Research paper

Influence of sand substitution with waste lime powder on the concrete carbonation

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Abstract: The concept of sustainability requires that waste-modified materials also demonstrate adequate sustainability. This paper examines the effect of modifying cement concrete with waste lime dust on the course of concrete carbonation. The waste dust comes from the dedusting of aggregate for use in HMA – Hot Mixture Asphalt. The aim of the study was to examine whether the partial replacement of sand with waste powder would have a negative effect on the potential durability of a reinforced concrete element made of this concrete. To determine the extent of carbonation, an experimental plan was prepared including the execution of concretes with varying levels of substitution and a variable water/cement ratio. In order to identify long term influence the test was performed as indicated in EN 12390-12, but with the test time extended to 560 days. The results obtained were statistically analysed and the predicted maximum extent of carbonation depending on the level of substitution and the water/cement ratio was determined. The analysis indicates that it is possible to substitute sand with waste limestone dust without having a negative impact on the extent of carbonation, and thus on the durability of the reinforced concrete structure.

Keywords: lime waste powder, carbonation, statistical analysis, long term examination, cement concrete, durability

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1. Introduction

One of the directions of material engineering development is to fulfill the idea of sustainable development. On the one hand, this involves managing the waste products, and on the other hand, the production of building materials that meet the requirements of achieving durability under the foreseen exposure conditions. One of the key environmental impacts under the rules of EN 206 standard is carbonation. Therefore, the analysis of the possibility of using the waste for the production of cement concrete should also include studies of the impact of using this waste on the carbonation process, because it influences the durability of reinforced concrete. The aim of the paper is to examine whether the partial sand replacement with waste powder would have a negative effect on the potential durability of a reinforced concrete element made of this material.

1.1. The waste lime powder

One of the preparatory processes in the production of Hot Mixture Asphalt (HMA) is the preparation of the aggregate. This process includes drying, dusting and heating. As a result, waste is collected in the filters of the dryers as a mineral powder. Due to the most commonly used aggregates for the production of HMA the waste in the form of lime or basalt powder is obtained.

The limestone powder chemical composition tests shows that the main component of the powder is CaO, which content calculated as $CaCO_3$ is 84.9% wt. Content of SiO_2 is 7.1% wt. and Al_2O_3 content is 2.4% wt. (their carriers are aluminosilicates occurring in the form of feldspars and clay minerals, and quartz) [1]. The scanning electron microscope was used for current investigation and it turned out that the shape of the grains is smooth and uniform (Fig. 1, [2]).
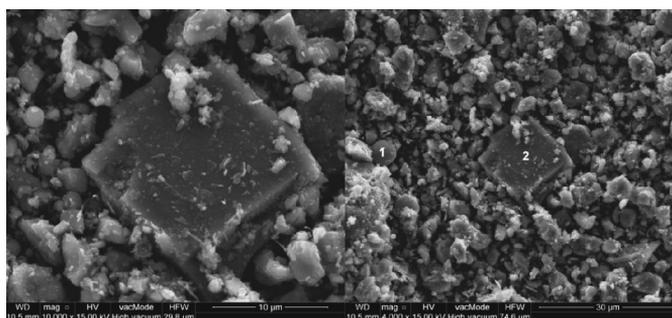
Fig. 1. Microscopic photograph of waste limestone powder grain shape,
1 – aluminium silicate grain, 2 –l imestone grain [2]

The obtained specific area for tested powder is $26\,444$ cm^2/cm^3. The average diameter is 27.5 μm and the mode diameter was 14.2 μm [1].

Research conducted on basalt powder indicates its potentially high usefulness in the production of cement concrete. [3–5]. Also conducted works on the analysis of features [2] and the use of lime powder [1, 6, 7] are promising. The conducted research indicates the possibility

of using a waste powder as a type I additive, i.e. an almost inert additive according to EN 206 regulations. The quoted studies concerning lime powder indicate that with the substitution of up to 20% – with regard to the weight of cement – powder does not significantly affect the reduction of durability in a chemically aggressive environment. The effect of replacing part of the sand with lime powder waste, depending on the water/cement ratio, has not been analyzed so far (Fig. 2).
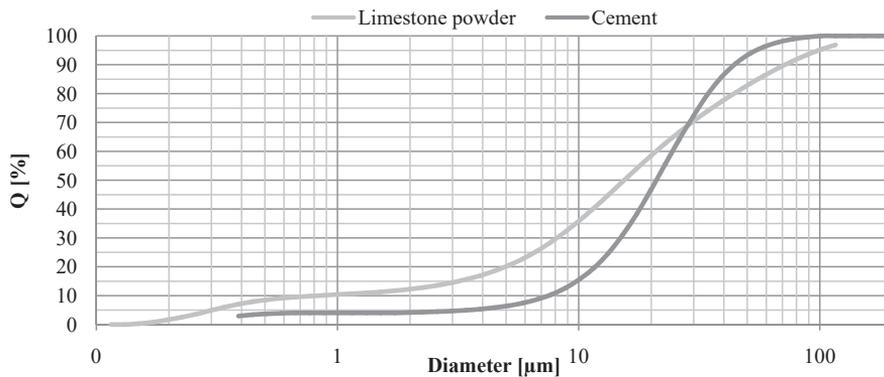
Fig. 2. Verification of grain size distribution curve of tested limestone powder in comparison to cement– cumulative frequency plot

1.2. Carbonation process

Carbonation leads to a reduction in the pH of the concrete cover and, consequently, to corrosion of the reinforcement due to steel deposition. Carbonation is a complex of physicochemical transformations of concrete under the influence of long-term influence of carbon dioxide, which is found both in the atmospheric air and in the internal atmosphere of buildings [8]. The factors influencing the carbonation of concrete can be divided into external and internal.

The basic internal factor on which the carbonation rate depends is the CO_2 diffusion coefficient [8, 9]. This, in turn, is a function of the pore distribution in hardened concrete, resulting, among other things, from the W/C coefficient [9]. Other internal factors affecting the course of carbonation are: cement content – the higher content effects in the lower susceptibility to carbonation [10,11], clinker characteristics, degree of fineness of cement – the higher specific surface the slower the carbonation course [12].

Another internal factor on which carbonation depends is the type and amount of additives to cement and concrete. Type I additives (almost inert) similarly to aggregate grains do not undergo carbonation. However, they affect the CO_2 diffusion coefficient by densifying the structure [8].

A more complex process is the influence of type II additives (with pozzolanic or latent hydraulic properties). On the one hand, they have an inhibiting effect by tightening the structure, and on the other hand, they may increase the depth of carbon dioxide penetration. Increasing the penetration depth of CO_2 according to [8] is associated with a reduced content of $Ca(OH)_2$ in concrete with additives.

Therefore, although the tested waste dust was initially classified as a type I additive, it is necessary to check its impact on the concrete carbonation process.

2. Materials and methods

In order to assess the influence of limestone powder waste concrete modification on the course of carbonation, an experimental plan was prepared. On its basis concrete samples of 10 compositions were prepared, and then after 28 days of curing in water, the effect of carbonation in accelerated conditions was tested. The tests were carried out after the time provided for in the EN 12390-12 standard and additionally within 560 days. A statistical analysis of the results was then carried out.

2.1. Examination of the carbonation depth

An accelerated test for the carbonation resistance of concrete was used in our study using a method based on the draft EN 12390-12 standard. Before placing the samples in the chamber, the depth of carbonisation was checked each time and was 0.0 mm. The samples were placed in the chamber so that air had free access to both unsealed sides of the sample. Gentle air circulation was forced in the chamber. The environmental parameters in the chamber were: CO_2 concentration = 4%, T = (20 ± 2)°C, RH = (55 ± 5)%. The exposure time was 70 days. Measurements were also made at intermediate times: 56 and 63 days. In addition, testing was done after 120, 180 and 560 days of exposure (Fig. 3).
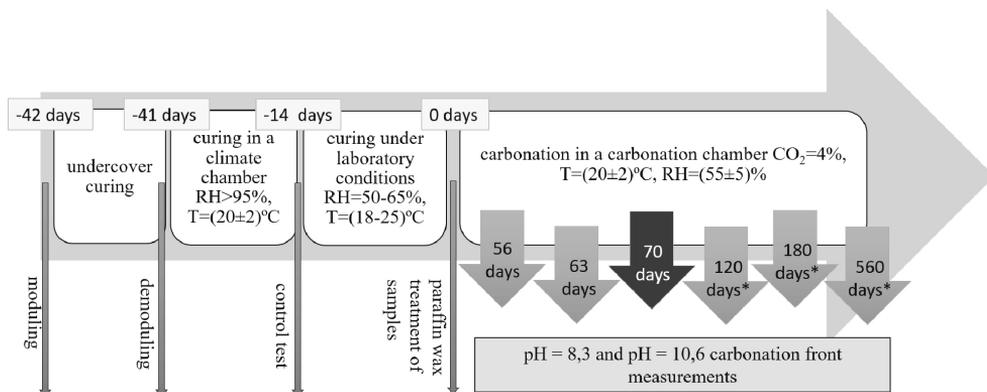
Fig. 3. Timeline diagram of the carbonation study, *additional testing dates not included in the standard

In order to measure the carbonation front, the sample was broken and then, immediately after the breakthrough was exposed, phenolphthalein solution was applied – 1 g phenolphthalein dissolved in 70 g ethyl alcohol and diluted with 30 g distilled water according to EN 14630. The change in colour to violet was checked after 30 s and the depth of the carbonation front was measured to the nearest 1 mm. The test result is the average of 20 measurements – 10 for each exposed edge of the breakthrough. The effect of aggregate grains interfering with the measurement was taken into account by determining the actual line of the carbonation front. In addition, measurements were made with a thymolphthalein solution – pH < 10.6.

2.2. Experimental design

The experimental design included two material variables: the water/cement coefficient (W/C) in the range of $0.35 \div 0.55$ and the level of sand substitution with the waste powder, expressed as a mass ratio of waste to cement in the range of $0 \div 20\%$ (P/C) which means $0 \div 10.8\%$ of natural sand mass. Due to the planned statistical analysis, a bi-factorial polyseckrotal-quasiuniformal plan was adopted with a two-fold repetition of the measurement at the central point (Fig. 4, Table 1) [13].
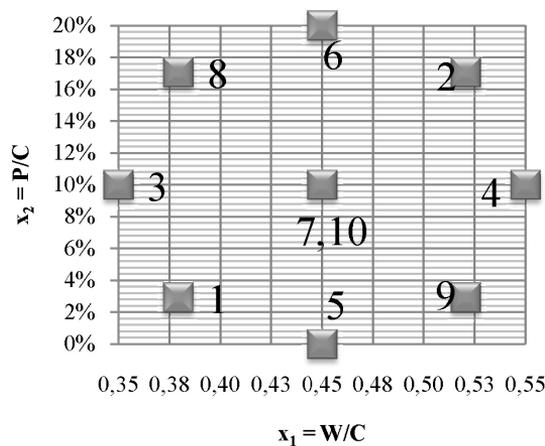
Fig. 4. Experimental plan – the input variables

Table 1. Coded variables and actual experimental design; W/C (x_1) – water to cement ratio, P/C (x_2) – level of fine aggregate substitution with waste limestone powder, C – cement content, W – water content, P – waste limestone powder content, 0/2 – sand content, 2/8 and 8/16 coarse aggregate content

Composition	Coded variables		Actual variables		Concrete mix compositions [kg/m^3]					
No.	x_1	x_2	W/C [kg/kg]	P/C [%]	C	W	P	0/2	2/8	8/16
1	−1	−1	0.38	2.93	375	142	11	711	742	488
2	1	1	0.52	17.07	375	195	64	606	689	453
3	−1.414	0	0.35	10.00	375	131	38	696	753	495
4	1.414	0	0.55	10.00	375	206	38	622	677	446
5	0	−1.414	0.45	0.00	375	169	0	696	715	471
6	0	1.414	0.45	20.00	375	169	75	621	715	471
7	0	0	0.45	10.00	375	169	38	659	715	471
8	−1	1	0.38	17.07	375	142	64	658	742	488
9	1	−1	0.52	2.93	375	195	11	659	689	453
10	0	0	0.45	10.00	375	169	38	659	715	471

The composition of the reference, unmodified concrete mix (composition No. 5 in the experimental design) was adopted in accordance with EN 1766:2001. Therefore, the concrete mixes were prepared with the use of a fixed amount of cement CEM I 42.5 R NA HSR amounting to 375 kg/m^3. The crushed granite aggregate of the group of fractions 2/16 and river sand were used. The grain size curve was selected according to the standard composition by EN 1766:2001 (0/2 – 37%, 2/8 – 38%, 8/16 – 25%). An air-entraining admixture in a constant amount of 0.2% of cement mass was applied, as well as a water reducing admixture in a variable amount dosed to obtain a constant consistency of S3 for all mixes.

3. Results and statistical analysis

To determine the resistance to carbonation, the progress of the front of pH = 8.3 and pH = 10.6 deep into the concrete under exposure to 1% CO_2 was investigated. *A* summary of the test results is presented in Table 2.

Table 2. Overview of carbonation depth values over time of concrete. Bold – values suggested by the standard as a basis for assessment

Exposure time [days]	pH	Carbonation depth [mm]									
		1	2	3	4	5	6	7	8	9	10
56	8.3	0	5	0	4	5	5	6	7	10	10
	10.6	5	9	2	12	20	10	12	12	18	15
63	8.3	0	6	0	5	10	10	10	8	12	11
	10.6	7	11	4	20	25	12	14	10	19	18
70	**8.3**	**3**	**8**	**0**	**8**	**13**	**12**	**10**	**10**	**13**	**12**
	10.6	8	12	10	22	30	16	19	12	20	20
120	8.3	10	10	4	10	18	16	11	12	14	13
	10.6	12	18	15	30	35	23	21	14	25	22
180	8.3	12	12	10	11	20	19	13	14	16	15
	10.6	15	20	18	36	40	28	26	19	30	23
560	8.3	18	17	22	21	25	25	18	20	20	19
	10.6	25	35	30	45	50	33	33	33	35	30

Due to the suggestion of a test with phenolphthalein in the standard and the fact that the results of this test have been analysed in the literature [8, 14–16] an analysis of the results obtained by this test was performed (Fig. 5).

For each of the compositions, a separate analysis was made to fit an equation of carbonatation of the type Eq. (3.1). Such an equation for the course of carbonatation is used in publications by Czarnecki and Woyciechowski, both for unmodified and for concretes modified with mineral additives [8, 14–16]. A list of obtained values together with R^2 coefficients is given in Table 3.

(a)   (b)
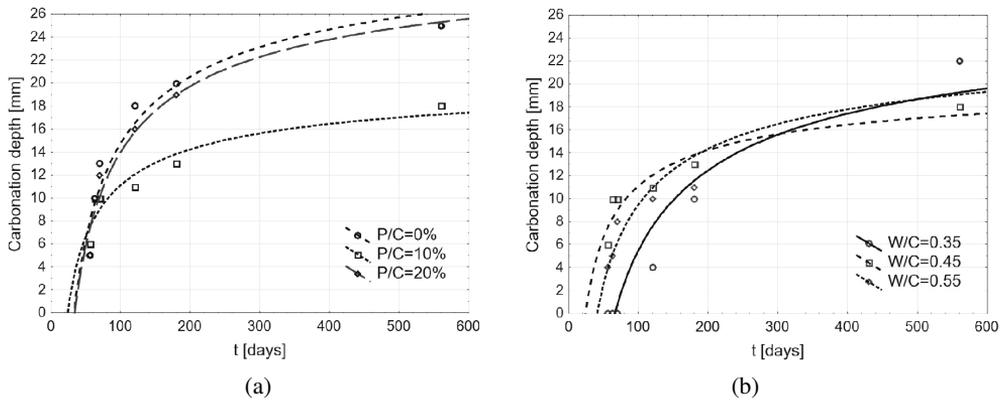
Fig. 5. Dependence of carbonation depth determined with phenolphthalein test as a function of time:
a) W/C constants = 0.45, b) P/C constants = 10%

Table 3. Summary of the values of parameters A and B of the function Eq. (3.1) depending on the
composition according to the experimental plan

Composition number of the experimental plan	A	B	R^2
1	27.341	−206.13	0.9829
2	21.793	−123.90	0.9809
3	29.341	−238.49	0.9212
4	26.099	−166.19	0.9194
5	34.588	−198.44	0.9317
6	33.687	−196.05	0.9580
7	21.791	−106.86	0.8951
8	24.835	−132.85	0.9727
9	23.629	−96.686	0.9556
10	22.233	−90.935	0.9642

Example matches are shown in Fig. 5. The smallest range of carbonatation depth was observed for concretes with W/C = 0.45 and P/C = 10% (composition 7). These were compositions that had a sealed structure and also had very good workability.

$$(3.1) \qquad h = A + B \cdot t^{-0.5}$$

where: h – carbonation depth in mm, A, B – directional coefficients of the equation t – time of exposure in days.

Due to a strong dependence of the carbonation depth on $t^{-0.5}$, an attempt was also made to apply a dynamic model to describe the development of carbonation in time. The depth of carbonation measured with phenolphthalein was analysed. Using the GAM method, the values of A and B coefficients as functions of W/C and P/C were determined as $a(\text{W/C}, \text{P/C})$ and

b(W/C, P/C), respectively. A set of equations describing the development of carbonation in time as a function of W/C and P/C Eq. (3.1) was obtained. For Eq. (3.2), a conformity analysis was carried out and MAPE = 8.14 and R^2 = 0.95 was obtained, which indicates that the prediction can be considered accurate.

$$(3.2) \qquad h = a\left(\frac{W}{C}\frac{P}{C}\right) + b\left(\frac{W}{C}\frac{P}{C}\right) \cdot t^{-0.5}$$

where: h – carbonation depth in mm, $a\left(\frac{W}{C}\frac{P}{C}\right)$, $b\left(\frac{W}{C}\frac{P}{C}\right)$ – functions of W/C and P/C determined in Eq. (3.3) and Eq. (3.4), t – time of exposure in days

$$(3.3) \quad a\left(\frac{W}{C}\frac{P}{C}\right) =$$

$$= \begin{cases} 610 - 2803\frac{W}{C} - 332 \cdot \frac{P}{C} + 3388 \cdot \left(\frac{W}{C}\right)^2 + 189 \cdot \frac{P}{C} \cdot \frac{W}{C} + 1213 \cdot \left(\frac{P}{C}\right)^2 \\ \hspace{9cm} \text{for } \frac{W}{C} < 0.45 \\ 980 - 3863\frac{W}{C} - 193 \cdot \frac{P}{C} + 3916 \cdot \left(\frac{W}{C}\right)^2 - 121 \cdot \frac{P}{C} \cdot \frac{W}{C} + 1213 \cdot \left(\frac{P}{C}\right)^2 \\ \hspace{9cm} \text{for } \frac{W}{C} \geq 0.45 \end{cases}$$

where: W/C – water/cement ratio, P/C – lime powder/cement ratio;

$$(3.4) \quad b\left(\frac{W}{C}\frac{P}{C}\right) =$$

$$= \begin{cases} -6873 - 31179\frac{W}{C} + 5234 \cdot \frac{P}{C} - 36324 \cdot \left(\frac{W}{C}\right)^2 - 7233 \cdot \frac{P}{C} \cdot \frac{W}{C} - 9835 \cdot \left(\frac{P}{C}\right)^2 \\ \hspace{9cm} \text{for } \frac{W}{C} < 0.45 \\ -10029 + 40032\frac{W}{C} + 3293 \cdot \frac{P}{C} - 40413 \cdot \left(\frac{W}{C}\right)^2 - 2920 \cdot \frac{P}{C} \cdot \frac{W}{C} - 9835 \cdot \left(\frac{P}{C}\right)^2 \\ \hspace{9cm} \text{for } \frac{W}{C} \geq 0.45 \end{cases}$$

where: W/C – water/cement ratio, P/C – lime powder/ cement ratio.

The function Eq. (3.2) for a defined composition with constant W/C and P/C tends asymptotically to specific values of the carbonation range. The function reached its minimum for P/C = 10% and up to W/C = 0.45. This was the composition in which the structure was sealed without significant worsening of mix workability.

The conclusions of the analysis of the phenolphthalein and thymolphthalein tests are different. This may indicate a variable dependence of the extent of carbonation in time or a variable nature of the dependence as measured by different indicators.
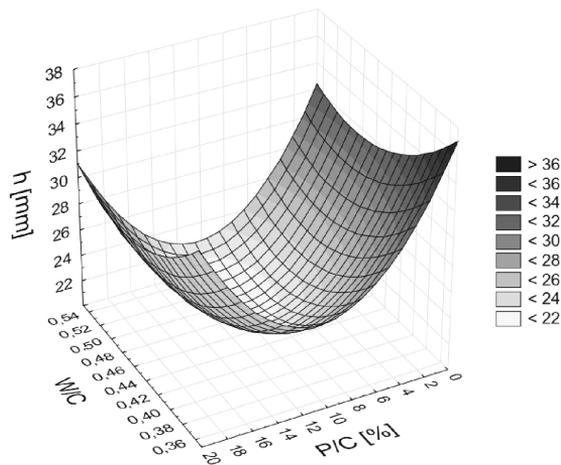
Fig. 6. Range of maximum carbonation – *h* measured with phenolphthalein as a function of W/C and P/C

4. Conclusions

The process of carbonation of concrete is very important, especially when considering the durability of reinforced concrete structures. Modification with waste material must not lead to a reduction in durability. Thanks to the preparation of an appropriate experimental plan, conducting long-term tests – beyond the time limits suggested by the standard, it is possible to prepare a model of the dynamic development of the extent of carbonation in time. This makes it possible to assess the modified concrete within the ranges of long-term durability. In the case of the analysed waste – limestone dust, modified concretes not only turn out not to be worse, but at $P/C = 10\%$ they reach the lowest ranges of carbonation, which will affect better passivation protection of steel in reinforced concrete structures. The results obtained are very promising and further research should include the use of mineralogically different waste dusts from aggregate dusting for the production of HMA mixtures.

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Wpływ substytucji piasku odpadowym pyłem wapiennym na przebieg karbonatyzacji

Streszczenie:

Koncepcja zrównoważonego rozwoju wymaga, aby materiały modyfikowane odpadami również wykazywały odpowiednią trwałość. W niniejszej pracy zbadano wpływ modyfikacji betonu cementowego odpadowym pyłem wapiennym na przebieg karbonatyzacji betonu. Analizowany pył odpadowy pochodzi z odpylania kruszywa przeznaczonego do stosowania w mieszankach MMA – Mieszanek Mineralno-Asfaltowych. Celem pracy było zbadanie, czy częściowe zastąpienie piasku pyłem odpadowym będzie miało negatywny wpływ na potencjalną trwałość elementu żelbetowego wykonanego z tego betonu. W celu określenia stopnia karbonatyzacji przygotowano plan eksperymentu obejmujący wykonanie betonów o różnym stopniu substytucji i zmiennym stosunku wodno-cementowym. W celu określenia wpływu długiego czasu ekspozycji badanie przeprowadzono zgodnie z normą PN-EN 12390-12, ale z wydłużonym czasem badania do 560 dni. Uzyskane wyniki poddano analizie statystycznej i określono przewidywany maksymalny stopień karbonatyzacji w zależności od poziomu substytucji i stosunku wody do cementu. Przeprowadzona analiza wskazuje, że możliwe jest zastąpienie piasku odpadowym pyłem wapiennym bez negatywnego wpływu na stopień karbonatyzacji, a tym samym na trwałość konstrukcji żelbetowej.