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


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Carbonation Rate of Alkali-Activated Concretes: Effects of Compositional Parameters and Carbonation Conditions

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Abstract. The current ability to predict the carbonation resistance of alkali-activated materials (AAMs) is incomplete, partly because of widely varying AAM chemistries and variable testing conditions. To identify general correlations between mix design parameters and the carbonation rate of AAMs, RILEM TC 281-CCC Working Group 6 compiled and analysed carbonation data for alkali-activated concretes and mortars from the literature. For comparison purposes, data for blended Portland cement-based concretes with a high percentage of SCMs ($\geq 66\%$ of the binder) were also included in the database. The results show that the water/CaO ratio is not a reliable indicator of the carbonation rate of AAMs. A better indicator of the carbonation rate of AAMs under conditions approximating natural carbonation is their water/(CaO + MgO_{eq} + Na₂O_{eq} + K₂O_{eq}) ratio, where the index 'eq' indicates an equivalent amount based on molar masses. This finding can be explained by the CO₂ binding capacity of alkaline-earth and alkali metal ions; the obtained correlation also indicates an influence of the space-filling capability of the binding phases of AAMs, as for conventional cements. However, this ratio can serve only as an approximate indicator of carbonation resistance, as other parameters also affect the carbonation resistance of alkali-activated concretes. In addition, the analysis of the dataset revealed peculiarities of accelerated

tests using elevated CO₂ concentrations for low-Ca AAMs, indicating that even at the relatively modest concentration of 1% CO₂, accelerated testing may lead to inaccurate predictions of their carbonation resistance under natural exposure conditions.

Keywords: Alkali-activated materials · Durability · Carbonation · Pore structure · Accelerated testing

1 Introduction

The carbonation of cementitious materials such as concrete, *i.e.*, their reaction with atmospheric CO₂, modifies the chemistry and microstructure of the materials and their pore solution, potentially leading to the corrosion of embedded steel reinforcement. This degradation mechanism can have significant consequences, in many cases being the limiting factor for the service life of reinforced concrete structures [1]. For concretes based on conventional cements (Portland cements and blended Portland cements), it has been established that the water-to-reactive CaO ($w/\text{CaO}_{\text{reactive}}$) ratio is the major parameter determining their carbonation resistance [2, 3]. However, for concretes and mortars based on alkali-activated binders and other alternative cements this approach is not applicable without modifications [4–6]. Alternative approaches that apply specifically to alkali-activated concretes were previously not available, mainly because the mechanisms and factors determining the carbonation resistance of alkali-activated concretes are not yet completely understood. This is partly due to the wide range of chemistries of alkali-activated binders, notably their variability in CaO content, and the fact that individual carbonation studies were generally limited to only a few mix designs, curing conditions and exposure conditions.

To address these issues, the Working Group 6 of RILEM TC 281-CCC compiled and analysed carbonation data, for alkali-activated concretes and mortars with a wide range of chemistries, obtained from published studies. This has been done with the aim of identifying general correlations between the mix design parameters of alkali-activated concretes and mortars, and their carbonation resistance. For comparison purposes, data for blended Portland cement-based concretes and mortars with a high percentage of supplementary cementitious materials (SCMs) in the binder were included in the database. The analysis and the underlying dataset have been made available [7]. In the present contribution, the main results of the analysis will be summarised and discussed as regards the underlying microstructural characteristics and chemical processes.

2 Data Inventory and Evaluation

A full description of the dataset and its processing and evaluation is given in ref. [7]. In brief, carbonation depth (d_c)-versus-carbonation duration (t) data for alkali-activated concretes or mortars, for concretes or mortars based on blended Portland cements with at least 70% SCMs in the binder, as well as for concretes or mortars based on CEM III/B [66–80% ground granulated blast furnace slag (GGBS) in the binder according to EN

197-1] as well as their mix designs and the chemical compositions of their starting materials were compiled from the literature. The carbonation depths in those studies were generally determined by spraying a phenolphthalein solution (usually with a concentration of 1%) on fracture surfaces, except in two cases. (The limitations of this method have been discussed in ref. [7].) Only data obtained at controlled relative humidity were included in the database, while results for unsheltered natural exposure were not considered.

Data for concretes and mortars were treated jointly because previous studies indicate only a minor or no influence of aggregate content and grading, within the limits of normal concretes and mortars, on carbonation resistance [8–10]. Carbonation data for pastes were not included in the dataset.

Based on their mix designs, the alkali-activated concretes/mortars in the dataset were categorised as ‘GGBS-based AAC’ (fraction of GGBS in the solid binder precursors $\geq 50\%$; number of concretes/mortars in the dataset: 125), or ‘FA- and MK-based AAC’ (fraction of fly ash or metakaolin in the solid binder precursors $> 50\%$; number of concretes/mortars in the dataset: 12), or ‘other AAC’ (alkali-activated binders based on MSWI bottom ash, silico-manganese slag or natural pozzolans, as well as hybrid cements; number of concretes/mortars in the dataset: 18). Most of the AAC in the database were activated with sodium silicate solutions, while some were activated with NaOH, Na₂CO₃, or a mix of sodium silicate and Na₂CO₃. In some cases, portlandite or gypsum were employed as additional activators.

The concretes and mortars based on blended Portland cements in the database are referred to as ‘BCC’ (number of concretes/mortars in the dataset: 74). The SCMs in these materials were generally GGBS and/or FA; in one case, a natural pozzolan was added to a CEM III/B.

From the d_c -versus- t data, the carbonation coefficients (k_c) of the materials were calculated as usual, *i.e.*, as $k_c = d_c/\sqrt{t}$ when only one data point was available, or by fitting a linear function to the available data points plotted as $d_c(\sqrt{t})$ to a maximum of $t \approx 1$ year. Unphysical fits (*i.e.*, fits yielding a negative d_c at $t = 0$ d) were repeated with the linear function forced through the origin of the coordinates. (The limitations of this approach have been discussed in ref. [7].) For carbonation coefficients obtained under conditions approximating natural exposure (*i.e.*, CO₂ concentration: $\sim 0.04\%$) the symbol $k_{c\text{-nat}}$ will be used, while carbonation coefficients obtained under accelerated conditions will be denoted $k_{c\text{-acc}}$ or $k_{c\text{-acc}}[c_{\text{CO}_2,\text{acc}}]$, where $c_{\text{CO}_2,\text{acc}}$ is the CO₂ concentration, or concentration range, applied for accelerated testing.

From the mix designs of the concretes/mortars and the chemical compositions of their constituents, several parameters that may exert an influence on the carbonation resistance were computed. In cases where these parameters are water-to-oxide mass ratios (w/oxide), the value used for the oxide is the total amount of that oxide in the binder paste, including the contribution of the solid fraction of the activator. For example, $w/\text{Na}_2\text{O}$ is the ratio between the water and the amount of Na₂O in the paste, the latter being the sum of the Na₂O contributed by the activator and the Na₂O present in the solid precursors of the binder. No attempt was made to discriminate “reactive” and “non-reactive” oxides in the paste, as the information given in the reports containing the

carbonation data was usually not sufficient to calculate these fractions with reasonable accuracy.

For a combined consideration of the alkali metal and alkaline-earth metal oxides in the binder pastes, the masses of Na_2O , K_2O , and MgO were converted to equivalent masses (denoted $\text{Na}_2\text{O}_{\text{eq}}$, $\text{K}_2\text{O}_{\text{eq}}$, and MgO_{eq} , respectively) by multiplying them by the ratio of the molar mass of CaO to the molar mass of the respective oxide; for example, $\text{Na}_2\text{O}_{\text{eq}} = \text{Na}_2\text{O} \cdot M_{\text{CaO}} / M_{\text{Na}_2\text{O}}$. Through this conversion, the equivalent masses reflect the theoretical maximum CO_2 binding capacity of the respective oxides, referred to the binding capacity of CaO on a molar basis.

3 Results and Discussion

3.1 Carbonation Under Conditions Approximating Natural Exposure

Figure 1(A) shows a plot of the carbonation coefficients obtained under conditions approximating natural carbonation *versus* the w/CaO ratios of the binders of the concretes/mortars. The data for the FA-based AAC, and particularly the MK-based AAC, deviate considerably from a linear relationship that might otherwise be considered for the more Ca-rich binders (BCC and GGBS-based AAC) in the plot. Clearly, a relationship does not exist between the carbonation coefficients of AAC under conditions approximating natural exposure and their w/CaO .

This lack of a correlation between $k_{\text{c-nat}}$ and w/CaO for AAC is due to the fact that AAC generally contain significant amounts of alkali ions (in most cases Na^+) and that these have an important effect on the carbonation of these materials, including the carbonation products and the pH of the pore solution [4, 11, 12]. Taking this into account, and considering that Mg^{2+} is also able to bind CO_2 , it may be expected that a better correlation exists between $k_{\text{c-nat}}$ and $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}})$. This was indeed observed for the present dataset [Fig. 1(B)], though the scatter of the data is still considerable (coefficient of determination, assuming a linear relationship and excluding the data point for the MK-based AAC, $R^2 = 0.4748$). The deviations from a common linear relationship can be assigned to the influence of additional factors influencing the carbonation resistance of alkali-activated materials (e.g., type of activator, curing temperature, extent of reaction), not considered in this plot. It is noted, however, that the present dataset did not allow to identify a systematic influence of activator content or the silicate modulus ($\text{SiO}_2/\text{Na}_2\text{O}$ or $\text{SiO}_2/\text{K}_2\text{O}$ in mol/mol) of the activator on the carbonation resistance of the AAC [7].

The linear fit of the data, excluding the MK-based AAC, yields $k_{\text{c-nat}} = 0 \text{ mm}/\sqrt{\text{d}}$ at $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}}) = 0.45$ [Fig. 1(B)]. This can be rationalised by considering the processes occurring during the hydration of cements. The hydration of cements leads to an increase of the volume of solids in a cement paste, which gradually fills the initially water-filled capillary pores of the paste. At a certain degree of hydration, and thus pore filling, the capillary pore system may become discontinuous (depercolated), leading to a permeability that is orders of magnitude lower than the permeability of a paste with a fully continuous (percolated) capillary pore system [13]. This, in turn, would decrease the carbonation rate to values close to zero, because the progress of carbonation depends largely on the ingress of CO_2 into the paste.

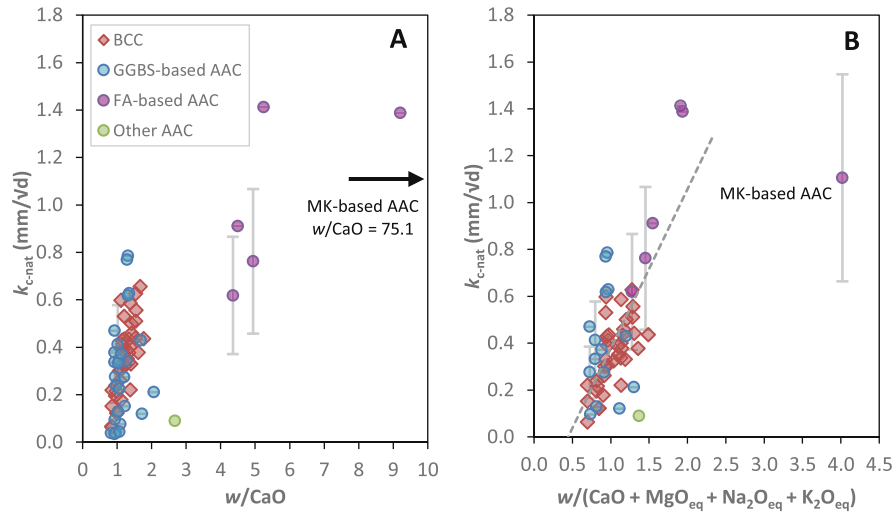


Fig. 1. Carbonation coefficients obtained under conditions approximating natural carbonation (indoor/sheltered) versus w/CaO (A) and versus $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}})$ (B). Error bars represent the estimated standard deviations of the results obtained in the round robin testing programme by RILEM TC 247-DTA. In panel A, the data point for the MK-based AAC with $w/\text{CaO} = 75.1$ and $k_{c\text{-nat}} = 1.1 \text{ mm}/\sqrt{\text{d}}$ is far outside the displayed range. In panel B, the dashed line is a linear fit through all data points except the MK-based AAC. (Data from ref. [7].)

Whether capillary discontinuity is reached depends on the water/cement ratio (w/c) of a paste and the degree of hydration of the cement. For a specific Portland cement, Powers [13] determined that the highest w/c that allows the capillary pore system to become discontinuous at full hydration is ~ 0.70 . Bentz and Garboczi [14] found that the capillary pore systems of C_3S and Portland cement pastes become discontinuous at a porosity of $\sim 18\%$, and computed combinations of w/c and degree of hydration at which this porosity is reached; for example, for $w/c = 0.50$, capillary discontinuity is reached at a degree of hydration of 86%, and for $w/c = 0.35$, capillary discontinuity is reached at a degree of hydration of 57%.

As the reactions of alkali-activated materials comprise the formation of reaction products with a volume higher than that of the precursors, depercolation of the capillary pore system can, in principle, occur also in AAC. Further, for each specific AAC, $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}})$ is related to the water/binder ratio (w/b). It follows that depercolation of the capillary pore systems can be expected for finite values of $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}})$ and the degree of reaction of an AAC.

The $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}})$ at which capillary discontinuity occurs will strongly depend on the starting materials and the mix design of a specific AAC. Numerical values have not yet been calculated systematically across the range of compositions of alkali-activated materials. However, for GGBS activated by sodium silicate or Na_2CO_3 , simulation results [15] indicate that at $w/b = 0.40$, a porosity of 18% is reached at $\sim 90\%$ extent of reaction, so it is expected that a comparatively low water content will be required to achieve capillary depercolation of AAC in practice, due to

the relatively low space-filling character of the key AAC binding phases compared to those in concretes based on conventional cements.

3.2 Carbonation Testing Under Accelerated Conditions

The carbonation resistance of cementitious materials is often assessed using accelerated tests, *i.e.*, tests at elevated CO₂ concentrations, to obtain results faster than what is possible under natural conditions. Assuming that the carbonation of concrete follows the square root-of-time law [8], the relationship between the carbonation coefficients obtained with accelerated carbonation testing and natural carbonation or conditions approximating natural exposure can be derived as:

$$\frac{k_{c-\text{acc}}}{k_{c-\text{nat}}} = \sqrt{\frac{c_{\text{CO}_2,\text{acc}}}{c_{\text{CO}_2,\text{nat}}}} \quad (1)$$

where $c_{\text{CO}_2,\text{acc}}$ is the CO₂ concentration applied in accelerated carbonation testing, and $c_{\text{CO}_2,\text{nat}}$ is the natural CO₂ concentration in air (0.04%).

There are some experimental data indicating at least an approximate validity of Eq. (1) for BCC, though in more cases the deviations between measured values and the values predicted by the equation are significant [8]. Whether Eq. (1) holds for alkali-activated materials had not previously been systematically investigated.

Figure 2 shows a comparison of the carbonation coefficients obtained under accelerated conditions ($c_{\text{CO}_2,\text{acc}} = 1\%$ and 3–4%, respectively) with the carbonation coefficients of the same materials obtained under conditions approximating natural exposure; the relationships between these values, predicted by Eq. (1), are also shown. The data points for the GGBS-based and the BCC appear to follow Eq. (1) (albeit with considerable scatter), while the data for the FA- and MK-based AAC clearly deviate from the predicted relationships. Specifically, the carbonation coefficients obtained under accelerated conditions were always significantly smaller than what would be expected from application of the square root-of-time law, *i.e.*, Eq. (1).

This result indicates that the outcomes of accelerated carbonation testing of FA- and MK-based AAC (*i.e.*, low-Ca alkali-activated materials) lead to inaccurate predictions of their carbonation resistance under natural exposure; *viz.*, a measurement of $k_{c-\text{acc}}$ and its conversion *via* Eq. (1) yields a $k_{c-\text{nat}}$ that is lower than the result of a measurement of $k_{c-\text{nat}}$. However, Fig. 2(A) shows that the *relative* performance of the FA- and MK-based AAC in accelerated and natural testing, *i.e.*, the order of the obtained carbonation coefficients, is the same for testing at a CO₂ concentration of 1% and under conditions approximating natural exposure. This has been noted earlier in the round robin testing programme of RILEM TC 247-DTA [6], and the present dataset contains an additional data point confirming this finding. However, the number of available studies of low-Ca alkali-activated materials that include both accelerated and natural carbonation testing results for the same materials is currently very limited; thus, additional data are required to verify these conclusions.

In line with the identified shortcomings of accelerated carbonation testing of FA- and MK-based AAC, the coefficient of correlation for the relationship between $k_{c-\text{acc}}[1\%]$ of the materials in the dataset and their $w/(\text{CaO} + \text{MgO}_{\text{eq}} + \text{Na}_2\text{O}_{\text{eq}} + \text{K}_2\text{O}_{\text{eq}})$ was lower

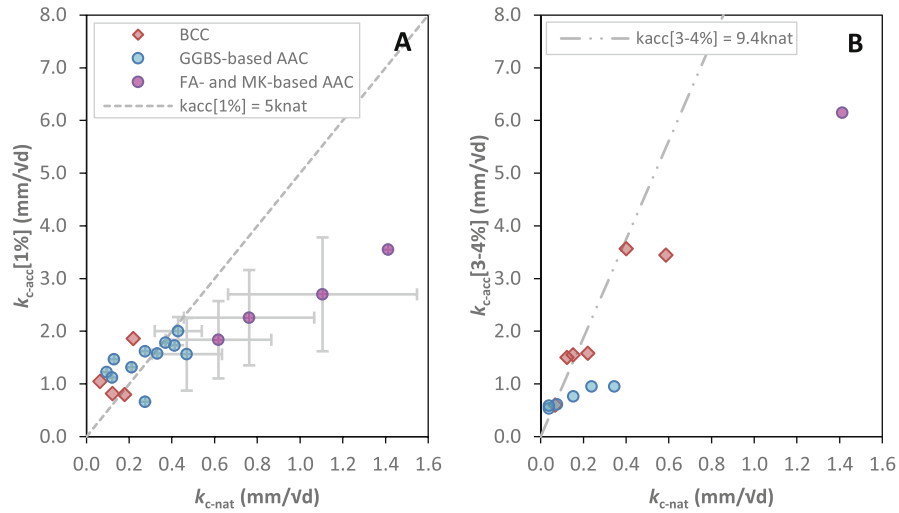


Fig. 2. Carbonation coefficients obtained under accelerated conditions (**A**, CO₂ concentration: 1%; **B**, CO₂ concentration: 3–4%) versus carbonation coefficients obtained under conditions approximating natural carbonation (indoor/sheltered). Error bars represent the estimated standard deviations of the results obtained in the round robin testing programme by RILEM TC 247-DTA. The dashed line (panel **A**) and the dash-dotted line (panel **B**) represent the relationship predicted by Eq. (1). (Data from ref. [7].)

than for the relationship between k_{c-nat} and $w/(\text{CaO} + \text{MgO}_{eq} + \text{Na}_2\text{O}_{eq} + \text{K}_2\text{O}_{eq})$ [7]. For carbonation testing under higher CO₂ concentrations ($c_{\text{CO}_2} \geq 3\%$), no correlation between k_{c-acc} and $w/(\text{CaO} + \text{MgO}_{eq} + \text{Na}_2\text{O}_{eq} + \text{K}_2\text{O}_{eq})$ or any other examined compositional parameter was observed, indicating that the significance of these tests is very limited [7].

4 Conclusions

The carbonation coefficients of GGBS- and FA-based AAC obtained under conditions approximating natural exposure exhibited a noticeable, though weak, correlation with their $w/(\text{CaO} + \text{MgO}_{eq} + \text{Na}_2\text{O}_{eq} + \text{K}_2\text{O}_{eq})$ ratio. This can be attributed to the CO₂ binding capacity of alkali metal and alkaline-earth metal ions. The correlation included also the high-volume SCM concretes (BCC) in the present dataset; thus, alkali-activated concretes and high-volume SCM concretes appear to behave similarly in this respect. However, the scatter of the pertinent data was considerable; thus, the $w/(\text{CaO} + \text{MgO}_{eq} + \text{Na}_2\text{O}_{eq} + \text{K}_2\text{O}_{eq})$ ratio can serve only as an approximate indicator of the carbonation resistance, and other factors that influence the carbonation rate must be considered in addition when accurate predictions of the carbonation resistance of alkali-activated concretes are to be obtained.

Accelerated carbonation testing at CO₂ concentrations in the range 1–4% of FA- and MK-based AAC (low-Ca alkali-activated materials) consistently yielded significantly lower carbonation coefficients than what would be expected from carbonation testing

under conditions approximating natural exposure and application of the square root-of-time law. Thus, the present data indicates that accelerated carbonation testing and conversion of the obtained values using the square root-of-time law leads to inaccurate predictions of the carbonation resistance of FA- and MK-based AAC materials under natural conditions. However, there is only limited data available in this regard; thus, more studies comparing the results obtained under accelerated carbonation conditions and conditions approximating natural exposure of low-Ca alkali-activated materials are required.

For CO₂ concentrations $\geq 3\%$, the present dataset showed no correlation with any of the examined mix design parameters, including $w/(CaO + MgO_{eq} + Na_2O_{eq} + K_2O_{eq})$, indicating that the establishment of a relationship between these results and the carbonation resistance under natural conditions will be difficult to achieve. However, current developments in cement and concrete testing indicate that these highly accelerated carbonation tests will be largely abandoned anyways, due to their unrealistic impacts on material chemistry and microstructure compared to natural conditions.

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