A service life based global warming potential for high-volume fly ash concrete exposed to carbonation

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HIGHLIGHTS

- HVFA concrete is less resistant to carbonation than OPC concrete but still is durable.
- Colorimetric assessment underestimates the actual (microscopic) carbonation depth.
- Service life of HVFA concrete estimated with a simplified model exceeds 100 years.
- When curing and weather effects are considered, service life also exceeds 100 years.
- The GWP of carbonation exposed HVFA concrete is 18–27% lower than OPC concrete's GWP.

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ABSTRACT

To evaluate the global warming potential (GWP) of carbonation exposed high-volume fly ash (HVFA) concrete, its expected service life should be known. In the early stages of product development, this is done with rudimentary prediction models based on simple colorimetric carbonation testing. More sophisticated methods (e.g. thin section analysis) and prediction models that consider concrete curing and meteorological conditions (cf. Fib Bulletin 34) can be used later on if the former predictions look promising. This paper shows that both rudimentary and advanced prediction models result in significant GWP decrease (−18% to 27%) for HVFA concrete, regardless the underlying carbonation assessment method.

1. Introduction

Within the development process of a new potentially 'green' concrete type, it is at all time imperative to keep an eye on its expected reduced cement related greenhouse gas (GHG) emissions in comparison with traditional concrete. It is known that the production of ordinary Portland cement involves considerable CO_2 emissions. Based on in depth literature study this is around 842 kg per ton on average [1]. Now, the environmental benefit that could be achieved by replacing a considerable portion of the cement with supplementary cementitious materials such as fly ash (FA) will depend greatly on the strength and durability performance of the material. In case its compressive strength is lower in comparison with traditional concrete, structure dimensions will evidently be larger. As a result, more concrete will need to be manufactured to meet the structural requirements. If the concrete in its expected future natural environment – in this case an environment where carbonation-induced corrosion is at risk – will require regular maintenance, repair or even full replacement, the impact of the necessary rehabilitation actions will add on to the impact of the original concrete amount needed to build the structure [1].

Evidently, little information on the actual durability performance of a new concrete type is readily available in the early stages of product development, and this for two reasons. (i) The new material has not been applied yet in any real structure that could be regularly inspected for damage. (ii) Accelerated laboratory durability tests, e.g. experiments done to predict the time to carbonation-induced depassivation of the reinforcing steel, are time consuming. True, exposing the concrete at its proper testing age to an increased CO_2 concentration followed by a time-dependent evaluation of the carbonation depth may very well result in the experimental determination of a carbonation rate for the concrete composition under investigation. These carbonation depths could

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simply be visualized by using the well-known colour indicator phenolphthalein [2] or even more accurately with microscopy on thin sections [3,4]. However, without a full understanding of the concrete's curing behaviour and the influencing environmental factors of its natural environment even the most accurately determined experimental carbonation rate will not result in a realistic service life prediction on the short term. This is because the effect of curing can only be evaluated by performing carbonation tests at multiple testing ages. The expected meteorological conditions (relative humidity, amount of precipitation, etc.) of a specific environment in which the concrete would be used once on the market, should also be assessed more thoroughly. Logically, a quantification of these influencing factors asks for more effort and time within the product development process. Both items will be addressed only if a first rough service life and sustainability assessment based on rather easy-to-obtain data already gives promising results.

Another important aspect that deserves attention is the applied CO_2 concentration during an accelerated carbonation test. This concentration can vary considerably with the chosen test method. For instance, NBN B15-100 suggests a CO_2 concentration of 1% during the carbonation test for equivalent performance assessment of concrete with type II additions, but gives the freedom to work with other CO_2 concentrations as well. According to Fib Bulletin 34 concrete needs to be stored in a carbonation cabinet at 2% CO_2 for 28 days to determine its inverse effective carbonation resistance [5]. Lo and Lee applied the same CO_2 concentration [6]. NT Build 357 on the other hand specifies an atmosphere of 3% CO_2 [7] and Borges et al. expose concrete to a CO_2 concentration of 5% to accelerate carbonation [8]. In our research, the applied CO_2 concentration was 10% by volume cf. [9,10]. Literature survey also shows that still higher CO_2 concentrations are inherent to some test methods for accelerated carbonation. For instance, a CO_2 concentration of 20% was applied in [11]. The AFPC-AFREM procedure on accelerated carbonation testing is even more extreme as it requires a carbonation chamber conditioned at 50% CO_2 [12]. Thus, it is clear that there is not much uniformity among the available carbonation test methods. Nevertheless, some researchers are convinced that increasing the CO_2 concentration too much will result in important chemical changes of the concrete's paste fraction that would not occur if the concrete could naturally carbonate at 0.03%. Castellote et al. found that the polymerisation of the calcium silicate hydrate (C–S–H) after carbonation of OPC pastes increases with the increase of the CO_2 concentration applied [13]. When carbonating at 0.03% CO_2 and 3% CO_2 there is remaining C–S–H gel, although with a lower Ca/Si ratio than that of an uncarbonated sample (Ca/Si = 1.87), but still with the characteristics of this phase. The obtained Ca/Si ratios for the C–S–H gels of samples carbonated at 0.03% CO_2 and 3% are quite similar and amount to 1.23% and 1.18%, respectively. When carbonating at 10% and 100% CO_2 the C–S–H gel completely disappears. It is completely transformed into polymerised Ca-modified silica gel. In a CO_2 atmosphere of 3% or less, this transformation is only partial. The presence of remaining unhydrated cement and ettringite also highly depends on the imposed CO_2 concentration. After carbonation at 0.03% CO_2 and 3% CO_2, unhydrated cement and ettringite are still there. Testing at 10% CO_2, makes the ettringite completely disappear and leaves only a small amount of unhydrated cement. A CO_2 concentration of 100% results in a complete disappearance of both the ettringite and the unhydrated cement. Based on these findings Castellote et al. conclude that the maximum allowable CO_2 concentration for acceleration of the carbonation process is 3% [13]. Only under these conditions, a dramatical change in microstructure of the pastes is avoided. Nevertheless, they emphasize that these observations only hold true for OPC pastes and they advise that their validity should also be checked for pastes consisting of alternative binder materials (e.g. FA).

Borges et al. studied the carbonation behaviour of pastes containing high amounts of blast-furnace slag (BFS) by exposing them at 5% CO_2. When dealing with OPC + BFS pastes attention needs to be paid to both the carbonation of calcium hydroxide (CH) and C–S–H. The carbonation of CH usually results in a densification of the microstructure because the volume of the carbonates (calcite) formed is 11–12% greater than the volume of CH. Carbonation of C–S–H on the other hand causes a polymerisation of the silica chains in the C–S–H, which may be responsible for a volumetric decrease (shrinkage), a coarsening of the matrix and cracking [8]. There is evidence that CO_2 reacts simultaneously with the CH and the C–S–H [14]. This statement is also confirmed by Thiery et al. [15]. According to the latter, CH carbonation is initially more rapid than that of the C–S–H gel, but this situation soon reverses because of the formation of a layer of $CaCO_3$ microcrystals on the surface of CH crystals. Now, in blended cement pastes there is less CH available due to pozzolanic reaction. As a consequence, rapid decalcification of the C–S–H is expected when the paste is highly permeable to CO_2. This will result in carbonation shrinkage which is accelerated when the Ca/Si ratio drops below 1.2 [16]. In a blended paste characterised by a low permeability, carbonation of the low Ca/Si ratio C–S–H is believed to be less at risk. Borges et al. observed from permeability experiments that OPC + BFS paste is very sensitive to the carbonation shrinkage phenomenon, although carbonates filled some of the pores which increased the overall density and reduced the overall porosity [8]. Similar findings could be expected for blended pastes consisting of OPC and FA.

Yet, when considering the findings of Castellote et al. [13], at least part of the shrinkage due to the carbonation of C–S–H observed by Borges et al. [8] may also be attributed to the fact that the carbonation test was performed in an atmosphere with 5% CO_2 instead of the proposed maximum value of 3%. Thus, performing carbonation tests at a CO_2 concentration above 3% may overestimate the carbonation of C–S–H and thus the measured carbonation depths and rates that result from it.

However, this effect does not take into account another phenomenon inherent to carbonation. The production of calcium carbonate always coincides with the release of water. When carbonating concrete at a high CO_2 concentration, the amount of water produced could be more than the porous matrix is capable of expelling in the same time interval. The time needed to establish a condition of equilibrium again is believed to slow down the propagation of the carbonation depth [17,18]. da Silva et al. also mention that CO_2 solubility is low when high CO_2 concentrations are used. The penetrating CO_2 first needs to transform into acid in the presence of water before the actual carbonation reaction can take place and the amount of CO_2 capable of dissolving in water is limited [17]. If these mechanisms would turn out more dominant than the coarsening of the pore structure due to the carbonation shrinkage attributable to the acceleration of the carbonation test, applying a high CO_2 concentration would rather underestimate the carbonation depth and rate under field conditions.

Thus, depending on the prevailing mechanism – either the increase in permeability due to carbonation shrinkage [8] or the blocking effect of the water released during carbonation [18] together with the limited solubility of CO_2 in water [17] – experimentally estimated field carbonation rates from highly accelerated tests either over- or underestimate the carbonation resistance and expected service life of a concrete with a high content of supplementary cementitious materials in its natural environment. A calculation of the global warming potential based on this service life assessment may attribute a higher or lower environmental burden to the material than when the service life assessment would be based on a more natural carbonation test.