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# Concrete with Accelerated Naturally Carbonated Recycled Concrete Aggregates

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**Abstract**

*Recycled concrete tends to be increasingly investigated with respect to the environmental needs, such as resource depletion, circularity and lack of landfill disposal spaces. A naturally accelerated carbonation procedure by using atmospheric $CO_2$ concentration level and a cyclic wetting and drying of the recycled concrete aggregates was applied. A two stages carbonation process was applied. The recycled concretes exhibited similar mechanical properties of the reference concrete or even better durability as compared to the reference concrete prepared with natural rock aggregates. The carbonation of the outer layer of the recycled concrete aggregates was achieved with a simple method that can be widely applied and is sufficient to significantly increase the properties of the recycled concrete aggregates.*

**Keywords:** Concrete, Recycling, Aggregates, Acceleration, Carbonation.

## Introduction

Recycled concrete is a component of the sustainability of cementitious materials that may contribute to a lower environmental impact and save precious landfill disposal space and pollutants leaching into the ground. The quality of the recycled concrete depends on the properties of the recycled aggregates. Actually, the demolition process represents the end of a service life of a structure. In some countries the material is still landfilled, and a marginal verification of its pollutant content is done. Conversely, when the lack of disposal space or the costs are high and the recycling capability, such as mobile crushing and aggregate treatment plants are present, the recycling process tends to occur. The demolition of high quality infrastructure concrete, such as from highway bridges or tunnels may provide high grade recycled aggregates. An adequate dosage of these latter aggregates below 50% by concrete mass still allows to reproduce durable concrete (Antonietti et al., 2024). The recycling plants collect all type of concrete that derives from different demolition sites and no controls of the concrete source can be done in detail. However, a simple and fast differentiation between infrastructure concrete and all the rest of the concrete may help to distinguish between potentially high and low quality aggregates. This separation allows a more targeted exploitation of the recycled aggregates towards durable infrastructure recycled concretes and conventional recycled concretes to be used for buildings, where no particular high environmental durability requirements are necessary.

In the case no minimal distinction about the quality of the source concrete brought to the recycling plants can be done, the properties of the aggregates must be tested. The water adsorption of the recycled concrete aggregates is a main parameter that requires a special attention for recycled mixtures, although with adequate and sustainable mix designs a satisfactory mechanical and durability performance can be accomplished (Paglia et al., 2022). The water adsorption of the aggregates can reach relatively high values beyond 9 % (Xiao et al., 2005). The fine component may reach even higher values up to 13%, due to the high specific surface area of the material, which promotes a higher water adsorption (Evangelista & Brito, 2007). Mixed granulates containing ceramic waste is also a main inert waste found during the demolition (Yan et al., 2017). Nonetheless, the ceramic component, especially the coarse aggregates are porous and adversely affect the mechanical and the durability properties of the recycled concrete. Therefore, the use of mixed granulate recycled concrete is not a long-term sustainable option (Paglia, 2022). The sorting of the ceramic component and even better, the rock components, may largely promote a more reliable and homogenous material with a higher potential for recycling.

Many different techniques are applied to increase the properties of recycled concrete aggregates (Tam et al., 2021), but only a few are reasonable in practical applications. Artificial accelerated carbonation by using a high concentration of $CO_2$ is one of the major methods to improve the quality of recycled concrete aggregates (Liang et al., 2020). This method is mainly used as a $CO_2$ sink and starts to be exploited along with the $CO_2$ production of cement production plants to reduce the $CO_2$ emissions. The accelerated carbonation process may almost completely close the porosity of the old cementitious paste around the natural aggregates (Zhang et al., 2015). Space availability to treat the recycled aggregates and the time are main concerns. Nonetheless, the recycled material does not need always to be used immediately. In fact, debris may require to be transported possibly along short distances to stationary plants to be prepared. This cannot always be done within highly populated urban areas, and the location of the future use may not be necessarily near to the demolition site. In this case, a slightly longer time may pass between the demolition and recycling stage.

The main goal of the project is to test a simple and natural, but accelerated carbonation method that use the atmospheric $CO_2$ concentration and promotes the carbonation process through a periodical wetting and drying of the recycled concrete aggregates. Furthermore, the differently exposed recycled aggregates to the naturally accelerated carbonation are used to prepare recycled concretes. The mechanical and the durability performance are tested.

## Experimental Procedure

A compressive strength class C 30 / 37 was produced with a mix of siliceous and calcareous natural aggregates. An ordinary Portland cement CEM I 42.5 N with a dosage of 320 Kg/m3 and the addition of 6% fly ash by weight of cement were used. The effective water / cement ratio was 0.47 by considering the natural water adsorption of the aggregates of 0.6%. The water reducing admixture dosage was 0.9 % by cement mass and the density of the fresh natural aggregate concrete mixture was 2468 Kg/$m^3$. The concrete block produced was demolished on site after approximately 3 months (Fig. 1 left) and the debris were crushed (Fig. 1 right) to prepare the recycled concrete aggregates.

**Figure 1:** Demolition and crushing procedure of the concrete produced with natural aggregates.

The crushed concrete aggregates with a diameter range 0-32 mm were exposed on a first stage (stage 1) at atmospheric $CO_2$ exposition for 8 months, where for the coarser aggregates after mechanical rupture exhibited an initial carbonation depth down to 3-4 mm (Fig. 2 top). On a second stage (stage 2) the material was exposed to the atmosphere with 450 ppm $CO_2$ for an additional 8 months. The recycled concrete aggregates were slightly widespread on a laboratory table and cyclically sprayed with water two times a week and naturally dried followed with a periodical mixing of the aggregates, in order to increase their contact with the atmosphere. After this latter stage, the carbonation increased up to 1 cm (Fig. 2 bottom).

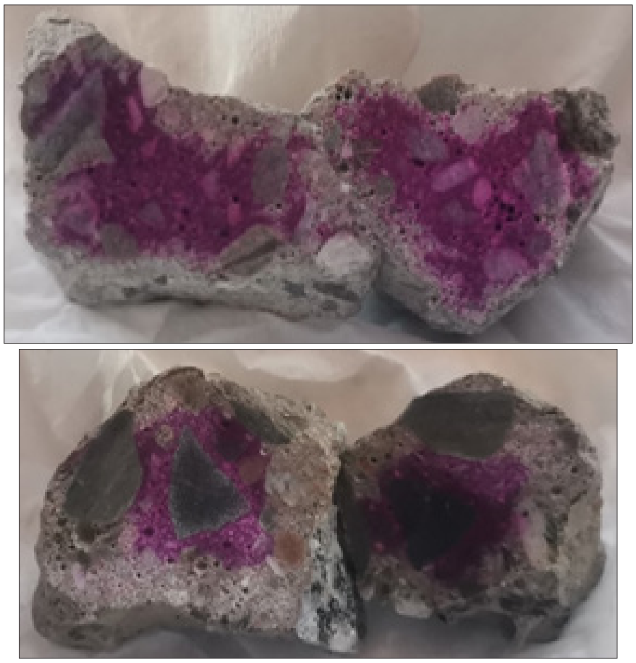

**Figure 2:** Naturally accelerated carbonation at atmospheric $CO_2$ concentration (400-500 ppm) with periodically wetting and drying cycles after the first (top) and second stage of the treatment (bottom) and respective carbonation depth of the 32 mm coarse recycled concrete aggregates.

The reference natural aggregate concrete blend, a mixture with 50% recycled concrete aggregate carbonated on the first stage, and a mixture with 50% recycled concrete aggregate successively treated on a second carbonation stage were prepared. The 50% addition referred to the total mass of the natural and recycled concrete aggregates.

The fresh and hardened state parameters, such as the effective water to cement ratios, the apparent density, the air content, the workability, the compressive strength, the modulus of elasticity, the porosity, the water permeability, the accelerated carbonation resistance, the accelerated chloride penetration and the freeze / thaw resistance were measured on concrete specimens (Standard EN 206 - Concrete - Specification, performance, production and conformity, 2021).

## Results and Discussion

The workability target of a consistency class C3 was reached for all mixtures that varied for 1.05 for the natural aggregate concrete to 1.1 for the mixture with 50% recycled carbonated aggregates (stage 2). A lower density and a higher air content for the recycled mixtures as compared to concrete with 100 % natural aggregates was seen (Table 1).

| Concreate made in laboratory | | | | |
|---|---|---|---|---|
| Fresh Concrete Characteristics | | | | |
| Mix Type | Density(kg/m$^3$) | Air Content (%) | w/c Ratio | Walz index |
| Natural mix | 2429 | 1.0 | 0.47 | 1.05 |
| Mix 50% recycled (stage 1) | 2261 | 3.7 | 0.35 | 1.08 |
| Mix 50% recycled with Carbonated aggregates (stage 2) | 2272 | 2.8 | 0.42 | 1.1 |

**Table 1:** Fresh state properties of the blends.

All blends exhibit a satisfactory compressive strength after 28 days of hydration. The mixture with the carbonated recycled aggregate treated after the second stage reached the reference strength. However, the w/c ratio of the concrete with natural aggregate was slightly higher. Nonetheless, the mixture with carbonated recycled aggregates after the first carbonation stage exhibits lower water / cement ratio, but also the lowest strength (Fig. 3 left). Hence, a direct relationship between strength and water / cement ratio is not always seen. The type and the carbonation level of the recycled concrete aggregates also influence the mechanical parameters. The concretes with 50% of the first and the second stage carbonated recycled concrete aggregates show slightly higher modulus of elasticity as compared to the reference. In spite of the relative difference in the water / cement ratios for the second stage carbonated recycled concrete aggregate concrete, the similar modulus of elasticity values as the reference blend can be positively considered. Hence, the reduction of the modulus of elasticity and the reduction of the mechanical properties by using recycled concrete aggregates (Wang et al., 2021) can be partially mitigated by the carbonation of the recycled concrete aggregates.

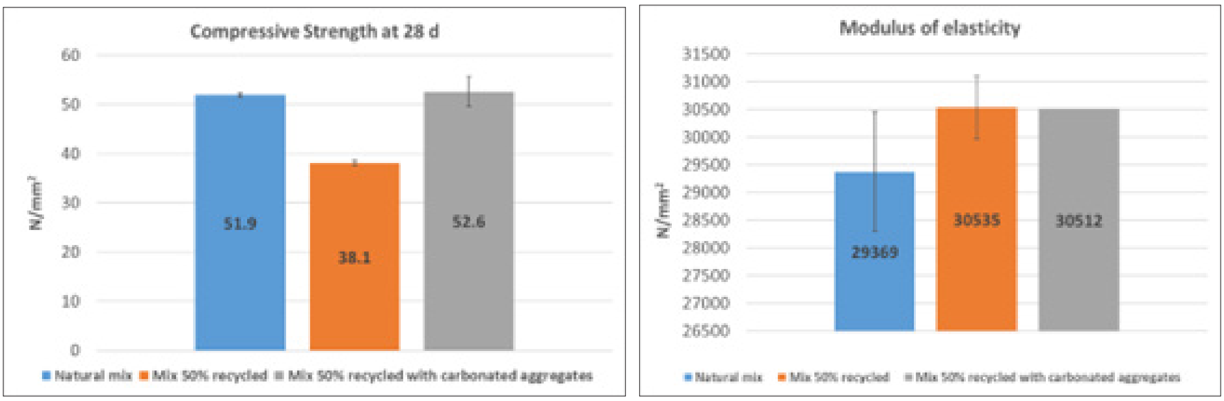


**Figure 3:** Compressive strength (left) and modulus of elasticity (right) of the blends.

The durability (Standard EN 206 - Concrete - Specification, performance, production and conformity, 2021), measured with the resistance to accelerated carbonation, indicates the concretes with 50% of carbonated recycled concrete aggregates after the first stage to have a higher carbonation resistance as the reference natural aggregate concrete. This fact can be related to both the lower water / cement ratio and the carbonation of the outer shell of the recycled concrete aggregates. The carbonation may close the porosity and hinder the $CO_2$ penetration. In fact, the concrete with the carbonated recycled aggregates after the second stage exhibit a higher resistance to accelerated carbonation tests, in spite of the similar water to cement ratio as compared to the reference natural aggregate concrete. This may partially be due to the increased filling of the porosity and the lower calcium hydroxide availability through the prolonged naturally accelerated carbonation reaction (Fig. 4 left). The freeze and thaw tests exhibit a higher resistance of the recycled concrete blends as compared to the concrete prepared with the natural aggregates. The further naturally accelerated carbonation with a second stage carbonation treatment slightly improves the carbonation resistance, although not significantly. Nonetheless, in this case, the water to cement ratio of 0.42 for the second stage carbonation treatment of the concrete aggregates is very close to the reference natural aggregate concrete of 0.47. Hence, the positive effect of the natural accelerated carbonation of the recycled concrete aggregates can be observed (Fig 4 right).

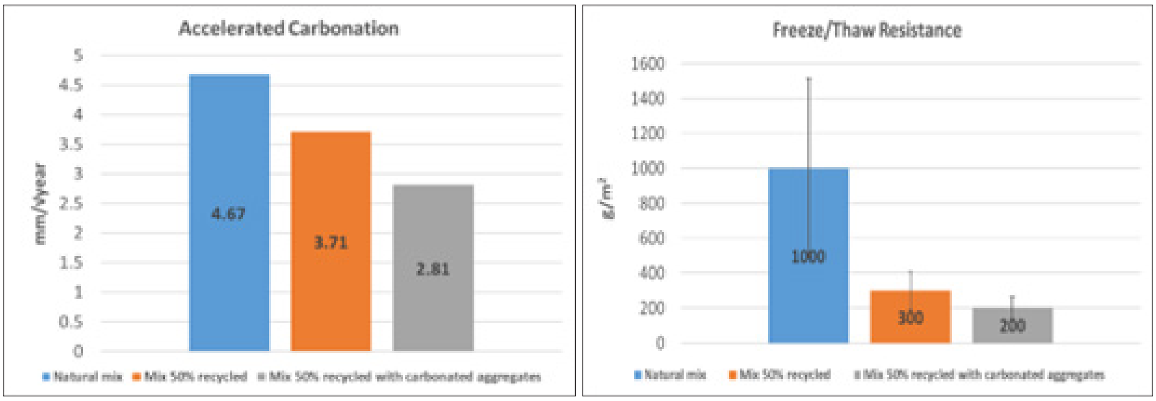


**Figure 4:** Accelerated carbonation depth (left) and frost resistance (right) of the concrete blends.

The mixtures with 50% carbonated recycled aggregates after the first and the second natural accelerated carbonation stage exhibit slight better resistance to chloride migration under an electric field (Standard EN 206 - Concrete - Specification, performance, production and conformity, 2021) as compared to the natural aggregate concrete. The further natural accelerated carbonation to a second stage does not significantly improve the resistance. The carbonates usually do not bind the chlorides, so that the chloride penetration appears mainly controlled by the porosity rather than the chemical bonding capacity. The porosity of the old mortar attached to the natural aggregates for the recycled concrete aggregates as compared to the virgin natural aggregates tends to be filled by the carbonation process through the formation of carbonates. Therefore, a similar behaviour with respect to the chloride penetration is seen for the blends, regardless of the different water cement / ratios (Fig. 5 left). The water permeability of the blends with natural aggregates is higher as compared to the recycled concrete blends and a further carbonation to the second stage does not relevantly reduce the permeability (Fig. 5 right). The permeability correlates with the main mechanical and durability performance of the blends, which indicates a lower water permeability of the recycled blends which in turn improves their mechanical and durability properties.

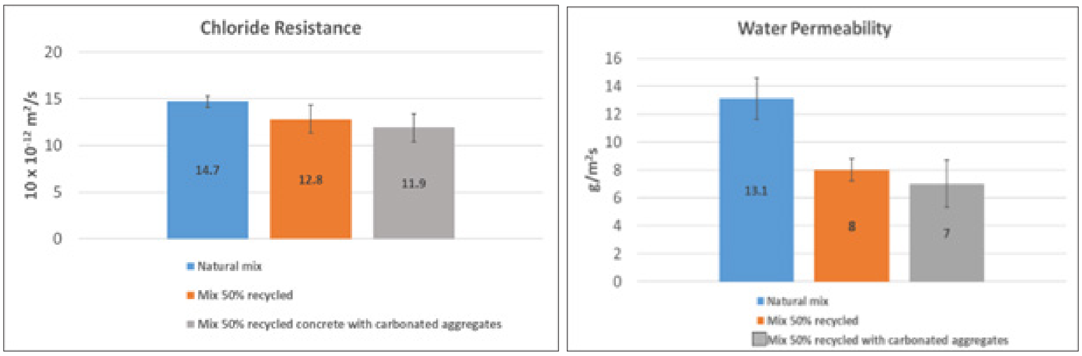


**Figure 5:** Chloride resistance (left) and water permeability (right) of the concrete blends.

The porosity values are quite similar between the mixes, with a slight increase for the recycled concretes. The carbonation likely closes some porosity, so that the recycled blends show similar values as the reference mixture.

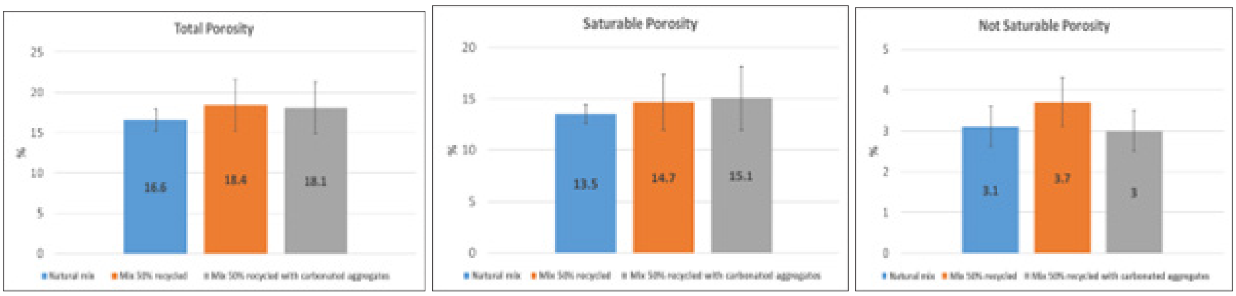


**Figure 6:** Porosity of the natural aggregates concrete and the naturally carbonated aggregates recycled concretes.

On the other hand, the water permeability does not correlate with the porosity. Nevertheless, the porosity values are not significantly different and the variation as well as the porosity present along the aggregate and the recycled aggregates interfaces and within the old mortar, cannot be clearly discriminated with conventional measurements techniques (Swiss standard association SN 505 262/1-SIA 262/1, 2013 - Concrete Structures – Supplementary specifications, Edition 2013, 2013).

Thus, the natural accelerated carbonation method can be applied on stationary or mobile recycling plants if enough space is available, although piles stocks can also be carbonated quite easily with adequate wetting and drying cycles. The accelerated carbonation under a natural $CO_2$ concentration by artificially increasing the wetting and drying cycle frequency improves the quality of the recycled concrete aggregates. The resulting recycled concretes exhibit satisfactory mechanical and durability performance, even if only an outer shell of carbonation with some millimeters thickness is present. From the point of view of the performance it is not necessary to totally carbonate the recycled concrete aggregates, unless you want to maximize the $CO_2$ sink.

## Conclusion

Concrete with recycled naturally carbonated aggregates in atmosphere with an increase in the wetting and drying cycles exhibit good mechanical properties by maintaining a good workability in the fresh state as for the reference concrete.

The durability parameters, such as the freeze / thaw resistance, the chloride penetration, the artificial accelerated carbonation and the water permeability exhibit better values for the concrete with the recycled naturally accelerated carbonated aggregates as compared to the natural aggregates concrete.

The partial closure of the porosity with the atmospheric natural accelerated carbonation may contribute to the positive effect on the mechanical and durability performance, although the first stage natural accelerated carbonation already significantly implements the properties. This in spite of the difference in the water / cement ratios of the blends.

A relative short-term naturally accelerated carbonation up to 8 months (maybe even lower) and the formation of an outer carbonated border (3-4 mm) on the recycled concrete aggregates is enough to reasonably implement the mechanical and durability performance of recycled concrete. The complete carbonation of the recycled aggregates may only act as an additional carbon sink.

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