

ASSESSING THE CO₂-BINDING OF CONCRETE DURING ITS SERVICE LIFE

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Abstract

In order to assess the impact of cement production on the atmospheric CO₂ concentration and with it on the global warming, not only the CO₂ emissions but also the CO₂ capture by carbonation of cementitious products during their service life needs to be considered. The key parameter for a reasonably accurate estimation is the carbonation coefficient that allows to calculate the temporal evolution of carbonation depth and the CO₂ adsorption by the cement hydrates. Current approaches use a strength-based concept linking carbonation coefficient and compressive strength. This approach has proven to be valid for Portland cement-based concrete, but appears to be questionable in the case of concrete produced with blended cements, which are prevalent nowadays. Based on recent scientific findings showing an excellent correlation between the CO₂ binding capacity of concrete and the carbonation coefficient, an alternative approach is proposed.

1. Introduction

The increasing CO₂ concentration in the atmosphere is regarded as a governing factor for the observed global temperature increase. The important contribution of cement production to anthropogenic CO₂ emission with 5-6 % is indisputable [1]. However, due to carbonation of cement hydrates, part of the emitted CO₂ is recaptured. Calcium-silicate-hydrate (C-S-H) is decalcified, leading to the formation of a phase with a lower Ca/Si-ratio and CaCO₃ and portlandite is transformed into CaCO₃. [2]. Only if CO₂ binding by carbonation of cementitious products during their service life is considered as well, the environmental impact of cement can thoroughly be assessed.

Carbonation starts at the surface of mortar or concrete components. The carbonation depth as a function of the square root of time is described by the carbonation coefficient K_N . It depends on various parameters like water-to-cement-ratio (w/c) of the mortar or concrete and relative humidity (RH) [3]. Therefore, an assessment of the carbonation coefficient K_N is mandatory to calculate CO₂ uptake by cementitious materials during their service life. This has been the subject of several studies, in which the carbonation coefficient K_N was assessed based on concrete strength [4-7]. In addition to CO₂ adsorption during its service life, a substantial amount of CO₂ is bound during the recycling phase of concrete [4-7]. This aspect is not considered in this study, as it solely focuses on service life.

A major drawback of the majority of studies dealing with CO₂ adsorption of cementitious materials is that the assessment of the carbonation coefficient K_N is based on concrete strength or that attempts are made to link the obtained experimental values only with strength-based concepts. Currently, CEN TC 229 WG 5 is following this path. However, mortar and concrete produced with blended cements do not show the same relation between carbonation coefficient K_N and compressive strength as mortar and concrete produced with Portland cement [8-10]. The share of blended cement used worldwide is steadily increasing in an attempt to decrease CO₂ emissions. Therefore, alternative approaches to assess the carbonation coefficient of mortar and concrete should be considered. In this paper, the CO₂ binding capacity of mortar and concrete as a tool to assess carbonation coefficient K_N of mortar and concrete is explored.

2. CO₂ binding capacity of mortar and concrete

The scientific background of the presented approach was published in Leemann et al. [9] and Leemann and Moro [10]. In these studies mortar and concrete mixtures containing various mineral additions were exposed both to accelerated and natural carbonation in sheltered and unsheltered conditions. They showed that it is possible to assess carbonation coefficient K_N based on the water-to-CaO-ratio (w/CaO) in concrete (Fig. 1). Hereby, only CaO present in clinker and reactive mineral additions like fly ash or slag but not in unreactive ones like limestone powder are taken into account. The w/CaO can easily be calculated, if the cement composition is known. It expresses the CO₂ binding capacity per volume of cement paste and, as it is a mass ratio between water and solid, it additionally contains information about porosity and microstructure. It shows a considerably better correlation with the carbonation coefficient K_N determined in sheltered ($R^2 = 0.90$) and unsheltered ($R^2 = 0.83$) outdoor exposure (Fig. 1) than the compressive strength. Based on these findings, the following empirical equations can be approximated for sheltered and unsheltered exposure [21]:

$$K_{N,S} = C \cdot (8.3 \cdot w/CaO - 4.7) \quad C = 1.00 \quad (1)$$

$$K_{N,US} = C \cdot (8.3 \cdot w/CaO - 4.7) \quad C = 0.47 \quad (2)$$

Of course, these equations are representative for the environmental conditions at the specific exposure site. In this case, yearly averages of temperature, RH and precipitation were 11.4 °C, 78 % and 1200 l/m², respectively. The slope of the linear regression is mainly dependent on the moisture state of the concrete. The value of C can be adapted to different humidity conditions dependent on the exposure site. Assessing carbonation coefficient K_N and

carbonation depth d_c based on the w/CaO is advantageous compared to a strength-based concept, because it is chemistry-based and as such takes into account cement composition and with it the cement-specific carbonation resistance.

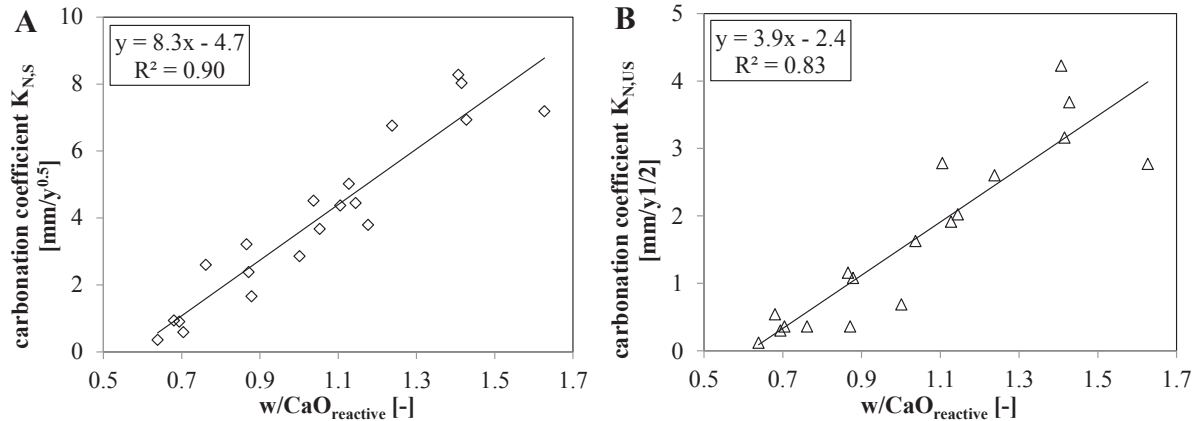


Figure 1: Carbonation coefficient $K_{N,S}$ in sheltered (A) and carbonation coefficient $K_{N,US}$ in unsheltered (B) outdoor exposure as a function of the w/CaO_{reactive} according to [10].

3. Calculation of CO₂-binding

As calculation examples, Swiss cements as described in Environmental Product Declarations (EPD) [11] are used, representing the average composition of the specific cement types (CEM I, CEM II/A, CEM II/B, CH-total). The key figures for the model calculation assumed typical Swiss concrete compositions used for house building. This type of concrete represents about 35 % of the concrete produced in Switzerland. For other types of concrete used for different types of structures, the input parameters like cement type and w/c leading to different values of w/CaO can easily be adapted using software as basic as an excel spreadsheet. The following comments provide additional information on some of the parameters used in the exemplified calculation shown in Table 1:

- The empirical value for the degree of carbonation a_c corresponds to the relative amount of carbonated CaO in concrete under natural conditions (1= CaO fully carbonated, 0= no CaO carbonated). It has been reported to be about 0.50 in [12].
- The content of the reactive CaO in Swiss cements has been determined in [9]. Only the CaO reacting during cement hydration is taken into account. Limestone powder for example is not included.
- Carbonation coefficients in sheltered ($K_{N,S}$) and unsheltered ($K_{N,US}$) conditions have been calculated according to Eq. 1 and Eq. 2, respectively. In the exemplified calculation, the ratio of sheltered to unsheltered surfaces has been assumed to be 1. This ratio has to be adapted for individual cases. The sum of sheltered and unsheltered carbonation depth is finally represented by $d_{c,tot}$.
- A service life of 100 years is used for this calculation corresponding to EN 16757 [13].
- The implications of carbonation on reinforcement corrosion are not elaborated in this publication, as it is focusing on CO₂-binding. Possible implications between carbonation and corrosion have to be considered in the structural design.

- The thickness of the concrete element is assumed to be 200 mm. This value refers to 1 m³ of concrete, allowing to derive the surface area exposed to carbonation.
- The ratio of CO₂-uptake to emission of concrete (U/E_{conc}) is calculated using the carbonation depth $d_{c,tot}$, the degree of carbonation a_c and the thickness of the concrete x .
- The CO₂ emission resulting from calcination (kg CO₂/kg cement) was published in the EPD for Swiss cements (www.cemsuisse.ch) following SN EN 15804+A1 [14].
- The final CO₂ uptake by concrete is calculated in kg CO₂/t of concrete.

Table 1: Exemplified calculation of CO₂-binding in concrete used for housing during its service life applied for Swiss cements (average composition corresponding to [11]).

parameter	abbr.	units	CEM I	CEM II/A	CEM II/B	CH-total	source / calculation
degree of carbonation (abs)	a_c	%	0.50	0.50	0.50	0.50	Thiery et al., 2013
water-cement ratio	w/c	-	0.60	0.60	0.60	0.60	typical Swiss value
cement content	c	kg/m ³	280	280	280	280	typical Swiss value
water content	w	kg/m ³	168	168	168	168	typical Swiss value
CaO content of cement	$CaO_{reactive}$	mass-%	0.61	0.53	0.44	0.53	Leemann et al., 2015
CaO content of concrete	$CaO_{reactive}$	kg/m ³	171	147	123	149	$CaO_{reactive} \cdot c$
water/CaO-ratio	$w/CaO_{reactive}$	-	0.98	1.14	1.36	1.13	$w/CaO_{reactive}$
carbonation coefficient sheltered	$K_{N,S}$	mm/y ^{0.5}	3.46	4.77	6.63	4.66	Eq. 1
carbonation coefficient unsheltered	$K_{N,US}$	mm/y ^{0.5}	1.63	2.24	3.12	2.19	Eq. 2
reference service life	RSL	y	100	100	100	100	see EN 16757 [25]
carbonation depth sheltered at end of service life	$d_{c,S}$	mm	34.6	47.7	66.3	46.6	$K_{N,S} \cdot y^{0.5}$
carbonation depth unsheltered at end of service life	$d_{c,US}$	mm	16.3	22.4	31.2	21.9	$K_{N,US} \cdot y^{0.5}$
sum of carbonation depth	$d_{c,tot}$	mm	50.9	70.1	97.4	68.5	$d_{c,S} + d_{c,US}$
thickness of concrete element	x	mm	200	200	200	200	related to 1 m ³ of concrete
CO ₂ emission from calcination of cement	E_{cem}	kg CO ₂ /kg of cement	0.474	0.413	0.357	0.422	EPD cemsuisse, 2015
ratio of CO ₂ uptake-to-emission of concrete	U/E_{conc}	-	0.127	0.175	0.244	0.171	$d_{c,tot} \cdot a_c / x$ (Eq. 3)
density of concrete	ρ_{conc}	t/m ³	2.4	2.4	2.4	2.4	
CO ₂ -uptake per volume of concrete	U	kg/m ³	16.9	20.3	24.3	20.2	$c \cdot E_{cem} \cdot (U/E_{conc})$
CO₂-uptake per mass of concrete	U	kg CO₂/t of concrete	7.0	8.4	10.1	8.4	U/ρ_{conc}

As shown in Table 1, the ratio of CO₂ uptake to CO₂ emission (U/E_{conc}) per volume of concrete can be calculated according to Eq. 3:

$$U/E_{conc} = (\text{CO}_2 \text{ uptake})/(\text{CO}_2 \text{ emission}) = (d_{c,tot} \cdot a_c)/x \quad (3)$$

The ratio U/E_{conc} increases with decreasing thickness of the concrete element as its surface-to-volume-ratio increases (Fig. 2). The higher $d_{c,tot}$ and a_c are, the higher is the ratio U/E_{conc} . The ratio U/E_{conc} was calculated for the three different cement types and the average of cement

types used in Switzerland as presented in Table 1 using Eq. 3. Concrete mix design (cement type, cement content, w/c) and dimensions of the concrete elements can easily be adapted in the model calculation. Fig. 2 shows how the ratio U/E_{conc} changes as a function of the thickness of a given concrete element.

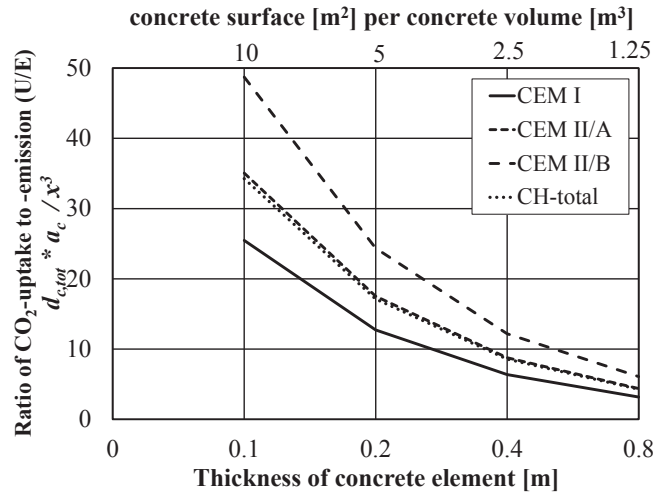


Fig. 2: Ratio of CO₂ uptake to CO₂ emission of concrete (U/E_{conc}) as a function of thickness of concrete element.

4. Limitations of assessing CO₂-binding

Any calculation of CO₂-binding has its limitations, as simplifications have to be made in order to achieve a manageable degree of complexity. This applies on the approach presented in this paper, as it does for strength-based concepts as well. One key question is the role of RH. Progress of carbonation shows a non-linear response to changes in RH [3,10]. Moreover, both porosity and RH change with concrete depth [15,16]. This point has to be studied further, because the carbonation depth as calculated in this study may be overestimated and therefore, an adaptation of the carbonation coefficients $K_{N,US}$ and $K_{N,S}$ used to calculate long-term CO₂-binding may be required.

5. Summary

CO₂-binding of concrete during its service life is an important point to be taken into account, when the impact of cement production on atmospheric CO₂ concentration and global warming is considered. It can be calculated by assessing the carbonation coefficient K_N and the carbonation depth d_c . This paper presents an approach based on the w/CaO of concrete, as this parameter governs the carbonation coefficient K_N . Examples for the implementation of this approach are given using boundary conditions of cement and concrete as used for housing in Switzerland. The two key parameters for the w/CaO are cement composition and w/c. They can easily be adapted for specific concrete compositions, climatic conditions and service life. The presented approach seems to be especially advantageous when the increasing use of blended cements is considered. In this scenario, the strength-based concept has clear limitations. Therefore, it is proposed that the presented approach is used instead of or in addition to existing strength-based concepts. This approach was not incorporated into EN

16757 [25], but it is currently under consideration to be included into the Technical Report “Carbonation” by CEN TC 104 SC1 TG20.

As shown in this paper, the amount of CO₂ bound by concrete during its service life is substantial compared to the emission of geogenic CO₂ during cement production. A comprehensible assessment of recaptured CO₂ is paramount for a realistic life cycle assessment.

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