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PHYSICAL-MECHANICAL AND DURABILITY PROPERTIES OF CONCRETE WITH LIGHTWEIGHT ARTIFICIAL AGGREGATE PRODUCED BY CONTROLLED SELF-BURNING OF WASTE COAL TAILINGS

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Abstract

The goal of this research focusing on the burning of coal tailings to produce a lightweight artificial aggregate is to compare the physical-mechanical and durability properties of concrete containing this new type of artificial aggregate with those of the reference concrete. The following parameters are measured on all the samples: compressive strength, volume mass, the static modulus of elasticity in compression, the frost resistance of this new type of concrete, and the resistance to CO_2. All the concretes tested with the new artificial aggregate exceeded the characteristic strength values in all the strength classes monitored. The volume mass of the concrete with the new artificial aggregate is tens of percentages lower than that of the conventional reference concrete. The frost resistance of the concrete tested was reliably proven by the non-destructive resonance method. It was also found that at the lower strength classes of concrete, carbonation occurred earlier, but there was no obvious direct relation to the porosity of the new artificial aggregate. The tests performed showed that it is possible to produce high - quality concrete using a new lightweight artificial aggregate produced from coal tailings

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Key words

- Coal piles;
- Self-burning;
- Lightweight artificial aggregate;
- Lightweight concrete;
- Physical-mechanical characteristics;
- Durability characteristics coal piles;
- self-burning;
- Lightweight artificial aggregate;
- Lightweight concrete;
- Physical-mechanical characteristics;
- Durability characteristics.

1 INTRODUCTION

Global coal production is still relatively high today. Coal is the second most used raw energy material, and the total world production is estimated at 5.3 billion tons per year (http://www.okd.cz/). Based on the resolution of the COP26 climate conference in Glasgow, "End of Coal in Sight at COP26" (https://ukcop26.org), a gradual reduction in the extraction of this raw material can be expected globally in the next decade. However, coal mining in the Czech Republic increased in 2022, despite the planned future slowdown until 2033. Approximately 35.2 million tons of black and brown coal were mined, which is a 10% year-on-year increase (http://www.okd.cz/). The reason for the increase was last year's energy crisis caused by the war in Ukraine, as well as the revival of industry after the Covid-19 pandemic. One of the major environmental problems of using this energy resource is the high content of sulfur and carbon, which produces a whole range of environmentally harmful oxides (CO, CO_2, SO_2, etc.) in the combustion process. Another considerable burden on the environment is linked directly to the mining of coal, during which the treatment process of the raw material produces a large amount of coal waste known as tailings. An important fact is that 1 ton of coal generates approximately 0.7 tons of waste, which is currently unused. Coal tailings normally contain 5-40% of combustible substances, a number of unstable clay minerals, and also large quantities of highly soluble metals (Xu, 2021). All these properties significantly reduce the possibility of their subsequent use (Ribeiro et al., 2010; Santos et al., 2023).

The global trend of "treating" the coal tailings is to store them in piles, which are subsequently recultivated, which means that the piles of industrial waste occupying vast areas are covered with soil and vegetation. A problem occurs in a situation where the high content of combustible substances combines with access to air, moisture, and other factors, which then causes the pile to steam up; this process can later lead to its spontaneous combustion. Inside the pile, a burning center can arise where temperatures can reach up to 1000°C and which can spread very slowly throughout the pile (Ribeiro et al., 2010). In some cases, however, gradual self-extinction occurs. Fighting pile fires is very expensive and difficult, and during the combustion, various harmful emissions are produced (Ribeiro et al., 2012). Another problem is associated with the release of harmful substances into the soil, which often results in groundwater contamination (see Fig. 1). These issues are largely dealt with in China and Portugal, where the possibility of reducing the amount of emissions produced has been discussed (Querol et al., 2011; Santos et al., 2023).

One of the options for the treatment of this industrial waste is so-called “self-burning”. This process involves placing the pre-treated coal tailings on a sinter grate and igniting them; the actual burning process is then initiated while air is sucked through to the grate. In this process, a new type of artificial aggregate is produced, which is then sorted and divided into selected fractions, which can subsequently be used in many applications, especially in the building industry. The temperatures from the burning process are around 1300°C, and the process results in the production of new, thermally stable minerals. The content of combustible substances in the burnt coal tailings is below 2%.

Fig. 1 *Pile of coal tailings; the source of production of new lightweight artificial aggregate, but also the source of harmful substances, which are released into the soil*

The goal of this research was to demonstrate the possibility of using a new type of artificial aggregate from sintered coal wastes as an aggregate to be used in concrete production and to examine what properties can be expected from this product in cases of its practical application. Another objective was to compare not only the mechanical but also the physical-chemical properties of concrete with the burnt coal tailings aggregate against the reference concrete with a natural aggregate. Furthermore, it was examined whether or not the new type of concrete degrades rapidly when exposed to CO_2.

According to the available resources, this topic has not been dealt with intensively. Coal tailings are mentioned in professional journals and articles only in terms of "piles" and their impact or on reducing their negative influence on the environment. For example, Querol et al. (2011) points out how to reduce emissions resulting from the piles that are at the stage of spontaneous combustion. Their proposed solution was to cover the pile of coal waste with a layer of loose clay and then to compact this layer. This proposal addresses only the local reduction of the emissions produced, which sooner or later will have to be released into the air. However, in the databases of scientific journals, a number of links are available concerning the development and testing of similar types of lightweight artificial aggregate, but these are produced based on different principles and prepared from different waste materials such as fly ash, slag, coal slurries, etc. For example, ceramsite (Babenko et al., 2022; Deng et al., 2023) and an artificial aggregate from reservoir sediments (Chen et al., 2012; Xing et al., 2012) are produced in rotary kilns where sintering heat is supplied from another source, which makes it significantly more expensive.

A range of possibilities of processing different types of waste into the form of a lightweight artificial aggregate have been examined round the world. For example, Ducman and Mirtič (2009) deal with the possibility of producing an artificial aggregate from silica sludge and superfluous clay. In some cases, the thermal conditions of the artificial aggregate manufacturing process were studied, including their effect on its resulting physical properties (Liao and Huang, 2011). A thermal process that is very similar to the one used in the production of artificial aggregate from burnt coal tailings is applied in the production of an artificial aggregates from sintered fly ash. The problems from this production, especially considering the final physical properties of the lightweight aggregate from sintered fly ash, have been discussed in various research articles (Anagnostopoulos and Stivanakis, 2009; Cheeseman et al., 2005; Kockal and Ozturan, 2011; Chang et al., 2007; Laursen et al., 2006; Mun, 2007; Wang et al., 2009).

A newly proposed solution to the processing of coal tailings during the manufacturing of an artificial aggregate on an agglomeration grate can significantly eliminate harmful emissions while using a large amount of energy. At the same time, we can get a new type of artificial aggregate with a relatively low bulk density and good thermal stability. It is important to note that coal tailings, due to the high amount of ash, do not burn under optimal conditions, like coal does. They produce much higher emissions, which need to be disposed of when producing this new type of artificial aggregate.

The issue of sintered aggregate production from coal tailings is especially beneficial in terms of environmental protection. It will not occupy a large amount of land; moreover, there will be no unregulated burning of the material stored in the piles, no escape of harmful substances into the soil, and finally, no contamination of groundwater. The processing of this industrial waste thus offers the possibility to produce a new type of artificial aggregate in the form of a secondary raw material that can be effectively used in the construction industry.

2 MATERIALS AND METHODS

As mentioned above, the basic raw material in the production process of this new type of artificial aggregate is coal tailings. This industrial waste is produced in large quantities during the extraction of coal. At that stage, it is a "side product" containing the residues of minerals and sedimentary rocks, e.g. sandstone, siltstone, claystone or clayey shale. When exposed to weathering, a number of unstable minerals will lose their strength and gradually create a substrate suitable for live cultures. Given that the content of combustible substances is in a range of 0-40%, the sulfur and carbon content is also higher (http://fast10.vsb.cz/). The absorption capacity of the coal waste is around 1-4%. The volume mass at this stage is not much different from a commonly mined aggregate, i.e., around 2400 kg/m^3; the bulk weight depends on the grain size and ranges from 900 to 1200 kg/m^3.

The intended method of handling this type of industrial waste is based on the principle of self-burning of the raw material on a sintering grid. The main advantage of this process is the fact that no other source of energy is needed in the production; the combustion is based on the actual content of combustible substances and the primary ignition is by a gas burner. On the other hand, the disadvantage is, as already mentioned, a higher amount of emissions generated, especially in the first stage of production when the ignition of the raw material starts. These emissions can be eliminated, but at a relatively high cost. The heating process can be seen in Figs. 2 and 4. In the production process of this new type of artificial aggregate, relatively high temperatures are reached; the structure is broken, and a highly open porosity of the resulting material is formed. Moreover, the bulk and normal weight is decreased, and the absorption is increased. The bulk weight of the aggregate from burnt coal tailings is around 1800 kg/m^3; the water absorption is up to 13%. The crushing resistance of the aggregate of 8/16 mm ranges from 4.8 to 5.6 MPa.

Other methodological procedures are mainly related to the testing of the mechanical properties, the frost resistance of the concrete, and the methodological procedure of accelerating the carbonation of the concrete.

The concrete formulas of the characteristic strengths given were supplied by a concrete company; based on an agreement, it is not possible to present them. It can only be stated that they are designed in such a way that the properties of the concrete significantly exceed their characteristic strengths.

Fig. 2 *Ignition of raw material in a furnace*

Fig. 3 *Horizontal mixing of a concrete batch*

Fig. 4 *Burning process of coal tailings*

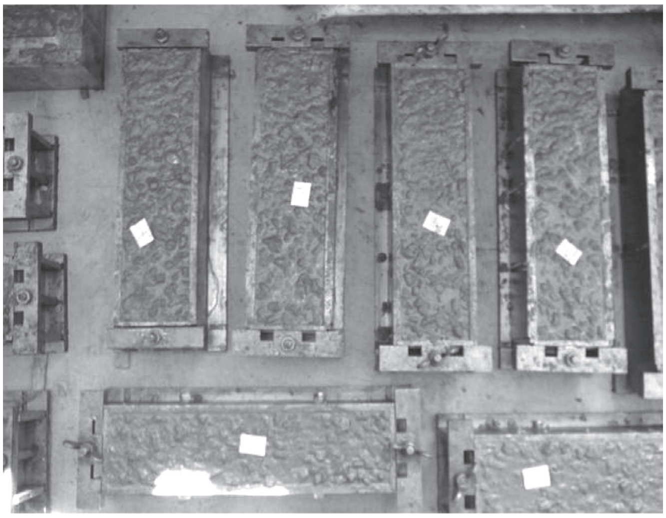

Fig. 5 *Concrete mixture in molds with dimensions of 100 x 100 x 400 mm*

Based on these formulas, the compositions of the concrete mixtures were calculated for the artificial lightweight aggregate in such a way that in the final stage, a comparison would be possible (based on the physical, mechanical, and other properties) between the artificial aggregate concrete and the concrete currently produced (with a natural aggregate). The concrete mixing process and the placement into molds can be seen in Figs. 3 and 5. A total of 60 standard cubes of 150 x 150 x 150 mm and 36 standard prisms of 100 x 100 x 400 mm (see Fig. 5) were produced to test the physical-mechanical and durability properties of the concrete with a lightweight artificial aggregate and the reference concrete with a natural dense aggregate. The properties of the concrete monitored were determined by averaging the results obtained by testing at least three corresponding test samples. Fig. 6 shows the structure of the concrete with burnt coal tailings as an aggregate, and Fig. 7 shows the concrete with a natural aggregate.

One of the most important parameters during the mixing of concrete is consistency, which is determined according to ČSN EN 206-1 Concrete - Part 1: Specification, performance, production and conformity, and ČSN EN 12350-2 Testing fresh concrete – Part 2: Slump-test. The compressive strength was determined using the standardized procedure according to EN 12390-3 "Testing hardened concrete – Part 3: Compressive strength of test specimens"; the determination of the static modulus of the elasticity of the concrete was done according to the valid ČSN ISO 6784 (731319) "Concrete - Determination of static modulus of elasticity in compression".

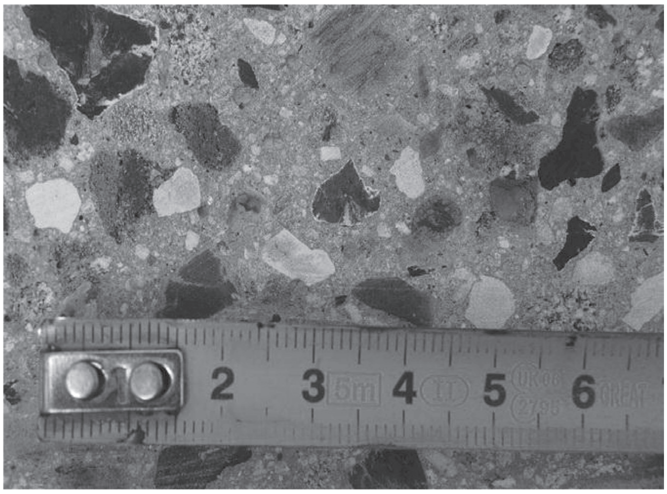

Fig. 6 *Saw cut of concrete with burnt coal tailings tested*

The determination of the frost resistance was based on ČSN 731380 "Testing resistance of concrete to freezing and thawing - Internal structure damage", which specifies how to proceed. The standard refers to the three available options for determining frost resistance. In our case, we chose the evaluation of damage to the internal structure of concrete using the resonance method, which determines the dynamic modulus of elasticity in tension and compression. The standard also prescribes the assessment of damage resulting from 56 freeze-thaw cycles. The approach we selected was to monitor the individual cases of damage over the following freeze cycles: 25, 50, 75, and 100. This allows for an easy comparison of any structural damage during testing.

The resistance to freezing and thawing of the concrete selected is expressed as a percentage change in the dynamic modulus of elasticity in tension and compression following the above-mentioned number of freeze cycles compared to the reference samples, which were not exposed to this process. In cases where there was a reduction compared to the reference sample, the values in the individual bar charts are expressed as a negative value (negatively orientated bar charts – see Fig. 14) and vice versa. The frost resistance of the samples tested could also be expressed by the determined values of the relative dynamic moduli (RDM) in percentages of the measured values of the resonance frequencies, but due to the number of samples, the processing of the changes in percentages of the dynamic moduli of the samples onto the bar charts is clearer.

To determine the depth of the concrete carbonation, we chose a method that was proposed as part of our research work at the Faculty of Civil Engineering, Brno University of Technology, the Czech Republic.

Tab. 1 *Water-cement ratio for each concrete mixture with the artificial aggregate*

Class of concrete with artificial aggregate	Water-cement ratio
C35/45	0.44
C30/37	0.42
C25/30	0.46
C20/25	0.58
C12/15	0.91
C8/10	1.14

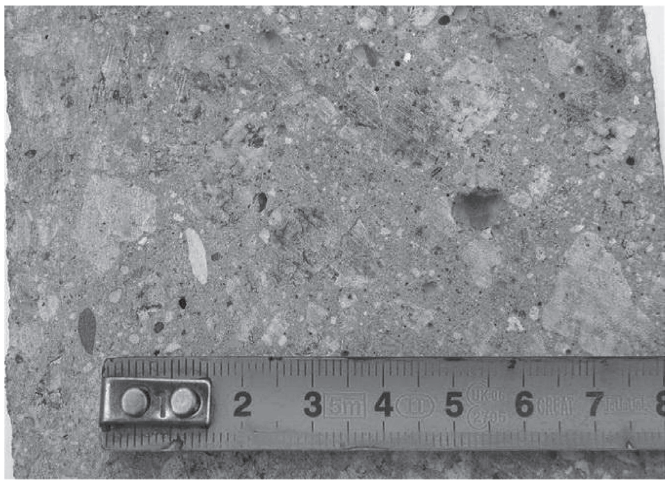

Fig. 7 *Saw cut of the reference concrete with the natural aggregate*

This is an accelerated test in which concentrated 98% CO_2 is used as the gas that causes carbonation (Stehlík, 2011). Based on previously performed correlations (Stehlík and Novák, 2011), this test can be used to estimate the time period during which the given thickness of the type of concrete tested will be carbonated.

3 RESULTS

The concrete formulas were designed in such a way that it could be possible to adequately compare the individual properties of the concrete mixtures and concrete from burnt coal tailings and a natural aggregate. It is important to note that the replacement of the aggregate was carried out only with

4 to 8 mm and 8 to 16 mm fractions. The fine fractions from 1 to 4, which were mined sand from Bratčice (Moravian region, Brno County), were identical in both cases.

During the concreting, important differences occurred; this section therefore receives more attention. A very important parameter is the amount of mixing water, which significantly affects both the consistency of a concrete mixture and the resulting properties of the concrete. Water was therefore added to the mixes in such a quantity that the resulting consistency according to ČSN EN 206-1 Concrete - Part 1 was in the range of a cone settlement of 80 mm to 110 mm.

Tab. 2 *Water-cement ratio for each concrete mixture with the natural aggregate*

Class of concrete with the natural aggregate	Water-cement ratio
C35/45	0.40
C30/37	0.40
C25/30	0.44
C20/25	0.49
C12/15	0.87
C8/10	1.12

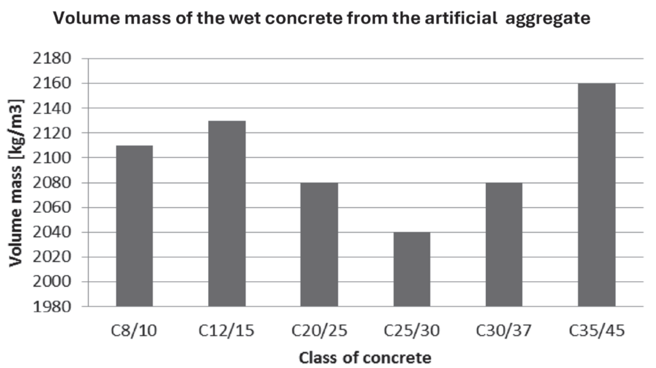

Fig. 8 *Volume mass of the wet concrete mixtures from the artificial porous aggregate (AA) tested on six triplets of concrete cubes with dimensions of 150 x 150 x 150 mm*

It is also important to note that the artificial aggregate has a high degree of porosity and that it was soaked before the concreting. Six percent of the estimated amount of water was used for this process; the artificial aggregate was then mixed for 5 minutes in a concrete mixer.

With regard to the physical properties of the artificial aggregate, the grains of which have very sharp edges after crushing and a higher degree of porosity, the mixture had worse rheological properties. After mixing the aggregate with water for a specified period of time, other ingredients, including a plasticizer, air-entraining admixtures, and other additives, were added into the mixing device. Mixing water was later added to the mixture in order to achieve the desired consistency. Based on the amount of water added in each mix, the values of the water-cement ratio were calculated. They are presented in Tabs. 1 and 2.

From the results it is evident that the water-cement ratio for both types of concrete with the natural and the artificial aggregate is nearly identical. This is due to the fact that the total value of the water-cement ratio does not include the water that is needed for pre-soaking. In the higher strength classes, the water coefficient varied between 0.4 to 0.46; this value in class C20/25 was approximately 0.55. Significantly higher values were obtained for the C12/15 and C8/10 classes. This is because a higher amount of fly ash was added to the mix as a replacement for cement.

Another important parameter is the volume mass of the raw concrete. The highest one was recorded in the case of class C20/25 (with the natural aggregate), namely 2390kg/m^3. In this class the largest difference was recorded in comparison with the volume mass of the concrete with the artificial aggregate. This is due to the fact that in this case, the largest quantity of the 4-16 mm aggregate fraction was used. These findings can be seen in Figs. 8 and 9.

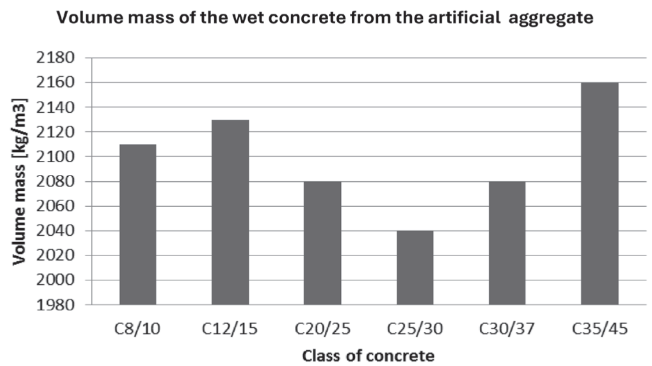

Fig. 9 *Volume mass of the concrete mixtures from the natural aggregate (NA) tested on six triplets of concrete cubes with dimensions of 150 x 150 x 150 mm*

The volume mass of the concrete with the new type of artificial aggregate corresponds to that of the lightweight and plain concrete (2000 kg/m^3). For the concrete with the natural aggregate, the volume mass values are up to 15% higher (see Fig. 10).

The tests performed were focused on curing the concrete, where, inter alia, the increase in compressive strength was monitored after 7, 28 and 90 days of curing. The gradual increase in strength during the curing process of the concrete with the artificial aggregate is represented in Fig. 11. In the lower strength classes of concrete, nearly 40% of the 90-day strength was achieved after 7 days; in the higher quality concrete, this value was actually 70%. When comparing the strengths after 28 and 90 days of curing, the differences were not so significant. In the lower strength classes of the concrete, 75% of the 90-day strength was achieved, while in the higher strength classes, it was 93%.

The situation with the natural aggregate concrete (Fig. 13) is similar. It is important to compare the two types of concrete in terms of the rate of increase in the compressive strength over time. In the lower strength classes, the increase was faster in the concrete with the natural aggregate than in the concrete with the artificial aggregate; in the better quality concretes, it was the other way round.

In the concrete with the artificial aggregate made from burnt coal tailings containing a large amount of open pores, it is necessary to count on a higher amount of water during concreting. In the higher strength concrete classes, a higher amount of cement is used; the water trapped in the pores of this aggregate produces a "self-curing" effect, where it is slowly released during the hydration process. The increase in strength is therefore slower than in the case of the concrete with the natural aggregate. This situation can usually be observed in all concrete, whether from a natural or artificial lightweight aggregate.

The modulus of elasticity in compression is a characteristic that is used mainly in static calculations of concrete structures. It describes the extent to which a structure is able to withstand mechanical stress without the occurrence of permanent deformation.

Looking at Fig. 12, it is apparent that concrete with an aggregate made from burnt coal tailings is far from achieving similar results as concrete with a natural aggregate. This is primarily due to the fact that in the case of exhaustion of the cement matrix´s mechanical strength, the porous aggregate is not able to transfer high stress without the occurrence of permanent deformation. This fact, however, is known about all types of concretes containing lightweight porous aggregate.

A comparison of the frost resistance of the individual types of concrete was performed, as mentioned above, using the non-destructive method described in ČSN 731380. The frost resistance was monitored on three selected concrete classes, i.e., C25/30, C30/37 and C35/45. The evaluation of frost resistance was based on a comparison of the changes in the dynamic modulus of elasticity in tension and compression before and after exposing the specimens to cyclic freezing (25, 50 and 100 F-T cycles). One F-T cycle consists of 4 hours of freezing in air, where the air temperature ranges from -15°C to -20°C, and two hours of thawing in water at a temperature of +20°C. One F-T cycle therefore lasts 6 hours; cooling to the desired temperature takes place continuously for 4.5 hours.

On the basis of a comparison of the values obtained of the percentual changes in the dynamic modulus of elasticity, it can be stated that all the concrete tested met the requirements after 100 F-T cycles. In almost all the cases, there was even a gradual increase in the dynamic modulus of elasticity, which amounted to 1-7% after 100 F-T cycles. In this respect, it is possible to say that no disruption of the structure of the concrete samples occurred due to cyclic freezing and thawing (see Fig. 14). However, in Fig. 14, we can observe a worsening of frost resistance expressed by a lower percentage increase in the dynamically determined modulus of elasticity for the concrete with the porous AA aggregate in contrast to the increase in their characteristic strengths (C25/30 – C30/37 – C35/45). This anomaly can be attributed to the porous nature of the artificial aggregate, which interacts better with a lower-quality

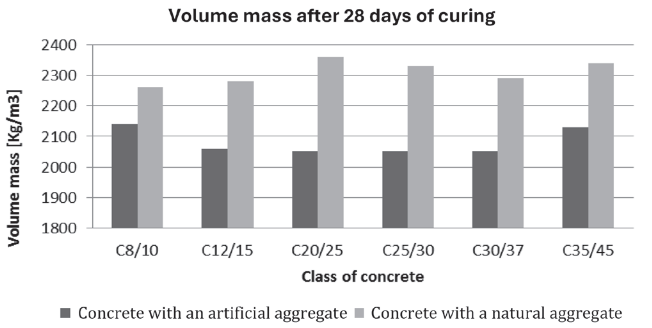

Fig. 10 *Volume mass of concrete after 28 days of curing, concrete with AA, and concrete with NA, tested on individual cubes with dimensions of 150 x 150 x 150 mm*

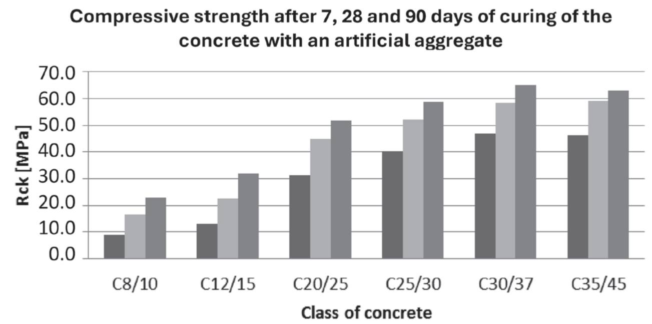

Fig. 11 *Compressive strength of concrete with AA after 7, 28 and 90 days of curing, tested on individual cubes with dimensions of 150 x 150 x 150 mm*

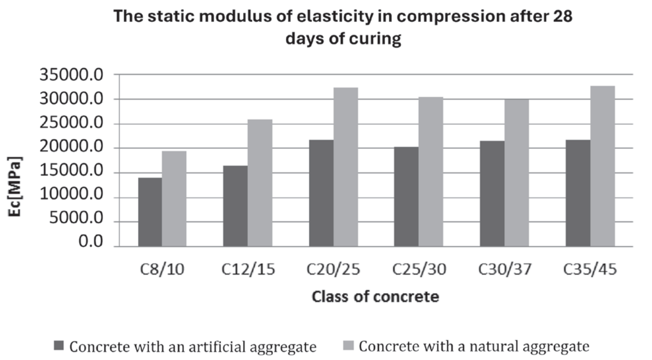

Fig. 12 *Static modulus of elasticity – concrete with AA and NA after 28 days of curing, tested on two sets of six triplets of concrete prisms with dimensions of 100 x 100 x 400 mm*

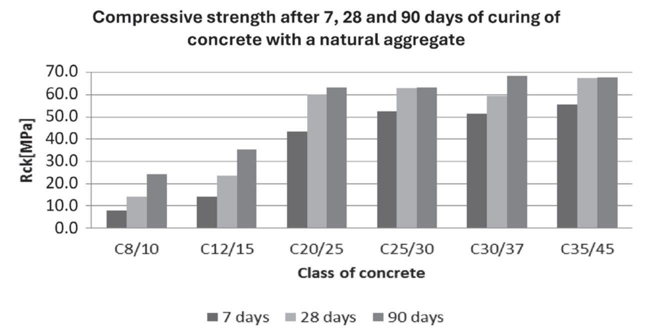

Fig. 13 *Compressive strength of concrete with NA after 7, 28 and 90 days of curing; individual cubes with dimensions of 150 x 150 x 150 mm were tested*

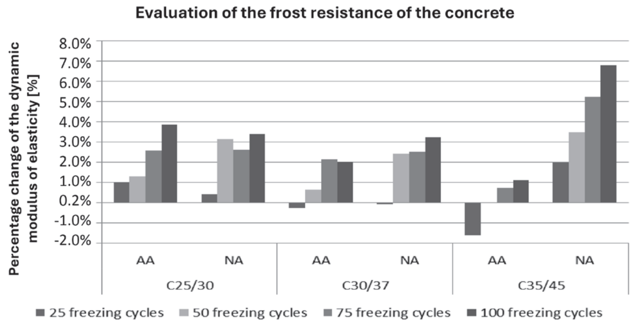

Fig. 14 *Percentage change of the dynamic modulus of elasticity, depending on the number of freezing cycles. It was tested on 3 AA + 3 NA prisms with dimensions of 100 x 100 x 400 mm for the given strength class and four freezing cycles*

cement paste in the formation of suitable reservoirs to compensate for changes in the state of the capillary water.

In the case of evaluating the frost resistance of the concrete by comparing the relative dynamic modulus of elasticity (RDM) values, the lowest drop in the dynamic modulus of elasticity of approximately 1.7% for the AA C35/45 formula (see Fig. 14) corresponds to the approximate RDM of 93%, while concrete is considered frost-resistant with a value of RDM ≥70%.

A comparison of the degree of degradation by CO_2 was done using a method that significantly accelerates the carbonation of the concrete layer. In the samples exposed to an environment of 98% CO_2 for 126 days, the depth of the carbonated layer was monitored (loss of purple color of the phenolphthalein indicator in a 0.1% aqueous solution), which gradually grew to the center of the samples. The goal was to evaluate the degree of carbonation of the samples, depending on the individual strength classes and types of aggregate.

From what has been said above, it is apparent that concrete of the lower classes selected is influenced relatively quickly by its exposure to CO_2, which decreases its pH and therefore decreases the ability to protect the reinforcement from corrosion. In this case the natural aggregate concrete showed a slightly lower resistance (see Figs. 15 and 17).

It can be concluded that the most important parameter of the carbonation process by far is the quality of the cement paste, where in the concrete of the higher strength classes, the degradation of the layer occurs after a much longer time. Likewise, as in our case, there was no disruption in both types of concrete in the layer due to CO_2 within the period monitored. The intact layer of concrete class C25/30 can be seen in Figs. 16 and 18. In Fig. 19, we can see the carbonation process of the concrete (class C20/25) with the artificial and natural aggregate over the course of time. It is apparent from the graphs in Fig. 19, that the effect of the type of aggregate on the carbonation process is negligible; the Achilles heel is mostly the cement paste.

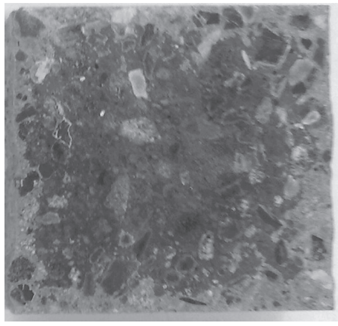

Fig. 15 *Carbonation progress of AA C20/25 concrete after 86 days of exposure to 98% CO_2; the healthy part is dark grey, while the average carbonation depth is 7 mm*

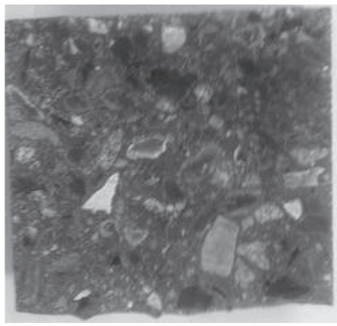

Fig. 16 *Carbonation progress of AA C25/30 concrete after 86 days of exposure to 98% CO_2; the healthy part is dark grey; the carbonation is negligible*

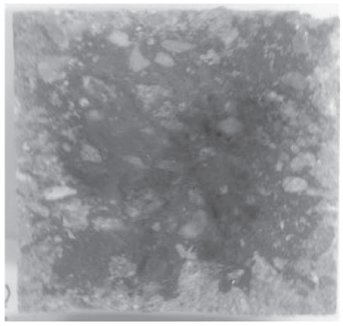

Fig. 17 *Carbonation progress of NA C20/25 concrete after 86 days of exposure to 98% CO_2; the healthy part is dark grey; the average carbonation depth is 15 mm*

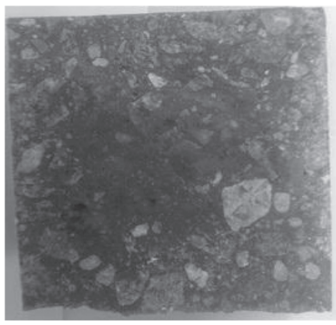

Fig. 18 *Carbonation progress of NA C25/30 concrete after 86 days of exposure to 98% CO_2; the healthy part is dark grey, while the carbonation is negligible.*

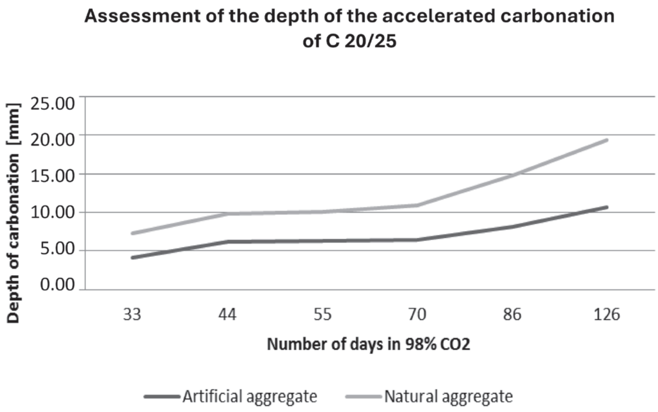

Fig. 19 *Increase in the carbonated depth of the AA C20/25 and NA C20/25 concrete*

4 CONCLUSIONS

One major benefit of this research work is the possible use of industrial waste, which is stored in large volumes in piles where many dangerous situations can occur. Processing coal tailings by producing an artificial aggregate can result in a quality product that can be used for various applications in the construction industry. The results suggest that this type of artificial aggregate is suitable for producing high quality concrete that has a good resistance against the effects of cyclic freezing. It has a lower density, which is an advantage in many applications, especially for progressive lightweight constructions. The moduli of elasticity of concrete with this new type of artificial aggregate from burnt coal tailings are usually up to 26 GPa.

Further research should mainly be focused on the possibility of using this aggregate in refractory structures. Aggregates from burnt coal tailings are produced at temperatures of around 1300°C. Many minerals in the original material are converted into stable forms, which gives a new perspective on the use of these aggregate. The production of these aggregates is not as expensive as the production of artificial refractory aggregates and heat-resistant additives. When producing an artificial refractory aggregate, much more energy must be added to the process, in comparison with the burnt coal tailing aggregate.

Acknowledgements

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