the assumptions for the analysis of the environmental impact of the used ingredients. The adopted unit cost of ingredients was estimated based on the prices offered by producers in Poland in the first quarter of 2021 (the average of three offers). ECO_2, defined as the CO_2 produced over a defined period of the life cycle of a product, was estimated based on a publication by Chu in 2021.

Table 3: Assumptions for the analysis of the embodied CO_2 emission of the raw materials used in the tests

Material	OPC	GP	FA	Water	SP	Aggregate
Cost (€/t)	74.66	12.25	34.25	5.00	2,000	12.20
ECO_2 (kg CO_2/kg)	0.88	0.028	0.57	2.5×10^{-7}	0.01	0.008

Cement is the main, and most expensive material used in cementitious mixes. Granite powder is significantly cheaper, and also has a very low ECO_2/kg value. Fly ash is described by providing its average price and the produced ECO_2. Based on the compositions of the tested series and the factors presented in Table 3, the cost and embodied CO_2 (ECO_2) emission for all the investigated series is presented in Table 4.

Table 4: Compressive strength, cost of materials, and embodied CO_2 of the tested series

Test series	Compressive strength after 28 days (MPa)	Cost of materials (€/m^3)	Cost Reduction (%)	ECO_2 of the mix (kg CO_2/m^3)	Reduction of ECO_2 of the mix (%)
OPC	48.54	50.55	0	346.59	0
GP10	45.28	48.18	-4.69	314.21	-9.34
GP20	47.25	45.81	-9.38	281.84	-18.68
GP30	42.56	43.44	-14.07	249.46	-28.02
FA10	41.25	47.72	-5.61	334.81	-3.40
FA20	38.95	44.88	-11.22	323.03	-6.80
FA30	36.86	42.04	-16.84	311.25	-10.20
FA20GP10	42.54	43.35	-14.26	290.65	-16.14
FA10GP20	48.56	44.65	-11.69	257.87	-25.60

Choosing the most optimal mix composition is a challenging task. It is usually performed with the help of computational algorithms, but thanks to the conducted research it is possible to present graphs of the relationship between the compressive strength (mechanical properties) and the ecological and price properties of cementitious composites (Figure 3a and 3b).

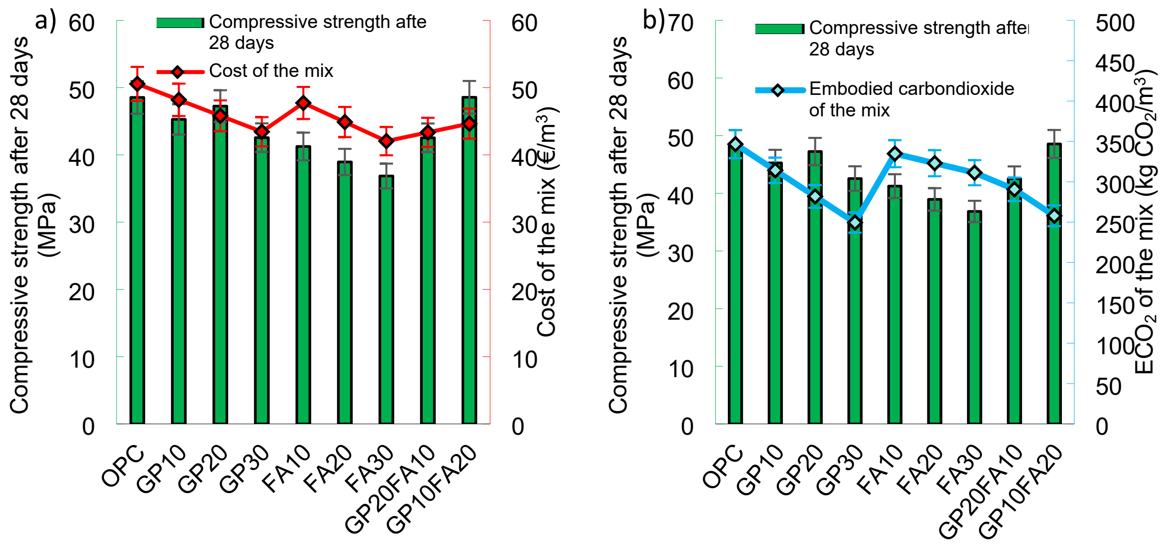

Figure 3: The comparison of the compressive strength and: a) cost of the mix, b) embodied carbon dioxide of the tested series

The mechanical properties of cementitious composites depend on the properties of the ingredients used in mixes. Replacing cement with granite powder (with appropriately selected graining) leads to nonsignificant reduction the compressive strength of the obtained material (Figure 3). A higher decrease of compressive strength is observed in the composites with the addition of fly ash, which were tested after 28 days. Opposite conclusion was drawn by researchers Nepomuceno et al. (2012). They observed improved mechanical properties (tested after 90 days) in composites modified with fly ash, which could be due to the pozzolanic reactivity of the fly ash. Synergistic use of FA and GP leads to improve compressive strength after 28 days (GP10FA20 research series) compared with OPC, GP, and FA series. It is desirable in using two different materials and it indicates a high potential for industrial use of those materials as supplementary cementitious materials to decrease the consumption of cement. Analysing the costs of the production of mixes we can observe that cement is the most expensive material used in mixes (Figure 3a). It may be observed that the addition of granite powder waste and fly ash allows the cost of producing composites to be reduced. Synergistic use of GP and FA leads to an increase in the strength/cost ratio of composites. Figure 3b presents the relation between the compressive strength of composites and the embodied carbon dioxide emitted during the production of mixes. Granite powder and fly ash allow a lower emission of ECO_2 to be achieved in the production of mixes. Especially use of GP leads to a significant decrease in ECO_2 emission of composites. We observed the positive effect of synergistic use of granite powder and fly ash in mixes. The use of supplementary cementitious materials allowed the cost of cementitious mixes to be reduced by even 17 %, and the emission of ECO_2 in the composite production process to be decreased by 28 %. The increase in mechanical properties of composites modified with the addition of granite powder (especially observed for the GP10FA20 series) is due to the synergistic effect of the addition of granite powder and fly ash. Gupta and Vyas (2018) observed that the addition of granite powder in cementitious mixes causes an improved packing density and the filling of the air pores of the mixes which may influence the strength properties of composites. Supit et al. (2014) stated that the use of fly ash in cement mixes increases the mechanical properties of cementitious composites. A high volume (more than 10 %) of fly ash provides for increased consumption of hydrogen carbonates in a cementitious system. It improves the compressive strength and the degree of hydration in composites. Due to the joint application of granite powder and fly ash, it was possible to obtain a synergistic effect that increased the compressive strength of the synergistic series to a greater extent than the series where GP and FA were used separately.

The mechanical properties of cementitious composites are the main characteristics considered when designing building structures. The materials to be used in a structure are chosen based on the compressive strength and class of concrete or mortar. Comparing the economic aspects of the process and ecological properties (cost and ECO_2 of mixes) to mechanical properties is crucial to evaluate the possibility of using the mix in the building. Figure 3a allows for the conclusion that the cost of a mix is not directly connected with the mechanical properties of composites. Similarly, analysing Figure 3b leads to conclusions that environmental impact is not strictly associated with the strength properties of composites.

There are currently many research gaps related to the optimisation of the environmental footprint of cementitious materials modified with the addition of granite powder waste. This waste should be commonly used, especially to reduce the amount of cement in cementitious mixes. To discuss the results obtained in this research, Table 5 presents the comparison of the results of different authors who examined the impact of granite powder on cementitious composites.

Table 5: Comparison of literature results of granite powder waste-modified cementitious composites

Reference	Scale in which the composite was tested	Curing conditions	Investigated after (days)	Additive	Replacement ingredient	Compressive strength development / regress (%)
				GP20		**-5.18**
This research	**Mortar**	**Water cured**	**28**	**GP10FA20**	**Cement**	**+1.28**
				GP30		+0.89
(Gupta and Vyas, 2018)	Mortar	Water cured	28	GP40	Sand	+2.04
				GP10		+8.64
(Zhang et al., 2019)	Concrete	Autoclaved cured	28	GP25	Cement	+9.14
(Rojo-López et al., 2020)	Concrete	Water cured	28	GP8	Cement	-2.84
				GP20	Cement	-21.25
(Asadi Shamsabadi et al., 2018)	Concrete	Water cured	28	GP40	Cement	-30.87

The use of granite powder waste in cementitious mixes has recently significantly increased, especially in the technology of replacing cement. Using granite powder as a replacement for cement has great potential. The authors of the cited publications noted that it is possible to further optimise this technology using the method of improving packing density. This research showed that the synergistic addition of granite powder and fly ash leads to the best mechanical properties of the obtained composite.

4. Conclusions

Granite powder and fly ash have a great potential of being used as Supplementary Cementitious Materials in cementitious mixes. The replacement of cement with granite powder leads to the improved mechanical properties of composites (if their composition was appropriately optimised). Granite powder improves the mechanical properties of cementitious composites when compared to fly ash. In turn, replacing cement with the addition of fly ash decreases the mechanical properties of cementitious composites. Both used SCMs allow for the cost and emission of ECO_2 to be reduced. This research proved that granite powder and fly ash, when used separately in the mix, do not increase the analysed properties when compared with the reference series. However, when used together, these materials enable the mechanical properties of cementitious composites to be improved. In the future, additional research on composites with the addition of granite powder waste, as well as a detailed analysis of the footprint of granite powder waste-modified composites, should be performed. Granite powder waste has an exceptional potential to improve the impact of cement-based materials on the environment.

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