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CO_2 uptake by carbonation of concrete during life cycle of building structures

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Abstract The cement manufacturing process has been responsible for about 5 to 7 % CO_2 emissions. However, during its life cycle, concrete structures are submitted to carbonation and can uptake part of CO_2 emitted during its construction. In order to corroborate this potential, this study applied mathematical modeling to evaluate the performance of compression strength from a 20, 30 and 40 MPa concrete produced with different types of cements (CP III, CP IV and CP V, ARI) from 0 to 100 years of age. Based on these parameters, it was found out that CO_2 uptake is directly ratable to the concrete superficial area exposed to CO_2, influenced by the type of cement and concrete strength. CO_2 emissions balance may become an indicator of sustainability and, in the future, it can be considered a compensatory measure in the concrete structures project.

Keywords CO_2 uptake · Carbonation · Concrete structures · Sustainability

1 Introduction

The increasing global consumption of cement and concrete has led to some discussions concerning possibilities to minimize emissions related to Portland cement production. It is estimated that more than 2 billion tons of carbon dioxide have been emitted in the last decade due to Portland cement process [1]. This fact ranks cement as one of the products that emits a huge amount of CO_2. Thus, it represents almost 5 % total anthropogenic emissions [2]. Another important fact to be highlighted is that 50 % of these emissions are a result of limestone calcination (decarbonation) and the other part comes from fuel combustion during power generation to produce clinker. However, during cement lifetime, due to the reaction between CO_2 that takes part of this reaction through concrete and calcium hydroxide ($Ca(OH)_2$) in the presence of water, cement-based materials are submitted to carbonation. In cementitious structures, carbonation is considered a deterioration mechanism, because it decreases pH of a concrete. Thus, it allows concrete susceptibility to corrosion [3].

However, in non-structural concrete, carbonation can uptake CO_2 from the atmosphere, to make up for partially emissions generated by cement production [4–6]. This compensation can be considered during the balance concerning greenhouse gases emissions, associated with concrete structures. In order to ensure this ability, further researches are required, which led to the development of this study.

2 CO_2 uptake due to concrete carbonation

Carbonation is a result of CO_2 chemical reactions with alkaline products of cement hydration, in order to form calcium carbonate ($CaCO_3$) and water. This reaction ($Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$) reduces concrete pH, so, steel becomes susceptible to corrosion. On the other hand, the same reaction uptakes CO_2, since carbon dioxide that enters through concrete pore solution reacts with calcium hydroxide ($Ca(OH)_2$) to produce $CaCO_3$ in an

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opposite process to cement production ($CaCO_3 \rightarrow CaO + CO_2$). Carbonation reaction, and consequently CO_2 uptake, occurs throughout concrete structure lifetime. And according to Pade and Guimarães [7], it continues after demolition, where the process can be strengthened.

However, there is some disagreement defended by some authors regarding the potential of carbon sequestration/uptake ratio due to concrete carbonation (Table 1). This kind of research could be observed in a study carried out in Norway. Jacobsen and Jahren [8] estimated that 16 % CO_2 emissions in cement production are reabsorbed by lifetime of concrete and carbonation process.

Gajda [14] reports that 7.6 % of emitted CO_2 can be absorbed, but Pade and Guimarães [7] evaluate, in a study carried out in Denmark, an approach of 100 years and took into account a building demolishing, that concrete can absorb up to 57 % CO_2 emissions in cement production due to carbonation. But, if building demolishing is not considered, this value is reduced to 24 %.

The disagreement in literature is mainly due to the factors that have influenced concrete carbonation (as compressive strength, exposure zone, amount of cement, age structure, among others) and are dependent upon the methodology used by researchers. Then, each structure requires specific studies for these estimates thus, other studies should be carried out to confirm that CO_2 sequestration could be considered a compensatory measure due to concrete carbonation.

3 Mathematical modeling of concrete carbonation

Fib [15] stated that the durability is essentially a retrospective view of the structure performance. The expectation that a structure may be durable or not can only be evaluated through models that represent the deterioration processes, thus for the project assurance it is required the use of lifetime preview methods.

In order to understand and to model the deterioration mechanisms and their kinetic and action in the concrete structures, countless studies have emerged in the technical and scientific community. As a result, many models to estimate carbonation depth and lifetime preview were established in recent years. These models enabled considerable advances for understanding the behavior of the exposed concrete structures over time. With the aim of estimate the carbonation depth, Possan [16] compiled 39 models, which in general were based on Tuutii [17] model, that propound two periods to degradation process (initiation and propagation), separately modeled in most cases.

As the carbonation modeling can be physicochemical, numerical, analytical, or combined [18, 19] it results in empirical mathematical models [16, 20] or complexes models [19, 21]. The empirical ones, also named as simplified models, are based on adjustments of field or laboratory data or expert's experiences, and usually have analytical solution. The complexes or phenomenological models are based on physicochemical laws, as the conservation of mass law for water and carbon dioxide. Given the complexity of these models, numerical solutions are required.

Commonly, empirical models are simpler to apply, however they may generate less accurate results due to simplifications. On the other hand, the complexes models tend to provide a major precision and generalization for the degradation process, due to the large number of considered influence variables, but they are harder to apply [16].

When it refers to concrete carbonation, both modeling segments have limitations. Empirical modeling do not conduct to a deep understanding of the involved degradation mechanisms, difficulting extrapolation and generalization, because normally the model database is limited.

Table 1 CO_2 uptake due to concrete carbonation in different studies

Local	References	Scenario (type, structure characteristics, time)	CO_2 uptake	
			Lifetime/age	Demolition
Global	[4]	Concrete structures (100 years of lifetime)	33–57 %	–
Denmark	[7]	70 years of lifetime + 30 years after demolition	24 %	57 %
Norway	[8]	Concrete structures (age structure—20 years)	11 %	–
Nordic countries	[10]	Unprotected structure, exposed to rain (50 years of service life)	47 kg CaO	–
Sweden	[11]	Internal environment, sheltered from rain (100 years of lifetime)	6.1 kgCO$_2$/m^2	–
	[11]	Unprotected structure, exposed to rain (100 years of lifetime)	0.5 kgCO$_2$/m^2	–
Brazil	[6]	Itaipu Dam (old structure—34 years)	13.384 Tons	–
	[12]	Residues from construction and demolition	–	1 kg CO$_2$/1000 kg RCD
USA	[13]	Concrete pavements	28.2 %	–

Fonte: Possan et al. [14]

Besides, this models (as "k.$\sqrt{t}$" [17], where "k" is the carbonation coefficient and "t" is the structure exposure time) don't take into consideration all the main variables that influence the phenomenon, not describing it as the reality. Simulations with a model of this nature can conduct an error at the estimative. On the contrary, the complexes models consider a larger number of input variables, describing better the studied phenomenon, despite the difficult simulations. Many of the complexes models show few practical applications, since most of the input parameters are difficult to measure. In general, these models demand application of performance tests to specific material properties, as diffusion coefficient of CO_2, $Ca(OH)_2$ content, and others [16].

The Fig. 1 compares some literature models with similar characteristics [17, 20, 22–24]. These compared models contain simpler input data, as compressive strength, air humidity, among others.

It's remarkable that the analyzed models predict distinct carbonation depths. The proposed models by EHE [24] and Smolczyk [22] are the ones that predict bigger carbonation depths, followed by Langerblad [10], Tuutti [17] and Vesikari [23]. These models have a few input variables (in general just one) and do not consider many factors related to the carbonation process. On the other hand, the more complexes formulations such as proposed by Possan [16] and CEB 238 [20] include a larger number of input variables, enabling a better description of the the degradation process by carbonation. It's notable that the estimated carbonation depths generated through these models are very close, which can be related with the similar number of variables used in both.

Therefore, it was decided to apply in this study the Possan [16] model, which has input data easily obtainable [25], as concrete compressive strength, cement type, type and content of pozzolanic additions employed, CO_2 content of the exposure environment, air humidity and structure environment exposure (if indoors or outdoors, protect or not from the rain).
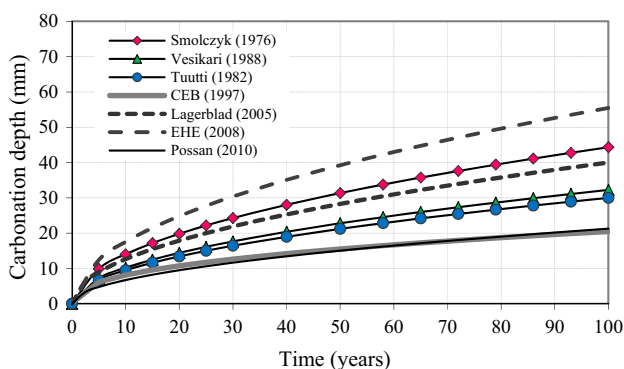

Fig. 1 Comparison between estimative models of concrete carbonation depth [16]

4 Estimation of CO_2 uptake due to concrete carbonation

In order to calculate CO_2 uptake rate during lifetime cycle of structures produced with different types of concrete, for a period from 0 to 100 years, this study was divided into six stages:

- *Step 1*: Selection of concrete;
- *Step 2*: Estimation of CO_2 emissions resulting from cement manufacturing process, aggregates and concrete;
- *Step 3*: Estimation of carbonation depth;
- *Step 4*: Calculation of CO_2 uptake by concrete structures during its lifetime period (0–70 years);
- *Step 5*: Calculation of CO_2 uptake by concrete structures after its demolition (70–100 years);
- *Step 6*: CO_2 balance during lifetime cycle of concrete structures.

4.1 Step 1: selection of concrete

In order to obtain the simulations, it was considered that the structure will be built in an urban area with an average relative humidity of 65 and 0.038 % CO_2 content. Table 2 shows the concrete mixing ratio used in the simulations, measured by the ABCP/ACI method. A basaltic coarse aggregate is employed whose maximum diameter size is 19 mm, Specific mass of 2700 kg/m³ and unit weight of 1515 kg/m³. Some natural quartz sand was applied as fine aggregate with fineness 2.9 modulus, specific mass of 2660 kg/m³ and unit weight of 1490 kg/m³.

4.2 Step 2: estimation of CO_2 emissions resulting from the cement manufacturing process, aggregates and concrete

Carbon dioxide emissions from concrete production were estimated up from the cement manufacturing process until its availability at construction area, whether it is from batching plant or produced during the construction, including emissions from aggregates production ABNT 11578 [26]. Based on methodologies from IPCC [27] and CSI [28], emissions by energy use (E_{ene}), raw material decomposition (decarbonizing) (E_{desc}) and by transportation of cement manufacturing process (E_{trans}) were considered to estimate CO_2 emissions relative to cement manufacturing process (E_{cim}), using Eq. 1.

$$E_{cim} = E_{ene} + E_{desc} + E_{trans} \, (\text{kgCO}_2/\text{t}) \qquad (1)$$

For E_{ene}, E_{desc} e E_{trans} calculations, it was used the methodology applied by Lima [29]. So, in order to estimate emissions from aggregates production, indicators found out

Table 2 Mixtures employed in the simulations

Type of cement	Additions		Compressive strength (MPa)[b]	Dosage (kg/m³)				Simulated scenarios
	Type[a]	Content[a]		Cement	Crushed Stone	Sand	Water	
CP III	Slag	35–70	20	370.90	1112.60	741.80	185.00	1
			30	385.20	1155.50	770.30	154.00	2
			40	392.70	1178.20	785.50	137.00	3
CP IV	Fly ash	15–50	20	352.00	1055.90	703.90	211.00	4
			30	371.60	1114.80	743.20	167.00	5
			40	393.50	1180.60	787.10	118.00	6
CP V	Limestone filler	0–5	20	364.80	1094.40	729.60	182.00	7
			30	378.60	1135.90	757.30	151.00	8
			40	392.00	1175.90	784.00	122.00	9

[a] Kinds and levels in Standard ABNT 11578 [26]

[b] The compressive strength refers to 28 days

in specific literature the use of 5.81 kgCO₂/t for natural sand, and 15.46 kgCO₂/t for crushed stone (MARCOS) [30]. Also, in order to estimate CO_2 emissions by transportation, 8.4 kgCO₂/t were used LIMA [29]. The energy data were collected from the National Energy Balance—NEB [31]. All calculations and estimations were performed in Excel spreadsheets.

4.3 Step 3: estimation of carbonation depth

Using the Excel® software, carbonation depth estimation of concrete through time was carried out with the degradation model proposed by Possan [16] (see Eq. 2), selected because it has readily available input variables (compressive strength, kind of cement, CO_2 content in the environment) and good representativeness of damage (it represents almost 85 % of tested cases).

$$y(t) = k_c . \left(\frac{20}{f_c}\right)^{k_{fc}} . \left(\frac{t}{20}\right)^{\frac{1}{2}} . exp\left[\left(\frac{k_{ad} . ad^{\frac{3}{2}}}{40 + f_c}\right) + \left(\frac{k_{CO_2} . CO_2^{\frac{1}{2}}}{60 + f_c}\right)\right. \\ \left. - \left(\frac{k_{RU} . (UR - 0.58)^2}{100 + f_c}\right)\right] . k_{ce} \qquad (2)$$

where ($y(t)$) is the average of carbonation depth (mm); (f_c) is the characteristic of compressive strength in concrete (in MPa); (k_c) is the factor according to the kind of cement (Table 3a); (k_{fc}) is the factor relating to compressive strength of concrete, depending on which cement is used (Table 3a); (t) is the concrete age (year); (ad) is the percentage of pozzolanic in concrete (relative to cement mass); (k_{ad}) is the factor to pozzolanic additions of concrete–silica fume, metakaolin and rice husk ash, according to the used cement (Table 3a); (RH) is the average of relative humidity (in % * 0.01) (k_{rh}) is the factor related to

relative humidity, according to cement used (Table 3a); (CO_2) is the atmospheric CO_2 concentration, (in %); (K_{CO2}) is the factor related to the environment CO_2 content, according to the cement used (Table 3a); and (K_{CE}) is related to exposure to rain factor, according to exposure conditions of the structure (Table 3b).

4.4 Step 4: calculation of CO₂ uptake by concrete structures during its lifetime (0–70 years)

Calculations were carried out concerning carbonation by concrete on a structural element, shown in Fig. 2, to estimate the amount of CO_2 uptake during the structures' lifetime (70 years) for different scenarios (Table 1). Based on stoichiometric calculations, CO_2 uptake was estimated due to concrete carbonation by Eq. 3.

Where (y) is carbonation depth of concrete (in meters); (c) is the amount of used cement to produce one m³ of concrete (without considering additions); (CaO) is the amount of calcium oxide contained in cement; (r) is CaO ratio that is totally carbonated; in other words, it is transformed into $CaCO_3$; (A) is the surface area of concrete exposed to CO_2 action (m²) and (M) is molar fraction of CO_2/CaO.

$$CO_2 = y.c.Cao.r.A.M \; CO_2 (kg/m^3) \qquad (3)$$

4.5 Step 5: calculation of CO₂ uptake by concrete structures after its demolition (70–100 years)

Estimate of CO_2 uptake amount by concrete during post-demolition period was obtained by simulations of carbonation that were carried out from demolishing structure residues. So, the carbonated layer was removed over the

Table 3 Coefficients of the model (a) according to concrete properties and environmental conditions and (b) exposure conditions [16]

(a) Type of cement	Properties of concrete			Environmental conditions	
	Cement k_c	f_c k_{fc}	Addition k_{ad}	CO_2 k_{co_2}	RH k_{RH}
CP I	19.80	1.70	0.24	18.00	1300
CP II E	22.48	1.50	0.32	15.50	1300
CP II F	21.68	1.50	0.24	18.00	1100
CP II Z	23.66	1.50	0.32	15.50	1300
CP III	30.50	1.70	0.32	15.50	1300
CP IV	33.27	1.70	0.32	15.50	1000
CP V ARI	19.80	1.70	0.24	18.00	1300

(b) Exposure conditions of structures	
Sheltered from rain	k_{ce}
Indoor sheltered from rain	1.30
Outdoor, sheltered from rain	1.00
Outdoor, exposed to rain	0.65

Fig. 2 Concrete structure

Fig. 3 Concrete elements after demolition
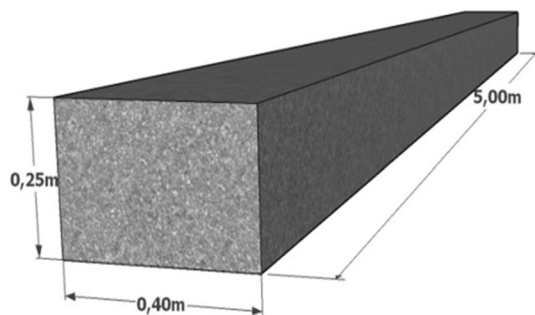
lifetime and became "non-carbonated concrete" in 30 × 30 × 30 mm cubes (Fig. 3), considering that they would be exposed to CO_2 for a period of 30 years. Therefore, the following steps were employed to estimate CO_2 uptake rate by concrete after demolition:

(a) Estimation of carbonation depth at the end of lifetime of a building structure (70 years);
(b) Volume estimation of carbonated and non-carbonated concrete during its lifetime;
(c) Definition of approximate dimensions of concrete elements after demolition (Fig. 3);
(d) Calculation of the new total surface area of concrete elements after demolition, according to Eq. 4;
(e) Use of Eq. 3 with the new surface area to determine CO_2 (kg/m^3) uptake amount after demolition.

$$A_{st} = V_{nc} \cdot \left(\frac{A_{sc}}{V_c} \right) \ (m^2) \tag{4}$$

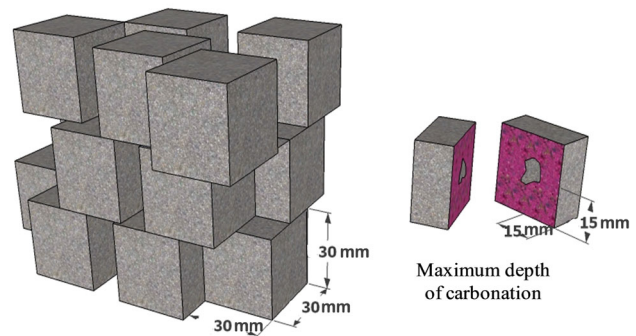where (A_{st}) is the total surface area that is in contact with the atmosphere (m^2); (A_{sc}) is the concrete surface area (parallelepiped) (m^2); (V_c) is concrete volume (parallelepiped) (m^3) and (V_{nc}) is the concrete volume that has not occurred carbonation (m^3) yet.

4.6 Step 6: CO_2 balance during the life cycle of concrete structures

Equation 5 was used to estimate CO_2 balance during the life cycle of concrete structures. The computed value refers to the amount of CO_2 that has been emitted to manufacture each concrete (scenarios 1 to 9—Table 1) subtracted from the uptake amount along the life cycle due to carbonation, according to Eq. 5.

$$B_{CO_2} = E_{con} - C_{cap} CO_2 (kg) \tag{5}$$

where (B_{co_2}) is CO_2 balance, (E_{con}) is CO_2 amount emitted during concrete manufacture process and (C_{cap}) is an amount of CO_2 uptake by concrete.

5 Results

Table 4 shows carbonation depth of the structures over time, calculated by Eq. 2.

In some cases, during post-demolition period, concrete was fully carbonated before concluding the estimated life cycle of 100 years. This fact is due to shape and size of residue particles used in simulations (cube sides are equal to 30 mm). Whereas CO_2 can enter through all the faces, maximum carbonation depth of each particle is 15 mm (see details in Fig. 3)

As it is expected (see Table 3), the increase of compression strength of concrete reduces carbonation depth [16, 32], so, there is some reduction on the amount of carbon absorbed from the atmosphere (see Figs. 4, 6, 8). According to Kumar et al. [33], concretes with higher compressive strength have lower porosity due to the influence of water/cement ratio, and this makes CO_2 difficult to be captured.

There is also some influence concerning cement during concrete carbonation. Concretes produced with cements without (CP V) additions tend to have higher compressive strength to carbonation than with (CP III and CP I) additions. This is because advance of carbonation is inversely proportional to alkalinity reserve, available in the matrix of hydrated cement paste: a role of cement chemical composition [34], influenced by its kind and content additions.

Within limits set by NBR 11578 [26] in Brazil, CP V AR I cement has a maximum of 5 % carbonate additions (Limestone) while CP III and CP IV have, respectively, 35–70 % of slag blast furnace additions and 15–50 % of fly ash contents, which alter chemical composition of cement, with direct influence on concrete carbonation as well as CO_2 uptake.

Figures 4, 6 and 8 show CO_2 (kg/m³) values, absorbed from the atmosphere, by concrete structures during its lifetime (0 to 70 years) and post-demolition period (70–100 years) according to strength contents. While Figs. 5, 7 and 9 show the amount of CO_2 emitted to produce concrete and register the uptake amount due to carbonation during life cycle depending on the cement (according to Eqs. 3, 4).

It should be pointed out that at 20 MPa compressive strength of concrete, CO_2 potential and uptake due to carbonation is on average 60 % higher than the uptake one during the lifetime period, if post-demolition is considered. It is shown the importance of considering structure demolition in CO_2 balance. Similar results are observed for other concretes. This fact is due to the greater surface area of exposed concrete to CO_2 action, which increases carbonated area, and therefore carbonation reaction of CO_2 uptake. The considered structure according to estimates of lifetime has 6.5 m² surface area, while during post-demolition period, an average surface area estimated of waste generated is approximately 64.47 ± 1 m² (the area is variable in post-demolition period due to carbonated layer deduction during its lifetime, which varies with strength and cement used).

There can be seen in Figs. 5, 7 and 9 CO_2 emissions as a result from concrete manufacturing process, where the positive quadrant was related to the produced emissions and the negative quadrant concerned about CO_2 uptake due to concrete carbonation during the structure life cycle

Table 4 Carbonation depth over time, in mm, depending on cement and compressive strength of concrete

Type of cement	f_c (MPa)	Carbonation depth (mm)													
		Structure (As = 6.5 m²)								Waste (post-demolition)[a] (As = 64 ± 1 m²)					
		5	10	20	30	40	50	60	70	75	80	85	90	95	100
CP III	20	14.9	21.1	29.9	36.6	42.3	47.3	51.8	55.9	14.9	[b]	[b]	[b]	[b]	[b]
	30	7.4	10.5	14.9	18.3	21.1	23.6	25.9	28.0	7.4	10.5	12.9	14.9	Carb.	Carb.
	40	4.5	6.4	9.1	11.2	12.9	14.4	15.8	17.1	4.5	6.4	7.9	9.1	10.2	11.2
CP IV	20	16.9	23.9	33.8	41.4	47.8	53.5	58.6	63.3	13.1	[b]	[b]	[b]	[b]	[b]
	30	8.2	11.6	16.5	20.2	23.3	26.1	28.5	30.8	8.2	11.6	14.2	[b]	[b]	[b]
	40	5.0	7.1	10.1	12.4	14.3	16.0	17.6	19.0	4.7	6.7	8.3	9.5	10.7	11.7
CP V	20	9.9	14.0	19.8	24.3	28.0	31.4	34.3	37.1	9.8	13.9	[b]	[b]	[b]	[b]
	30	4.9	7.0	9.9	12.1	14.0	15.6	17.1	18.5	4.9	7.0	8.5	9.9	11.0	12.1
	40	3.0	4.2	6.0	7.4	8.5	9.5	10.4	11.3	3.0	4.2	5.2	6.0	6.7	7.4

(fc) compressive axial strength of concrete (MPa), (As) surface area of concrete exposed to CO_2 action (m²)

[a] Refers to the age of the structure. Carbonation depth during simulations is based on waste production. Therefore, the times of 75, 80, 85, 90, 95 and 100 years correspond to 5, 10, 15, 20, 25 and 30 years of residue exposure to CO_2 action

[b] 100 % carbonated aggregate particles (carbonation depth higher than or equal to 15 mm)
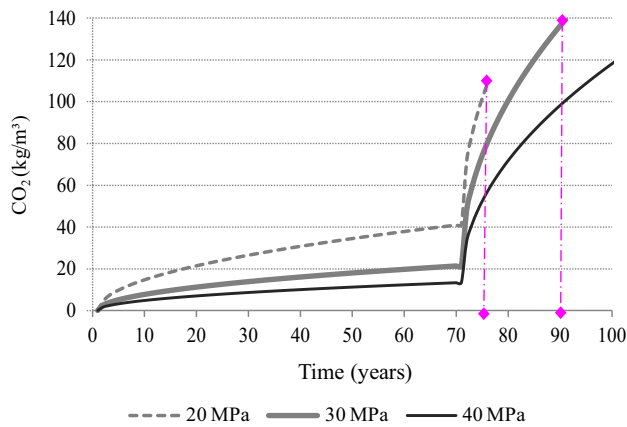
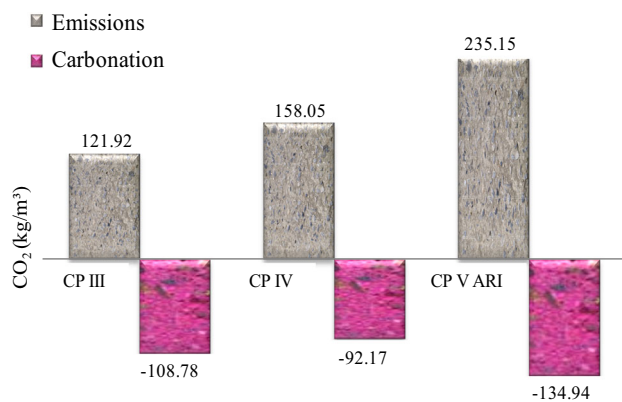**Fig. 4** Evolution of CO_2 uptake over time in concrete with cement CP III

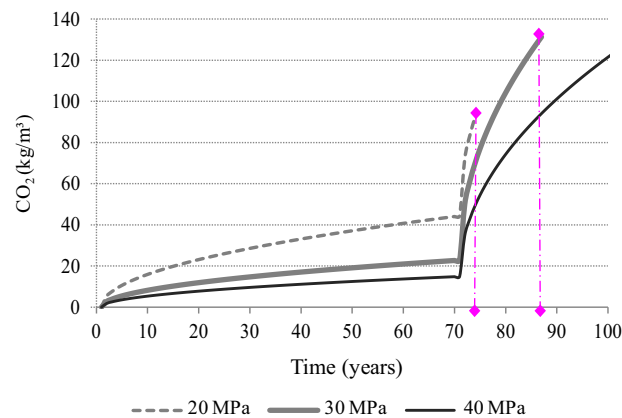**Fig. 5** Quantities of CO_2 emitted and carbonated in concretes—20 MPa

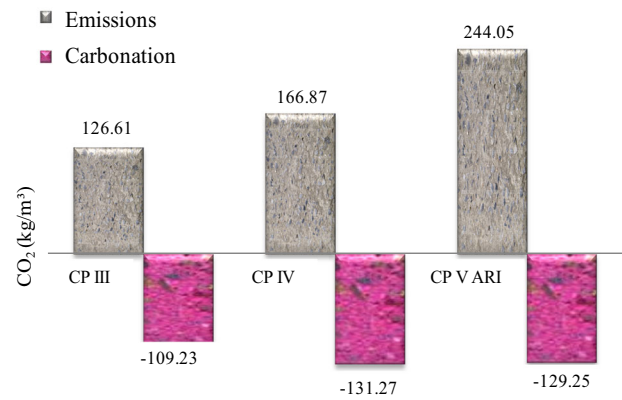**Fig. 6** Evolution of CO_2 uptake over time in concrete—cement CP IV

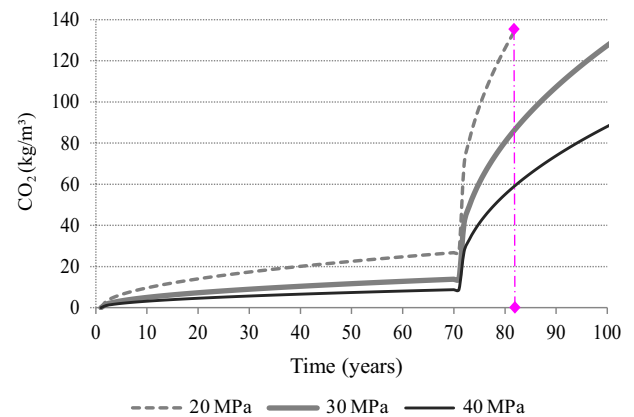**Fig. 7** Quantities of CO_2 emitted and carbonated in concrete—30 MPa

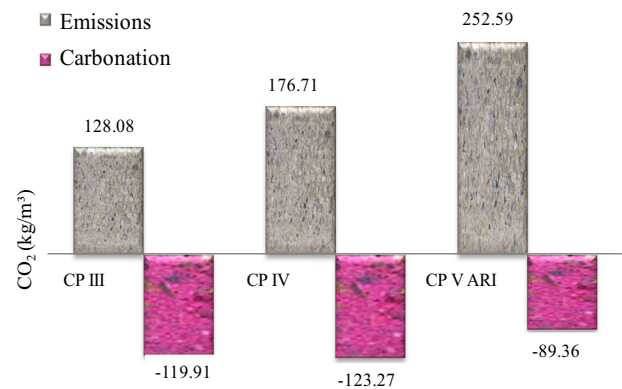**Fig. 8** Evolution of CO_2 uptake over time in concrete—cement CP V ARI

Fig. 9 Quantities of CO_2 emitted and carbonated in concrete—40 MPa

(lifetime and post-demolition). It was also considered the kind of cement and its compressive strength. It seems that concrete during its lifetime can uptake from 40 to 90 % CO_2 emitted in its manufacturing process. In some cases, considering the structure demolition, its uptake is nearly 100 %. An example of this is a concrete made from cement CP III, where the emissions of concrete manufacture are 128 kgCO_2/t while its uptake is 120 kgCO_2/t (see Fig. 9).

Fig. 10 Balance of CO_2 due to concrete carbonation
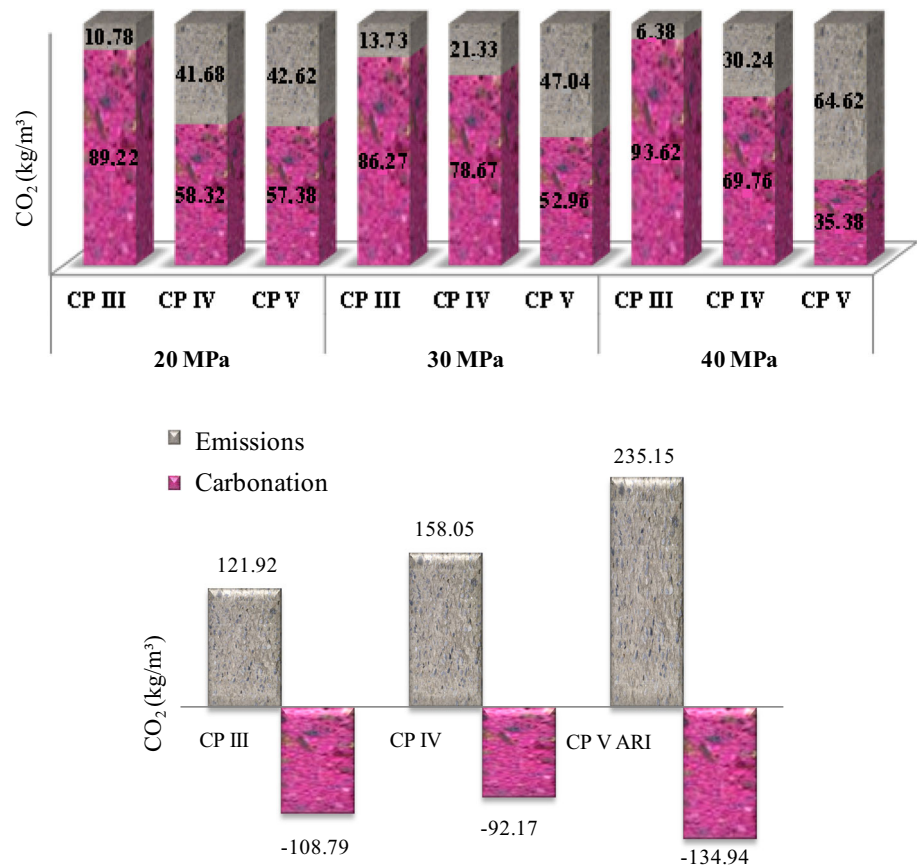
According to Eq. 5, in Fig. 10, CO_2 balance is shown in nine scenarios of study. It is presented carbon uptake percentage by concrete throughout its service life (70 years) and during the post-demolition period (up to 30 years), as well as CO_2 percentage that remains in the atmosphere, in other words, which has not been captured yet.

It has been observed that concrete produced with cement CP IV ARI has shown a balance of the least favorable emissions, although there was a better performance concerning durability, since it has greater compressive strength to carbonation. The best results for CO_2 balance in concrete produced with cement with additions (CP III and CP IV) are assigned to the cement manufacturing process, since part of the clinker is replaced by additions. Consequently, there were reductions on emissions per ton of produced cement. It was also due to alkalinity reduction in concrete with additions, which results in faster carbonation and, therefore, the largest CO_2 uptake.

It should be highlighted that concrete carbonation during post-demolition is highly dependent on exposure conditions and dimensions of demolished material, so, specific research is required to evaluate, in real scale, both form and time of waste exposure to CO_2 action.

6 Conclusions

According to the mathematical modeling, it was found out that concrete has potential regarding CO_2 uptake due to carbonation reaction. And, during the post-demolition period, its potential can be increased up to five times. Besides the factors that affect concrete carbonation (compressive strength, kind of cement, etc.), the surface area of a structure exposed to carbon dioxide action influences CO_2 uptake. The larger is the area in contact with CO_2, the largest is potential uptake.

CO_2 uptake, which is a result from concrete, has been important for industrial construction. Since this process is known, it is possible to indicate concrete by its performance, aiming at durability, with a minimum use of Portland cement, that can also CO_2 uptake from atmosphere during the structure lifetime. This denotes that the balance of emissions can be considered in the future, a compensatory measure in design of concrete structures.

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