50 YEARS OF MATERIALS AND STRUCTURES

Sustainability-based decision support framework for choosing concrete mixture proportions

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Abstract A framework is proposed, along with two objective indices, for the selection of concrete mixture proportions based on sustainability criteria. The indices combine energy demand and long-term strength as energy intensity, and carbon emissions and durability parameters as A-indices, which represent the apathy toward these essential features of sustainability. The decision support framework is demonstrated by considering a set of 30 concretes with different binders, including ordinary portland cement (OPC), fly ash, slag and limestone calcined clay cement (LC3). In addition to the experimental data on compressive strength, chloride diffusion and carbonation, life cycle assessment has been performed for the concretes considering typical situations in South India. The most sustainable of the concretes studied here, for service life limited by chloride ingress, are those with LC3, OPC replaced by 50% slag, and ternary blends with 20% each of slag and fly ash. In the case of applications where carbonation is critical, the appropriate concretes are those with OPC replaced by 15-30% slag or 15% fly ash, or with ternary blends having 20% slag and 20% Class F fly ash.

 $Carbonation \cdot Life \ cycle \ assessment \cdot Supplementary \\ cementitious \ materials \cdot Service \ life$

1 Introduction

Researchers, such as Mehta [1] have advocated since the 1990s, if not earlier, that concrete structures be designed and proportioned considering durability and environmental friendliness, in addition to the usual criteria of cost and strength. The insistence on the sustainability of construction has become exceedingly relevant considering that global process emissions related to cement production are at about 1.5 Gt-CO₂ per year [2], in addition to the growing requirement for limestone, water, aggregates and energy. If compelling sustainability measures are not adopted, all trends indicate that the environmental impact could continue to increase alarmingly, as emerging countries like India are still far from the peak per capita requirement for cement and concrete. Possible measures include the minimization of the raw materials used in concrete, substitution of the clinker, lowering the energy consumption and, most importantly, increasing the durability or service life [3]. Habert and Roussel [4] predict that CO₂ emissions associated with concrete can be reduced by up to 40% by using higher strength concretes and making appropriate substitutions for the clinker in cement. Miller et al. [5] anticipate a potential decrease in CO2 emissions of





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24% through straightforward changes in mixture proportions along with the integration of later-age strength gain in design.

In the context of the above arguments, it is necessary to facilitate the choice of concretes having the most appropriate composition by providing a framework based on objective sustainability considerations that can be effected through tender conditions and construction contracts. There are four essential features for such sustainability assessment [4-10]: (1) satisfying the requirements for mechanical performance, which is essential from the view of safety and serviceability; (2) economic viability, which is critical from the implementation standpoint, typical of most civil engineering applications; (3) durability, to ensure that the material used provides for the desired longevity of the structural system; and (4) minimum environmental impact, e.g., in terms of the carbon footprint and embodied energy.

The present work proposes a decision support framework based on indices that account for CO₂ emissions and energy consumed in the production of cement and concrete, along with compressive strength and durability parameters. The approach is illustrated through the assessment of various concretes having ordinary portland cement (OPC) blended with fly ash and slag, as well as a new limestone calcined clay cement, LC3. The most sustainable of the mixes studied for applications prone to chloride exposure and carbonation are identified through this procedure. Life cycle assessment (LCA) is performed [11, 12] for obtaining the parameters related to the environmental impact for a case study situated in India.

2 Sustainability indices and assessment framework

2.1 Review of sustainability indices

Many parameters have been proposed in the literature to represent the sustainability of concrete. They are mostly based on the carbon emissions during the production processes. Beyond the estimation of such quantities for functional units of volume or mass from LCA, several indices have been formulated incorporating performance metrics, mainly considering strength or load-carrying capacity and durability, to be more relevant for sustainability assessment [9].



The sustainability indices can be roughly classified into four types. One type of indices gives the impact, such as carbon emissions or energy consumed per unit compressive strength, mostly at 28 days [9, 10, 13–16]. By considering the strength as the functional unit, such indices combine the mechanical performance requirement with the environmental impact. The second type incorporates parameters representative of environmental impact, strength and durability, such as the complex functional unit formulated by Panesar et al. [9]. The third type combines an environmental impact parameter, strength and service life [16], such as in the sustainability potential of Müller et al. [7]. The application of the second and third types of indices requires the assessment of a range of concretes to facilitate the decision making as the physical significance of the index is not evident due to the combination of three or more distinct variables. The fourth type considers the environmental impact of the entire structure or a component designed for a certain set of loads [17], where the mechanical performance is taken as the functional unit. This can be extended further to include durability requirements in the design, as proposed by Muigai et al. [8]. Though this would be the ideal and comprehensive manner to choose the optimum concrete proportions, practical difficulties arise since the structural design has to be completed for each concrete before the decision can be made. Moreover, results of structure-level assessment would vary with the life estimation model, design strategy, codes of practice, exposure condition and so on.

2.2 Proposed decision support framework

Sustainability assessment of concrete should evidently include parameters related to cost, mechanical integrity, carbon footprint and durability. It is proposed here that two indices be used together to provide the decision support framework [14] for the choice of concrete composition. It is envisaged that the indices are determined, and even catalogued, by the concrete supplier so that the designer or specifier can select the most appropriate mixture proportions in terms of sustainability. The proposed indices, described in the following, are such that their minimization would lead to the most sustainable concrete, among the options provided.

2.2.1 Energy intensity

The first index used here is called the energy intensity $(ei_{cs,365})$, defined as the energy consumed in the production of unit volume of concrete, say 1 m³, divided by the compressive strength at 365 days or one year [14]. The basis for this is that the energy demand has significant cost and environmental implications, as well as reflecting the efficiency of the production processes, transportation distances, binder intensity and raw material usage. The functional unit of the index is taken here as the compressive strength at one year in light of the facts that most blended binders provide for the increase in strength much beyond the usual 28-day value, which is mostly ignored in conventional material selection and design, and that the long-term strength is more relevant for the mechanical integrity and performance of the structure during its service life. It has been shown that emissions could reduce by about 10% just by basing structural design on parameters defined at ages later than 28 days [5, 10].

2.2.2 A-indices

The second index in the framework aims to combine the carbon footprint and durability of concrete, which are commonly neglected in structural design, and is denoted here as the apathy index [14] or A-index. It is defined as the total carbon emissions attributed to a unit volume of concrete, say 1 m³, divided by a material durability parameter. A higher value of the A-index for a particular concrete composition would indicate the "apathy" towards sustainability, if that material were to be used instead of another with a lower A-index. The durability parameter for this index is expected to be one that can be correlated to service life, as an extension of previous approaches along these lines [7-9, 16]. Nevertheless, the parameter should be based on material characteristics and not require the structural design to be available, with the concomitant limitations.

Considering that the durability of reinforced concrete structures is often limited by the corrosion of the steel reinforcement caused by the ingress of chlorides, water, oxygen, carbon dioxide, etc., the most appropriate durability parameter would be one based on a diffusion or migration coefficient. Since chloride attack is a predominant cause for such loss of structural service life, durability is mostly represented by a material parameter related to chloride ingress [9, 14]. Also, carbonation of the cover concrete could lead to corrosion of the reinforcement; though this is not a predominant cause of structural degradation, it is of growing concern due to the vulnerability of blended binders in this regard. Therefore, two parameters have been proposed here to represent the durability of concrete exposed to chlorides and carbonation, and denoted, respectively, as the chloride resistance factor F_{chlor} and carbonation resistance F_{carb} :

$$F_{\rm chlor} = \exp\left(10^{-6}/\sqrt{D_{\rm cl}}\right) \tag{1}$$

$$F_{\rm carb} = \left(\frac{5}{k_{\rm CO2,nat}}\right)^2 \tag{2}$$

where D_{cl} is the coefficient of chloride diffusion, in m²/s, and $k_{CO2, nat}$ is the carbonation coefficient or rate, in mm/ \sqrt{y} ear. The parameters defined by Eqs. (1) and (2) are related to the rate of progress of the chloride and carbonation fronts within the concrete, respectively, and consequently reflect the corresponding service lives until the steel reinforcement begins to corrode. The numerators in the equations have been chosen to give values in the order of unity for the corresponding parameters.

Based on the resistance factors, the A-index is defined for chloride exposure and carbonation, respectively, as:

$$Ai_{\rm chlor} = \frac{\rm CO_2 \, emissions \, per \ cubic \, metre \ of \ concrete}{F_{\rm chlor}}$$
(3)

$$Ai_{\rm carb} = \frac{\rm CO_2 \, emissions \, per \, cubic \, metre \, of \, \, concrete}{F_{\rm carb}}$$
(4)

The concept of the A-index can be extended to other types of degradation and phenomena that could limit the service life of a structure.

3 Experimental study

3.1 Materials and mixture proportions used in the study

In order to demonstrate the proposed sustainability assessment framework, experimental data has been collected for a set of 30 concretes, ranging in 28-day characteristic cube compressive strength from 20 to 60 MPa, covering the grades commonly used in most applications. The binders include combinations of ordinary portland cement (OPC 53 Grade, i.e., Indian standard cement with 28-day mortar compressive strength of at least 53 MPa) with supplementary cementitious materials (SCMs), such as ground granulated blast furnace slag (GGBS; Slags A and B sourced from Goa and Nandyal, respectively), Class C fly ash (from the Neyveli thermal power plant) and Class F fly ash (from the North Chennai thermal power plant), as well as limestone calcined clay cement, LC3 (with 50% clinker, 30% calcined clay, 15% limestone and 5% gypsum) from a pilot industrial-scale production [18-20]. The fine aggregate used was river sand (5 mm down), and the coarse aggregate was crushed granite in two fractions (of 10 mm down and 20 mm down). The mixture proportions of the concretes are given in Table 1; the binders have 0, 15, 30, or 50% of either fly ash or slag, or are ternary blends with 40% of fly ash and slag; and the water/binder ratios (w/b) vary from 0.4 to 0.65. Note the notation for the concrete mixture "w/b-TBC- %SCM" indicates the w/b, total binder content (TBC) and the % of SCM in the binder (%SCM). A sulphonated naphthalene formaldehyde superplasticizer (SP) was used in all the concretes to obtain slump values between 80 and 150 mm. The mean compressive strengths at 28 days and 1 year are reported in Table 1.

3.2 Chloride diffusion coefficient

The sustainability assessment framework requires an estimate of the chloride diffusivity of the concrete (Eq. 1), as a representation of the ease with which chlorides can ingress and reach the steel reinforcement, causing it to eventually corrode. There are several methods in the literature and standards for the determination of the coefficient of chloride diffusion, and any one of those would be suitable for the purpose



of the assessment, as long as the same method and conditions are used across the study. Here, the rapid migration test (RMT) developed by Tang and Nilsson [21] has been used, as specified in the NT BUILD 492 [22] and ASTM C 1156 [23] standards. Three slices of 100 mm diameter and 50 mm thickness are cut from 300 mm long cylinders, which have been moist-cured for 90 days and later vacuum saturated with calcium hydroxide solution. Each slice is fitted tightly inside a PVC pipe and the edges are sealed with the help of a rubber ring, to form the anolyte chamber. The upper part of the pipe, above the slice, is filled with 0.3 M NaOH solution. Three such cells are placed, with a tilt, in a plastic container filled with 10% NaCl solution, which acts as the catholyte. The electrode immersed in the NaCl solution is connected to the negative terminal of the voltage source whereas the anode is connected to the positive terminal; nichrome meshes are used as the electrodes. Figure 1 shows the scheme of the test setup. To induce chloride migration, a 30 V potential gradient is initially applied across the slice, and subsequently adjusted based on the measured value of the initial current value, as specified in the standard.

After 24 h, each slice is split diametrically, and the depth of chloride penetration is measured after spraying the fractured surface with silver nitrate solution, which reacts with the chlorides to form a white precipitate of silver chloride. Further details of the testing and discussion of the data can be found elsewhere [20, 24]. The non-steady state migration coefficient D_{nssm} (in m²/s) is consequently obtained as:

$$D_{\rm nssm} = \frac{\rm RT}{zFE} \cdot \frac{X_d - \propto \sqrt{X_d}}{t} \tag{5}$$

where,

$$E = \frac{U-2}{L}$$
$$\propto = 2\sqrt{\frac{\text{RT}}{zFE}} \cdot erf^{-1} \left(1 - \frac{2c_d}{c_0}\right)$$

z = Absolute value of ion valence (for chloride, *z* = 1), *F* : Faraday constant = 9.648 × 10⁴ J/(V mol), *U* = Absolute value of the applied voltage (V), *R* : Gas constant = 8.314 J/(K mol), *T* = Average temperature in the anolyte solution (here, 300 K), *L* = Thickness of the specimen (0.1 m), X_d = Average penetration depth (m), *t* = Test duration (s), erf⁻¹

Table 1	Mixture proportion	s and p	properties of ve	urious concrete:	s used in	the study					
Mix ID	Notation (w/b-TBC- %SCM)	Quant	tity (kg/m ³)			Cube con strength (npressive MPa)	Non-steady state migration coefficient, $D_{\rm nsm}~(\times~10^{-11}~{\rm m^2/s})$	Chloride diffusion coefficient, D_{cl} (× 10^{-11} m ² /s)	Accelerated carbonation coefficient, <i>k</i> _{CO2,accl} (mm/, /vear)	Natural carbonation coefficient, k _{CO2,nat} (mm//year)
		Sand	$\underset{(\leq 10 \text{ mm})}{\text{CA}}$	$\mathop{\rm CA}_{(\leq 20 \text{ mm})}$	Water	28-day	1-year				
MI	0.65-280-0% SCM	760	499	749	182	30.0	34.3	2.36	2.67	16.0	4.0
M2	0.55-340-0% SCM	735	483	725	187	42.9	43.8	2.05	2.32	13.4	3.1
M3	0.50-310-0% SCM	759	498	748	155	43.8	48.7	1.93	2.18	10.2	3.2
M4	0.6-310-0% SCM	747	491	736	186	32.4	42.7	2.93	3.31	15.0	4.4 ^a
M5	0.6-310-15% FaC	747	491	736	186	31.7	41.1	2.04	2.31	16.9	5.1
M6	0.5-310-15% FaC	759	498	748	155	41.1	45.6	2.42	2.73	10.6	3.9
M7	0.55-340-15% FaC	735	483	725	187	41.3	45.1	1.18	1.33	16.6	3.8
M8	0.6-310-15% FaF	747	491	736	186	39.0	44.7	1.74	1.97	15.1	4.4
6M	0.55-340-15% FaF	735	483	725	187	40.0	49.4	1.43	1.62	15.0	5.2
M10	0.5-310-15% FaF	759	498	748	155	46.1	49.8	1.19	1.34	16.3	2.8
M11	0.6-310-15% SgA	747	491	736	186	36.1	43.9	1.60	1.81	14.8	4.6
M12	0.55-340-15% SgA	735	483	725	187	37.2	43.3	1.55	1.75	18.1	3.6
M13	0.5-310-15% SgA	759	498	748	155	44.7	40.8	1.31	1.48	11.9	2.7
M14	0.6-310-15% SgB	747	491	736	186	38.6	40.3	1.99	2.25	15.3	4.1
M15	0.55-340-15% SgB	735	483	725	187	44.5	52.9	9.64	1.09	15.4	3.6
M16	0.5-310-15% SgB	759	498	748	155	49.2	55.0	1.31	1.48	11.6	3.1
M17	0.5-310-30% FaC	759	498	748	155	37.9	46.8	1.08	1.22	17.7	5.4
M18	0.65-280-30% FaF	760	499	749	182	21.3	32.0	0.92	1.04	22.5	7.5
M19	0.5-310-30% FaF	759	498	748	155	35.2	40.9	0.60	0.68	17.3	5.6
M20	0.5-310-30% SgB	759	498	748	155	44.7	55.9	0.83	0.94	13.7	4.6
M21	0.65-280-30% SgB	760	499	749	182	28.7	36.0	0.78	0.89	19.3	5.6
M22	0.65-280-30% SgA	760	499	749	182	28.2	31.7	0.79	0.89	18.7	6.3
M23	0.5-310-(20% FaF + 20% FaC)	759	498	748	155	33.4	49.3	0.73	0.82	20.9	6.4
M24	0.5-310-(20% SgB + 20% FaC)	759	498	748	155	38.3	45.3	0.63	0.71	17.1	5.0
M25	0.5-310-(20% SgB + 20% FaF)	759	498	748	155	40.0	53.4	0.53	0.59	17.1	4.6
M26	0.5-310-50% FaF	759	498	748	155	23.2	33.1	0.41	0.46	24.4	8.4
M27	0.5-310-50% SgB	759	498	748	155	50.4	55.0	0.23	0.33	14.9	5.9
M28	0.5-310-LC3	708	491	736	155	44.9	48.0	0.11	0.13	n.a.	8.2

UI XIM	Notation (w/b-TBC- %SCM)	Quant	ity (kg/m ³)			strength ((MPa)	Non-steady state migration coefficient, $D_{\text{nssm}} (\times 10^{-11} \text{ m}^2/\text{s})$	Conoride diffusion coefficient, D_{cl} (× 10 ⁻¹¹ m ² /s)	Accelerated carbonation coefficient, $k_{CO2,accl}$	Natural carbonation coefficient, $k_{\rm CO2,nat}$ (mm/ \sqrt{y} ear)
		Sand	CA (≤ 10 mm)	CA ($\leq 20 \text{ mm}$)	Water	28-day	1-year			(mm/√year)	
M29	0.45-360-LC3	687	476	725	162	47.0	48.0	0.094	0.11	n.a.	7.1
M30	0.4-340-LC3	704	488	732	136	55.3	59.0	0.078	0.09	n.a.	6.6

'Natural carbonation coefficient estimated from the equation given in Fig. 3

Inverse of the error function, c_d = Chloride concentration at which silver nitrate solution changes colour (taken as 0.07 N), and c_0 = Chloride concentration in the catholyte solution (≈ 2 N). The coefficient of chloride diffusion, D_{cl} , can now be estimated from the $D_{\rm nssm}$ -value following the fib Bulletin 34 [25] procedure, as:

$$D_{\rm cl} = k_{\rm e} D_{\rm nssm} \tag{6}$$

 $k_{\rm e}$: Environmental where, transfer variable = exp $\left[4800\left[\frac{1}{T_{ref}} - \frac{1}{T_{real}}\right]\right]$, T_{ref} : Standard test temperature [K] (here, 300 K), and T_{real}: Ambient air temperature [K] (taken as 308 K).

Consequently, $D_{cl} = 1.13D_{nssm}$. The D_{cl} -values thus obtained are reported for all the concretes in Table 1. It can be seen that the lowest diffusion coefficients are found for the LC3 concretes (i.e., about 1×10^{-12} m²/s), and the concretes with 50% slag or fly ash or with 20% slag + 20% fly ash (i.e., 3– 8×10^{-12} m²/s), whereas the highest values are for concretes without any SCM (i.e., about $2-3 \times 10^{-11}$ m^{2}/s). Clearly, the concretes with lower D_{cl} -values can be expected to protect the reinforcement better under chloride attack.

Using the D_{cl} -value, the chloride resistance factor $F_{\rm chlor}$ defined in Eq. (1) can now be calculated for each concrete (see Table 2). In order to demonstrate the relevance of this factor, the D_{cl} -values have been used in the estimation of service life under chloride exposure (defined as the corrosion initiation time) for a reinforced concrete system having a cover depth (d) of 50 mm, chloride threshold for corrosion initiation (Cl_{th}) of 0.4% by weight of binder and surface chloride concentration of 0.8% by weight of binder, assuming a decay constant or ageing exponent (m) of 0.6 for the coefficient of diffusion [25]. The probabilistic service life was obtained using the solution of Fick's 2nd Law of Diffusion, in a MATLAB® programme SL-Chlor, considering d, D_{cl} and Cl_{th} as random variables, and a cumulative failure probability of 0.5. Figure 2 shows the relation between the estimated service life and F_{chlor} , which reflects the motivation for the use of the latter as a durability parameter of concrete in the proposed framework. The highest estimated service lives are for the LC3 concretes, followed by concretes with high SCM content, as expected.



Fig. 1 Setup for the rapid migration test

3.3 Carbonation coefficient

The carbonation coefficient of the concrete is required for the sustainability assessment corresponding to structures prone to corrosion of the steel reinforcement due to the carbonation of the cover concrete, just as it is necessary for the estimation of the service life of such conditions. It would be ideal to obtain the carbonation coefficient, $k_{CO2,nat}$, with the concrete exposed to the same environment as that of the structure. Since this would require several years of measurements, it is more practical to determine an accelerated carbonation coefficient, $k_{CO2,accl}$, from tests conducted in the laboratory under high CO2 concentrations. A suitable conversion factor to obtain $k_{\rm CO2,nat}$ from $k_{\rm CO2,accl}$ could be used, if available for the relevant environmental conditions; conversion factors reported in literature range from 0.07 to 0.5 [26-30], depending on the CO₂ concentrations in the accelerated and natural environments, and the climatic conditions.

In this study. concrete prisms of $100 \times 100 \times 500$ mm were cured for 28 days in a moist room followed by 14 days of air-curing at the temperature and relative humidity of 25 °C and 65% R.H., respectively, after which they were subjected to 1% CO₂ concentration in a chamber having the same conditions. Slices of 100 mm thickness were broken off perpendicular to the longitudinal axes after the exposure periods (t_{accl}) of 28, 56, 90 and 112 days of exposure, and the average carbonation depth on the fracture surface, $d_{CO2,accl}$, was measured (using phenolphthalein indicator). Further details of the accelerated carbonation tests are available elsewhere [24].

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The data is used to obtain the $k_{CO2,accl}$ -value by linear regression, following Tutti [31], as:

$$d_{\rm CO2,accl.} = k_{\rm CO2,accl.} \sqrt{t_{\rm accl}} \tag{7}$$

Table 1 gives the $k_{\text{CO2,accl}}$ -values of various concretes used in this study. The lowest accelerated carbonation coefficients (i.e., 10–12 mm/ $\sqrt{\text{year}}$) are found for the concretes without any SCM or with 15% slag and having low w/b and binder content, whereas the highest rates (i.e., $\geq 20 \text{ mm}/\sqrt{\text{year}}$) are for the concretes with high (i.e., $\geq 20\%$) fly ash dosages.

As mentioned earlier, it would be most appropriate to have the carbonation coefficients corresponding to the natural environment of the structure. For this purpose, the carbonation coefficients for the concretes considered here were determined in specimens maintained on a rooftop exposed to natural environment (i.e., unsheltered, exposed to direct rain and sun) in Chennai, over a period of up to 3 years [32]. The natural carbonation depth was measured at different ages, using the same methodology used in the accelerated carbonation tests, and the data were fitted to obtain the natural carbonation coefficient, $k_{CO2,nat}$, for each of the concretes. These values are also given in Table 1, where it can be seen that they range from 2.7 to 4 mm/ $\sqrt{\text{year}}$ for concretes without SCM or only 15% SCM content and having low w/b up to values greater than 6 mm/ $\sqrt{\text{year}}$ for concretes with higher $(\geq 30\%)$ SCM content or with LC3. As expected, concretes having binders with more carbonatable content exhibit slower progression of the carbonation front or rate, yielding lower carbonation coefficients. The relation between the natural and accelerated carbonation coefficients can be obtained, approximately, as (see Fig. 3):

$$k_{\rm CO2,nat} = 1.47 \sqrt{\frac{400 \,\mathrm{ppm}}{10000 \,\mathrm{ppm}}} k_{\rm CO2,accl}$$
 (8)

where, 400 and 10000 ppm are the CO_2 concentrations in the environment and accelerated conditions, respectively. The factor of 1.47 is similar to those obtained by previous researchers for other tropical climates [29, 30]. Such region or climate dependent factors could be used to convert short-term accelerated carbonation data to the natural carbonation coefficients.

Using the $k_{CO2,nat}$ -values, the carbonation resistance factor, F_{carb} is calculated for each concrete using



Tuble 2 Turameters and malees for sustainability assessment	Table 2	Parameters	and	indices	for	sustainability	assessment
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Mix ID	Notation (w/b-TBC- %SCM)	$\begin{array}{llllllllllllllllllllllllllllllllllll$		<i>ei</i> _{cs,365} (MJ/MPa) based on 1-year strength	Combined indicator footprint and durat	r for carbon bility
				e	Ai _{chlor}	Ai _{carb}
M1	0.65-280-0% SCM	1.21	1.56	87.8	284	221
M2	0.55-340-0% SCM	1.23	2.60	77.4	327	155
M3	0.50-310-0% SCM	1.24	2.44	66.0	302	153
M4	0.6-310-0% SCM	1.19	1.29	75.1	313	290
M5	0.6-310-15% FaC	1.23	0.96	70.9	267	342
M6	0.5-310-15% FaC	1.21	1.64	64.0	272	200
M7	0.55-340-15% FaC	1.32	1.73	68.2	268	204
M8	0.6-310-15% FaF	1.25	1.29	65.0	262	254
M9	0.55-340-15% FaF	1.28	0.92	62.0	274	381
M10	0.5-310-15% FaF	1.31	3.19	58.5	250	103
M11	0.6-310-15% SgA	1.27	1.18	68.9	266	284
M12	0.55-340-15% SgA	1.27	1.93	73.6	283	187
M13	0.5-310-15% SgA	1.30	3.43	74.2	259	98
M14	0.6-310-15% SgB	1.23	1.49	73.9	270	224
M15	0.55-340-15% SgB	1.35	1.93	59.4	264	186
M16	0.5-310-15% SgB	1.30	2.60	54.4	258	128
M17	0.5-310-30% FaC	1.33	0.86	56.2	214	332
M18	0.65-280-30% FaF	1.36	0.44	77.1	194	594
M19	0.5-310-30% FaF	1.47	0.80	63.8	193	356
M20	0.5-310-30% SgB	1.39	1.18	49.4	211	248
M21	0.65-280-30% SgB	1.40	0.80	72.4	194	341
M22	0.65-280-30% SgA	1.40	0.63	84.3	198	438
M23	0.5-310-(20% FaF + 20% FaC)	1.42	0.61	49.1	180	418
M24	0.5-310-(20% SgB + 20% FaC)	1.46	1.00	55.6	179	261
M25	0.5-310-(20% SgB + 20% FaF)	1.50	1.18	46.8	173	220
M26	0.5-310-50% FaF	1.59	0.35	66.5	141	632
M27	0.5-310-50% SgB	1.73	0.72	44.5	139	334
M28	0.5-310-LC3	2.47	0.37	58.7	110	732
M29	0.4-360-LC3	2.64	0.50	65.0	115	585
M30	0.45-340-LC3	2.90	0.57	51.0	100	530

Eq. (2) and reported in Table 2. As expected, the highest carbonation resistance is seen in concretes with no or only 15% SCMs and lower w/b, whereas the lower values occur in concretes with higher (\geq 30%) fly ash content or LC3, as expected.

4 Life cycle assessment (LCA)

The sustainability assessment framework considered here is based on two specific environmental impacts,



namely the total equivalent CO_2 emissions and energy consumed or embodied energy. Both these parameters can be estimated by following the guidelines for LCA in the ISO 14040 and ISO 14044 Standards [11, 12] or using appropriate databases. Obviously, the values will depend on the materials and fuel used, relative distances between the sources and manufacturing location, origin of the electricity, etc. For the purpose of illustration, LCA has been conducted for cement and concrete under generic considerations in South India. The parameters obtained correspond to concrete



Fig. 2 Relation between the chloride resistance factor and the service life estimate

made in Chennai, with materials that are typically used there. The analysis is done for the ground-to-gate (or cradle-to-gate or mine-to-gate) system, which includes all processes from the mines to the gate of the cement or concrete plant. The energy consumed and the (direct and indirect) emissions are considered from all processes involved in the production of concrete, including the extraction and transportation of fuels, raw materials, and the production of electricity. This system has been chosen here as it is most rational and in accordance with the scientific literature. However, the assessment can be made for other more delimited systems, as described elsewhere [33, 34].

All the cements considered here are taken to be manufactured in a plant having a double-string sixstage pre-heater with pre-calciner in Nandyal (Andhra Pradesh, India), where ordinary portland cement (OPC) and portland slag cement (PSC) are routinely produced and supplied to construction sites in South



Fig. 3 Correlation between the accelerated and natural carbonation coefficients

India. The limestone used by the plant is extracted, after blasting, from a quarry situated about 5 km away and brought by truck. The limestone has an average composition of about 65% CaO, 20% SiO₂ and 10% moisture, with 35% loss on ignition. Waste materials, such as slag, flue dust, red mud and laterite, are procured from nearby locations to supplement limestone as raw material for clinker production. The sources and distances of transportation of these materials are given in Table 3. Note that coal shipped from the USA is used along with pet coke and alternative fuels, such as pharmaceutical waste and carbon black. Electricity used in the cement plant is taken completely from the national grid. The conversion factors for calculating the impacts from the inventory of inputs have been derived from the cement plant reports and the ecoinvent database [35].

4.1 LCA of different binders

Based on the input data given in Table 3 and complementary information about the transportation, extraction of fuels, cement plant infrastructure, etc. obtained from the widely-used software SimaPro v8.0.5.13 [28], the total emissions and energy demand per tonne of clinker are obtained as 910 kg CO₂ eq. and 5290 MJ, respectively. The split-up of the impacts according to the different components (see Fig. 4) shows that the CO_2 emissions are dominated by the conversion of limestone and fossil fuels (accounting for 57% and 30% of the emissions, respectively), where the latter contributes to 73% of the energy consumed (see Fig. 4a). The impacts obtained in this case are significantly higher than those obtained in another case study also in South India (i.e., 850 kg-eq. CO₂ and 4450 MJ per tonne of clinker) [33], corresponding to production with a fuel mix of 1% coal, 53% pet coke, 32% lignite and 14% alternative fuels. The variation in impacts is attributed to differentiators including the higher clinker content, shipping of coal from USA, transportation of raw materials, lower usage of alternative fuels (8%) and higher consumption of coal (32%), pet coke (60%) and electricity. The energy demand value is, however, similar to the value of 4427 MJ/tonne [36], reported for the case of a precalciner kiln system using a fuel mix with 50% hard coal, 22% pet coke, 1% natural gas, 11% industrial waste, 13% refuse derived fuel, 1% waste rubber and 2% tires. The emissions are



Table 3	Data	related	to	the	production	of	1	tonne	of	clinker
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Input required for clinker (1 tonne)			CO ₂ emissions (kg)	Energy consumed (MI)
Material	Amount	Source (and distance from plant)	(15)	(1413)
Limestone	1420 kg	5 km	516 ^a	47.3 ^b
Flue Dust	30 kg	Bellary (225 km)		
Red Mud	25 kg	Belgavi (540 km)		
Laterite	46 kg	Rajamandri (500 km)		
Crushed slag	23 kg	Bellary (225 km)		
Coal (USA)	32 kg	USA (21,650 km)	107.2 ^a	1239 ^a
Pet coke	60 kg	Mangaluru (630 km)	165.1 ^b	2604 ^a
Pharmaceutical solid waste	3 kg	Hyderabad (300 km)	8.7^{a}	37.7 ^a
Carbon black	3 kg	Hyderabad (300 km)	9.2 ^a	75.4 ^a
Pharmaceutical liquid waste	2 kg	Hyderabad (300 km)	5.8 ^a	25.1 ^a
Electricity drawn from the grid				
For limestone crushing	1.77 kWh		2.4 ^b	35 ^b
For preparation of the raw meal	21.30 kWh		29.4 ^b	415 ^b
For the milling of coal	3.57 kWh		4.9 ^b	70^{b}
For kiln operation during clinkerization	25.59 kWh		35.3 ^b	500 ^b
Transportation				
By truck, in tonne-km	0.104	-	9.6 ^b	175 ^b
By rail, in tonne-km	0.033	_	1.5 ^b	25.3 ^b
By ship, in tonne-km	0.692	_	7.8 ^b	13.2 ^b

Conversion factors obtained from (a) cement plant data or (b) ecoinvent database [35]

lower than the corresponding production data [36] of 937 kg CO₂/tonne of clinker, which could be attributed to the differences in the processes (e.g., extent of heat recovery from the kiln) and the fuel mix.

In the production of cement at the plant considered here, the clinker is finely ground with about 5% of natural gypsum and phosphogypsum, in proportions of 50:50. The natural gypsum is transported from Oman (4754 kms away) via ship till Chennai port and then by truck, whereas phosphogypsum is procured from fertilizer industries near Tuticorin (400 km away). The PSC (53% OPC and 47% slag) is made with crushed slag brought from Bellary to Nandyal (225 km) by truck. It is further assumed, for the purpose of estimating the impacts of transportation, that PPC (65% clinker, 30% Class F fly ash and 5% gypsum) could be produced at the same plant with fly ash brought by truck from Vijayawada. Similarly, for the production of LC3, clay is assumed to be brought



from Dharmapuri to Nandyal, by truck, and calcined at the plant. Only one-way trips of loaded trucks to the plant are considered in the transportation process, as appropriate. For the LCA, the fly ash, slag and gypsum are considered as waste products and no burden is attached to them other than their transportation, in line with previous studies [33]. For the calcination of clay in LC3, the energy consumed is taken conservatively as 2.6 MJ/kg of clay based on preliminary calculations (of specific heat and calcination energy from thermogravimetric analysis of various kaolinitic clay samples) and estimates made by industry experts (including 30% losses), as in previous works [33]. The fuel mix for the clay calcination is taken to the same used for clinkerization. The data relevant to the transportation of the crushed slag, and the drying and grinding processes has been collected from the plant and used to conduct the LCA on the production of the ground granulated blastfurnace slag (GGBS). The



Fig. 4 Split-up of a energy demand (in MJ) and b CO₂ emissions (in kg-eq. CO₂) per tonne of clinker

impacts were found to be about 70 kg CO_2 eq./tonne and 1.03 GJ/tonne of GGBS, which compare well with the values given in the ecoinvent database (i.e., 63 kg CO_2 eq./tonne and 0.94 GJ/tonne). The split-up of the impacts, in Fig. 5, shows the role of the different processes, and highlights the significant contribution of grinding energy in the preparation of GGBS.

The impacts of the different types of cements hypothetically produced in the same plant have been obtained using the data corresponding to the clinker, as well as transportation, and electricity for grinding, blending, packaging, etc. As indicated earlier, the ground-to-gate system has been employed in the LCA, and the values obtained are given in Table 4, where it can be clearly seen that the impacts of OPC are much higher than those of other cements. In general, the impacts in this case, though within the general ranges given by Boesch and Hellweg [37], are higher than those obtained for a previous case study [33], primarily due to the higher clinker content of the OPC (i.e., 95% instead of 90%) and the greater energy contribution from coal imported from USA. However, the relative ranking in terms of the impacts is the same [33], with PSC and LC3 having lower carbon footprint due to the lower clinker content in these cements; see Fig. 6. Another reason for PSC to show the least impact in this case study is due to the proximity of the slag source to the cement plant. Obviously, LC3 needs



Fig. 5 Split-up of the a energy demand (in MJ) and b emissions (in kg CO₂ eq.) for the grinding of 1 tonne of GGBS

	• •			
Impact	OPC	PPC	PSC	LC3
Carbon footprint (kg CO ₂ eq./tonne of cement)	930	690	550	610
Energy demand (MJ/tonne of cement)	5945	4690	3840	4850





Fig. 6 Split-up of a CO₂ emissions and b energy demand per tonne of each cement considered

more energy than PSC and PPC for the fabrication due to the calcination process of the clay, though the emissions are less than those of PPC due to the lower clinker content.

4.2 LCA of the different concretes

Using the impacts obtained for the cements in the previous section, LCA for the ground-to-gate system [33] was conducted for the different concretes in this study, with the mixture proportions given in Table 1, considering materials typically used in Chennai, about 400 km from Nandyal, where the cements are deemed to be manufactured. Consequently, it is taken that sand is transported from Villupuram (192 km), coarse aggregate from Kanchipuram (75 km), GGBS from Goa (Slag A, 950 km) and Nandyal (Slag B, 400 km), fly ash from two sources (Class C - 275 km, from Nevveli; Class F – 50 km, from North Chennai). Further details of the LCA and steps involved can be found elsewhere [33, 34]. The impacts of each concrete are given in Fig. 7, which emphasizes the lower carbon footprint of concrete having low clinker binders (i.e., 220 kg CO₂/m³ vis-à-vis values of up to



400 kg CO₂/m³ for the concrete with only OPC). The least CO₂ emissions are obtained for concretes with OPC substituted with 30% or 50% slag or fly ash, or with combinations of 20% fly ash + 20% slag, and LC3. The energy consumed is least when large volumes of fly ash and slag are used, as these materials are considered as waste and no impact is assigned to their production; the embodied energy reduces from as high as 3400 MJ/m³ (M2) for concrete with only OPC down to 2200 MJ/m³ (M26) with 50% fly ash substitution. The impacts obtained for the different concretes are used as parameters in the calculations of the indices for the sustainability assessment in the following section.

5 Sustainability indices and decision framework

For the sustainability assessment, two scenarios are contemplated: (1) durability limited by chloride attack, and (2) durability limited by carbonation of the concrete, in both of which the steel reinforcement eventually loses the protection of the cover concrete and begins to corrode. Out of these two scenarios, the

	Cement Aggregates Electricity		Cement Aggregates Electricity
	Transportation Others Slag	2222	Transportation 📩 Others 🛛 📰 Slag
M1		M1	No SCM
M2	No SCM	M2	No SCM
M3	No SCM	M3	No SCM
M4	No SCM	M4	No SCM
M5	15% FaC	M5	15% FaC
MG	15% FaC	M6	15% FaC
M7	15%h FaC	M7	15% FaC
M8	15%h FaF	M8	15% FaF
M9	15% FaF	M9	15% FaF
M10	15% FaF	M10	15% FaF
M11	15% SgA	M11	15% SgA
M12	15% SgA	M12	15% SgA
M13	15%6SgA	M13	15%SgA
M14	15%SgB	M14	15%bSgB
M15	15% SeB	M15	15% SgB
M16	15% SgB	M16	15% SgB
M17	30% FaC	M17	30% FaC
M18	30% FaF	M18	30% FaF
M19	30% FaF	M19	30% FaF
M20	30% SgB	M20	30% SgB
M21	30%6 SgB	M21	30% SgB
M22	30% SgA	M22	30% SgA
M23	20% FaF + 20% FaC	M23	20% FaF + 20% FaC
M24	20% SgB + 20% FaC	M24	20% SgB + 20% FaC
M25	20% SgB + 20% FaF	M25	20% SgB + 20% FaF
M26	50% FaF	M26	50% FaF
M27	50% SgB	M27	50% SgB
M28	LC3	M28	LC3
M29	LC3	M29	LC3
M30	LC3	M30	LC3
	0 500 1000 1500 2000 2500 3000 3500		0 50 100 150 200 250 300 350 400
	Energy consumed (MJ/m ³ of concrete)		CO _s emissions (kg CO _s eq./m ³ of concrete)
	(a)		(b)

Fig. 7 Comparison of a energy demand and b CO₂ emissions for the different concretes

first is globally more common, especially in structures along coastal areas and those subjected to deicing salts, whereas the second may be a problem in regions with moderate humidity and higher temperatures or in urban areas with high CO_2 concentrations.

A sustainability index that is required for both scenarios, according to the proposed framework, is the energy intensity $ei_{cs,365}$, defined as the energy consumed in the production of 1 cubic metre of concrete per MPa compressive strength attained at 1 year. From the impacts obtained from the LCA (Fig. 7a), the energy intensities of the different concretes (Table 1) have been computed and given in Table 2. The lowest

 $ei_{cs,365}$ -values are for those concretes with binders where 50% of OPC has been replaced by Slag B, the concretes with ternary blends and those with LC3, highlighting the efficiency of binders that need low energy for fabrication and yield higher eventual strength gain that would benefit the structural integrity in the long-term.

The other index needed to assess sustainability in the proposed framework is Ai_{chlor} , that can be calculated from Eq. (3), to represent the combined effects of carbon footprint and chloride ingress. The values of Ai_{chlor} for the different concretes considered (Table 1) are given in Table 2, where it can be seen that the lowest values (i.e., about 100) are for the LC3 concretes and the highest (i.e., \geq 300) are for concretes with high binder contents, no SCMs and high w/b. This index reflects the high apathy toward sustainability when the latter class of concretes are used in structural applications, and shows concretes with higher clinker replacement with SCMs and LC3 in much better light.

The decision support framework for sustainability assessment of concrete for the scenario of service limited by chloride exposure is represented in Fig. 8. In this approach, the concretes closest to the lower left corner are the most sustainable among the proportions considered. It is seen that here the best concretes, in this sense, are those with LC3, OPC replaced with 50% Slag B or a combination of 20% fly ash and 20% slag.

For Scenario 2, where carbonation is more critical than chloride attack for reinforcement corrosion to initiate, the sustainability assessment requires values of Ai_{carb} that are dependent on the carbonation resistance as well as the carbon footprint, and calculated following Eq. (4). The Ai_{carb} -values given in Table 2 range from about 100–200 for concretes with no or lower (15%) dosages of SCM and low w/b up to much higher values of 600 or more for concretes with high (\geq 30%) Class F fly ash contents and LC3, reflecting the trade-off between carbon footprint and carbonation resistance. By applying the decision framework to choose the concretes that would be sustainable in environments conducive to aggressive carbonation, the Ai_{carb} - and $ei_{cs,365}$ -values are plotted for the different concretes, represented in Fig. 9. The concretes closest to the lower left corner of the plot are



Fig. 8 Decision framework based on the A-index and energy intensity for chloride exposure





Fig. 9 Decision framework based on the A-index and energy intensity for carbonation

the more sustainable choices, among the concretes considered. It can be seen that concretes with 15–30% slag and 15% fly ash, and with ternary blends having 20% slag and 20% Class F fly ash exhibit highest sustainability potential, in this case.

It is evident that the combined use of the energy intensity and A-index provides a framework to consider different aspects of sustainability in the choice of concrete composition. This could be extended to other A-indices that consider the resistance to degradation more extensively or bring in other degradation mechanisms that could be more relevant than those considered here. It is also worth noting that the sustainability potential of the same concrete could significantly differ with the exposure scenario, reiterating the need to use relevant parameters in the assessment.

6 Conclusions

A rational framework for choosing concrete mixture proportions through sustainability criteria has been proposed based on two indices: the energy intensity, $ei_{cs,365}$, taken as the energy consumed during the production of concrete divided by the compressive strength at 365 days; and an A-index, defined as the carbon emissions associated with the production of concrete divided by a parameter that can represent the durability of concrete. A high value of the A-index would reflect apathy toward environmental impact and durability considerations. The relevant experimental data for a set of thirty concretes, with strengths ranging over 20 to 60 MPa, have been obtained for providing

the parameters for the sustainability indices, including the compressive strengths at 28 days and 1 year, chloride migration coefficient and carbonation coefficients. The binders of these concretes have ordinary portland cement without and with fly ash or slag, and a new limestone calcined clay cement (LC3). The life cycle assessment (LCA) for these concretes has been performed to provide the carbon emissions and energy demand also needed for the indices, considering the mine-to-gate (or cradle-to-gate) system, for cements and concretes made in South India.

The application and results of the assessment have led to the following conclusions for the concretes studied here:

- The values of the chloride resistance factor, F_{chlor} , defined in terms of the coefficient of chloride diffusion, indicate that the concretes that could contribute most to service life under chloride attack are those with LC3 as the binder, followed by those with high SCM content.
- The values of the carbonation resistance factor, $F_{\rm carb}$, defined in terms of the carbonation coefficient, indicate that concretes with no or only 15% SCMs and low w/b would have the slowest carbonation, whereas the concretes with high (\geq 30%) fly ash content or LC3 would be those most affected.
- The lowest values of energy intensity, $ei_{cs,365}$, have been obtained for concretes with binders where 50% of OPC has been replaced by slag, ternary blends and those with LC3, highlighting the efficiency of binders that have low energy demand and yield higher long-term strength.
- The emissions from the LCA clearly show that the cements and concretes with the lowest clinker contents have the least carbon footprint. The lowest environmental impact is found in concretes with LC3 or OPC substituted with 30% or 50% slag or fly ash, or with combinations of 20% fly ash + 20% slag.
- For concretes exposed to chlorides, Ai_{chlor} -values are the lowest for the LC3 concretes, and the highest for concretes with high binder contents, no SCMs or high w/b. This reflects the apathy toward sustainability when the latter types of concrete are used in structural applications, and highlights the benefits of clinker replacement. For the carbonation scenario, the Ai_{carb} -values are lowest for concretes with no or low dosages of SCM and low w/b, and highest for concretes with high

 $(\geq 30\%)$ Class F fly ash content or LC3, reflecting the trade-off between carbon footprint and carbonation resistance.

- The most sustainable concretes under chloride exposure are those with LC3, and OPC replaced with 50% slag or a combination of 20% fly ash and 20% slag.
- For service life limited by carbonation, the most sustainable concretes are those with 15–30% slag or fly ash, and with ternary blends having 20% slag and 20% fly ash.
- It is evident that the sustainability potential of the concrete depends on the conditions that could limit its durability, reiterating the need to incorporate relevant durability parameters in the choice of mixture proportions.

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Compliance with ethical standards

Conflict of interest The authors declare that there is no conflict of interest in this work.

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