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Environmental Assessment of Limestone Calcined Clay Cement in Australia

Environmental Assessment of Limestone Calcined Clay Cement in Australia

Guangtong Huang, Yan Zhuge [*], Bernard Thomas (Tom) Benn, Yue Liu

UniSA STEM, University of South Australia, SA 5095, Australia

[*] Corresponding author's e-mail: Yan.Zhuge@unisa.edu.au

Abstract: The Australian government aims to achieve net-zero carbon emissions by 2050. Therefore, introducing a market-oriented carbon emissions trading scheme to offer a financial reward (or penalty) to those who emit below (or beyond) the allowed limits is expected. Under such a scheme, the cement industry is forced to reduce its energy consumption and carbon emissions. Limestone calcined clay (LC3) cement has been extensively studied and regarded as a promising solution to substitute ordinary cement clinker up to 50% without compromising the performance of concrete. In this paper, a comparative life cycle assessment (LCA) of the LC3 mortar considering cradle-to-gate system boundaries is conducted for the scenario in Australia. The LCA is undertaken on 122 collected LC3 mortar mix designs, and it includes the modification of traditional cement production to incorporate the calcined clay manufacture and evaluation of the environmental impact of different substitution levels. Results show that CO2 emissions associated with LC3 system production were reduced by up to 38% compared to Ordinary Portland cement mixtures.

1. Introduction

Concrete is a widely utilised material in construction, with Portland cement (PC) production contributing to about 8% of annual anthropogenic greenhouse gas emissions [1]. The increasing cement demand and lack of a cost-effective alternative to concrete necessitate strategies to reduce cement production. Using supplementary cementitious materials (SCMs) for partial replacement for cement clinker is currently the most effective approach, but the scarcity of traditional SCMs prompts the search for other cementitious materials. Limestone calcined clay (LC3) cement, which consists of a combination of PC, calcined clay, and limestone, has gained attention due to its higher cement clinker replacement and denser microstructure, which result in superior mechanical and durability properties [2].

Life cycle assessment (LCA) is a comprehensive tool that evaluates a product's environmental performance throughout its life cycle, from resource extraction to end-of-life disposal [3]. In this study, the LCA tool is used to quantify the CO_2 emission attribute to each mixture design. Previous studies have assessed the environmental impact of LC3 cement using LCA. Berriel, Favier [4] evaluated the environmental impact and the feasibility of producing three different types of cement (OPC, commercial blended cement & LC3) in Cuba, finding that LC3 mixture had the least adverse environmental impact [4]. The environmental impact of recycled aggregate concrete incorporating LC3 cement is also evaluated [5]. The results show the incorporation of LC3 cement not only significantly lower the environmental burden of recycled aggregate concrete production but also increases the durability performance. To enhance the understanding of eco-friendly cement production in Australia, this study utilises a LCA model tailored for cement manufacturing to assess the Global Warming Potential measured in CO2-eq (GWP) of the existing reported LC3 mortar design.

2. Assessment methodology

LCA analyses are often conducted on a limited number of concrete mix designs, restricting the ability to fully quantify the emissions range under specific assumptions [6]. To overcome this limitation, this study employs a modified version of Gursel's LCA model [7], implemented using Microsoft Excel. The LCA model follows ISO 14040-06 guidelines and consists of four major steps: 1) goal and scope definition, 2) inventory analysis, 3) impact assessment, and 4) results interpretation.

2.1. Goal and scope definition
The goal and scope describe the functional unit and model system boundary. The functional unit serves as the basis for linking all the inputs and outputs throughout the study. To quantify the $CO_{2\text{-eq}}$ emissions for each mix design, a simple volumetric unit of 1 m^3 was selected as the functional unit, and materials quantities were input in weight units (kg) in the model.

The system boundary was established according to ISO 14040/4 and can be classified into three types: cradle-to-gate, cradle-to-site, and cradle-to-grave [3]. This study omits maintenance and demolition impacts, as LC3 cement is a binder material in concrete, and the demolition of both OPC and LC3 systems results in similar inert waste materials. The LCA model's system boundary considers a cradle-to-gate scenario, covering manufacturing stages from raw materials extraction to LC3 mortar production, as depicted in Figure. 1. The process includes the raw materials extraction, raw materials blending and grinding, raw material pyroprocessing, clinker cooling, finish milling and grinding, materials conveying within the cement plant, aggregate production and admixture production. For LC3 system, a portion of OPC was replaced by calcined clay, limestone and gypsum, so the emission associated with producing these SCMs needs to be considered. Furthermore, unlike other by-products in the industry, such as fly ash and slag, which can be obtained directly from the plant, calcined clay used as one of the SCMs in LC3 cement requires thermal activation. This process is similar to Portland cement production, but the calcination temperature requirement is comparatively lower [8].

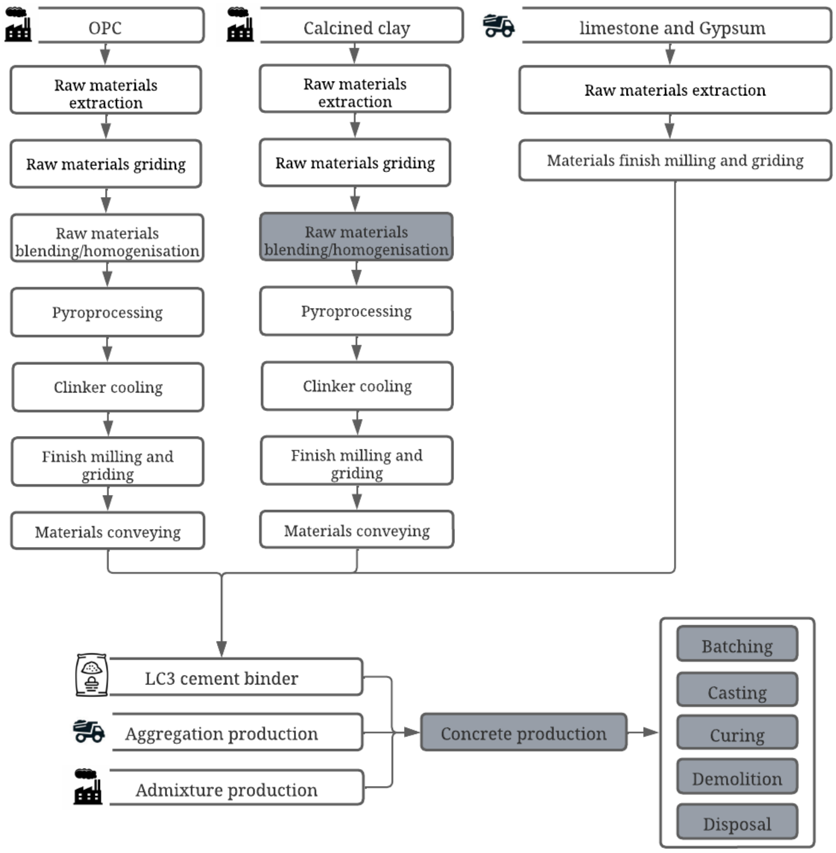

Figure 1. System boundary of the life-cycle assessment model: Cradle-to-gate production processes (the omitted processes are labelled in grey).

Gursel's model identifies four major processes responsible for emissions, namely kiln fuel pre-combustion and combustion, electricity consumption, and materials transportation. In addition, non-process related operations, such as on-site transportation and lighting within the cement plant, also contribute to emissions. The emissions associated with these processes are presented in Table 1.

Table 1. Emissions associated with OPC concrete manufacture process.

Related process	Fuel Pre-combustion	Fuel Combustion	Electricity	Transportation
Raw Materials Extraction	✓	✓	✓	✓
Raw Materials Preparation	✗	✗	✓	✗
Pyroprocessing	✓	✓	✓	✗
Clinker Cooling	✗	✗	✓	✗
Finish Milling & Grinding	✗	✗	✓	✗
Cement Conveying	✗	✗	✓	✗
Aggregate Production	✓	✓	✓	✓
Non-process Related Electricity	✗	✗	✓	✗
Admixture Production		European Federation of Concrete Admixture Association		

2.2. Life-cycle inventory analysis and model assumption

During the inventory analysis, the GWP was estimated using the modified LCA model developed by Gursel, which incorporates the evaluation of calcined clay production. Berriel, Favier [4] evaluated the environmental impact of LC3 blended cement production using three different clay calcination scenarios, including a traditional wet rotatory kiln, retrofitted calciner and optimised flash calciner. The study suggests that the retrofitted kiln with the lowest capital expenditure is the most suitable option forLC3 cement production. Therefore, the energy consumption and electricity used for clay calcination are based on the retrofitted kiln scenario. Additionally, the energy consumption and electricity use of retrofitted calciner are evaluated at a calcination temperature between 600 °C and 800 °C. In this case, an extrapolation process will be applied if the calcination temperature is above 800 °C to increase the accuracy of the calculation. In addition to the calcination temperature, the dehydroxylation process of clay results in a certain amount of mass loss, which corresponds to the mass of bound hydroxyl ions in kaolinite [8, 9], given by following

$$\mathrm{Al_2Si_2O_5(OH)_4} \rightarrow \mathrm{Al_2Si_2O_7} + 2\mathrm{H_2O} \tag{1}$$

Therefore, the kaolinite content (KC%) of clay materials can be obtained according to Eq.2. Where the dehydroxylation mass loss in percentage is defined as $\mathrm{KC-OH\%}$, the molecular mass of kaolinite and water is shown as $\mathrm{M_{kaolinite}}$ and $\mathrm{M_{water}}$, respectively. Once the kaolinite content is known, the mass loss percentage due to the removal of water from the kaolinite can be calculated.

$$\mathrm{KC\%} = \mathrm{KC-OH\%} \times \frac{\mathrm{M_{kaolinite}}}{2\mathrm{M_{water}}} \tag{2}$$

In this case, the input raw clay materials quantity ($\mathrm{Mass_{raw\ clay}}$) in LCA needs to consider the mass loss due to dehydroxylation. The mass of the raw clay materials as LCA input parameter is determined as follows.

$$\mathrm{Mass_{raw\ clay}} = \frac{7.17 \times 100 \times \mathrm{Mass_{calined\_clay}}}{7.17 \times 100 - \mathrm{KC\%}} \tag{3}$$

Where $\mathrm{Mass_{calined\_clay}}$ is the mass of calcined clay. The factor 7.17 refers to the ratio of molecular weight of kaolinite ($\mathrm{M_{kaolinite}} = 258.16\ \mathrm{g \cdot mol^{-1}}$) and two water molecules ($2 \times \mathrm{M_{water}} = 36.03\ \mathrm{g \cdot mol^{-1}}$).

Table 2 summarises the assumptions used in the LCA of LC3 mortar production. The technologies used for Portland cement manufacture were considered the average emissions scenario in the

Green_concrete_tools suggested by Gursel [7]. The average material transportation of material for cement and concrete manufacture in an Australian-based analysis suggested by [10] was employed for the base-case analysis in this study.

Table 2. Assumptions for the LC3 cement mortar production.

Material type	Assumption	
Cement	Portland cement type I	
SCMs	Calcined clay, Limestone, Gypsum	
Admixture	Superplasticizer	
Transportation	Type and model	Travel distance
Cement raw material to cement plant	Truck Class 8b	73
Raw kaolin clay to cement plant	Truck Class 8b	67
Limestone to cement plant	Truck Class 8b	25
Gypsum to cement plant	Truck Class 8b	73
Aggregate to concrete plant	Truck Class 8b	25
LC3 cement to concrete plant	Truck Class 8b	73
Admixture to concrete plant	Truck Class 8b	10
Technology options	Type of technology	
Raw materials pre-homogenisation	Dry, raw storing, non-preblending	
Raw materials grinding	Dry, raw storing, ball mill	
Raw materials blending/homogenisation	Dry, raw meal blending, storage	
Pyroprocessing	US average kiln for cement/Retrofitted kiln for calcined clay	
Clinker cooling	Reciprocating grate cooler	
Finish milling/grinding/blending	Roller press	
Materials conveying within cement plant	Screw pump (20 m between process stations)	
Concrete batching plant loading/mixing	Mix Loading (central mix)	
Concrete batching plant PM control	Fabric filter	

Table 3 shows the fuel used for electricity generation and kiln operation will consider the Australian average electricity grid [11] and the Australian average kiln fuel [6].

Table 3. Electricity grid mix and Kiln fuel percentage for Australian averages used.

Electricity Grid Mix Percentages for Australian average (%)						
Bituminous Coal	Natural Gas	Hydropower	Biomass	Solar	Wind	
34.75	37.35	17.53	0.68	0.35	8.29	
Pyroprocessing Fuel Use Options for Australian average (%)						
Bituminous Coal	Distillate (diesel or light) fuel oil	Petroleum coke (pet coke)	Natural gas	Waste oil	Waste tyre (whole)	Waste (other) (non-hazardous)
57	1	1.33	34	2.67	1.33	2.67

2.3. Impact assessment and results interpretation

Table 4 shows the unit cost and global warming potential (GWP) measured by CO2-eq for one kilogram of each component. Since the calcination temperature and clay purity (kaolinite content) can vary in different locations, the CO2-eq emissions of calcined clay production are calculated as a range, as shown in Table 4. Comparing the variations in CO2 emissions and cost, it is clear that there is a correlation between these variables and the potential carbon savings of using LC3 cement.

Table 4. CO_2 emission for different components of LC3 mortar.

Components	Unit $CO_{2\text{-eq}}$ (kg/kg)
Calcined clay	Range from 0.174 to 0.377
Limestone	0.006
Gypsum	0.002
OPC	0.886
Fine aggregates	0.003
Superplasticizer	0.786
Transportation	1.28E-04 (for Truck Class 8b)

2.4 Efficiency indicators

The binder intensity index (bi_{cs}) and CO_2 intensity index (ci_{cs}) are two performance indicators proposed by [12] to reveal binder efficiency and global warming potential. The bi_{cs} describe the amount of cement required in one cubic of mortar mass per 1 m^3 of mortar necessary to achieve 1 MPa strength. The equation is shown below. The ci_{cs}, on the other hand, show the carbon dioxide emitted during the production process of such a volume of cement that makes achieving a 1 MPa strength possible.

$$\mathrm{bi_{cs}} = \frac{\mathrm{b}}{\mathrm{p}} \tag{4}$$

$$\mathrm{ci_{cs}} = \frac{\mathrm{c}}{\mathrm{p}} \tag{5}$$

Where b is the total binder material in $kg \cdot m^{-3}$, c is the total $CO_{2\text{-eq}}$ in $kg \cdot m^{-3}$ from the production and transport of mortar materials, and p is the compressive strength (MPa) at 28 days. Both indicators allow rapid comparison of different mixes. A higher CO_2 intensity index reveals a lower environmental efficiency. While a higher binder intensity index indicates more binder materials are required to achieve the same function, contributing to a higher CO_2 intensity index.

3 LCA findings

3.1 Model assessment

A total volume of 1 m^3 of mortar sample was considered for LCA model assessment and results comparison, and 122 LC3 mortar mixes were collected from previous studies [13-25]. The statistics information for the collected dataset is tabulated in Table 5.

Table 5. Statistical description of the collected dataset.

Mixture components	**Unit**	**Unit weight (kg/m^3)**	**Min**	**Max**	**Mean**	**Median**	**Standard deviation**
Calcined clay	kg/m^3	2500	0	359.64	156.8	154.16	60.47
Limestone	kg/m^3	2710	0	179.82	76.15	77.08	36.18
Gypsum	kg/m^3	2320	0	40.73	12.92	10.49	7.94
OPC	kg/m^3	3120	107.8	465.81	289.11	282.63	66.81
Sand (Fine aggregate)	kg/m^3	2600	1293.75	1612.3	1508.84	1530.43	76.66
Water	kg/m^3	1000	188.1	262.27	228.3	227.96	19.64
Compressive strength	MPa	-	13	93.52	37.35	35.87	13.42

To better understand the carbon-saving potential of LC3 system, the dataset was divided into three subgroups based on the level of OPC substitution. For instant, LC3 (50%-80%), representing 50% to 80% of OPC was replaced with a mixture of calcined clay, limestone and gypsum. The resultant $CO_{2\text{-eq}}$ emission associated with the different LC3 systems and OPC is shown in Figure 2.

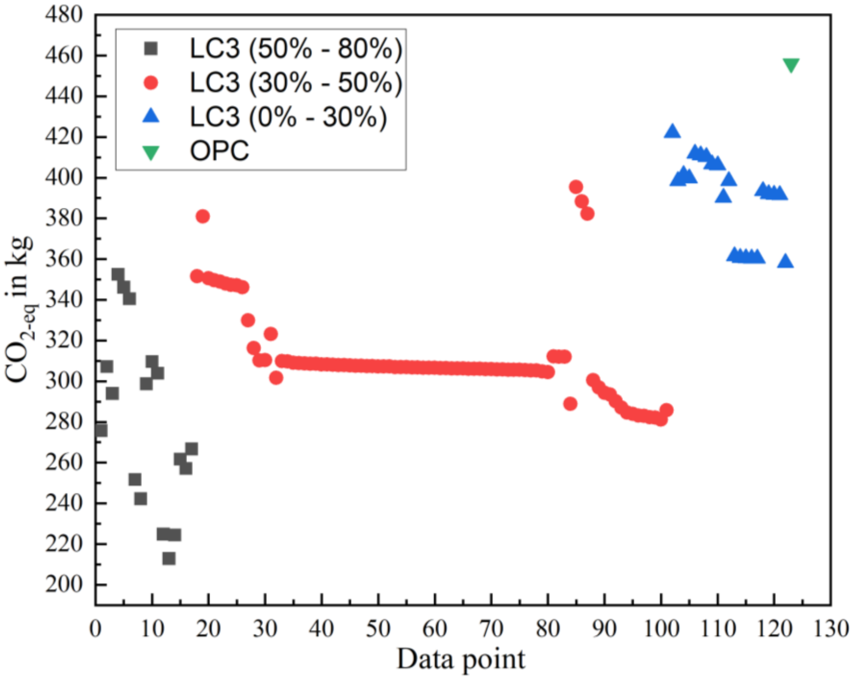

Figure 2. $CO_{2\text{-eq}}$ emission from the production of various LC3 systems and reference OPC sample.

The result shows that the reduction of cement clinker proportion in the LC3 system contributes a significant amount of $CO_{2\text{-eq}}$ emission reduction compared to the reference OPC sample. The average $CO_{2\text{-eq}}$ emission for different LC3 systems and the reduction percentage are shown in Table 6.

Table 6. Average $CO_{2\text{-eq}}$ emission for LC3 system under various substitution levels and reduction rates.

Type	$CO_{2\text{-eq}}$ (kg)	Reduction (%)
OPC	456.11	0%
LC3 (0% - 30%)	389.85	15%
LC3 (30% - 50%)	312.08	32%
LC3 (50% - 80%)	280.61	38%

3.2 Efficiency index

To understand the mixture performance of LC3 system in terms of binder intensity and Carbon (CO_2) intensity index, benchmark data from 29 countries [12] were used in this study for results comparison. The bi_{cs} index and ci_{cs} index are shown in Figure. 3 and Figure. 4, respectively.

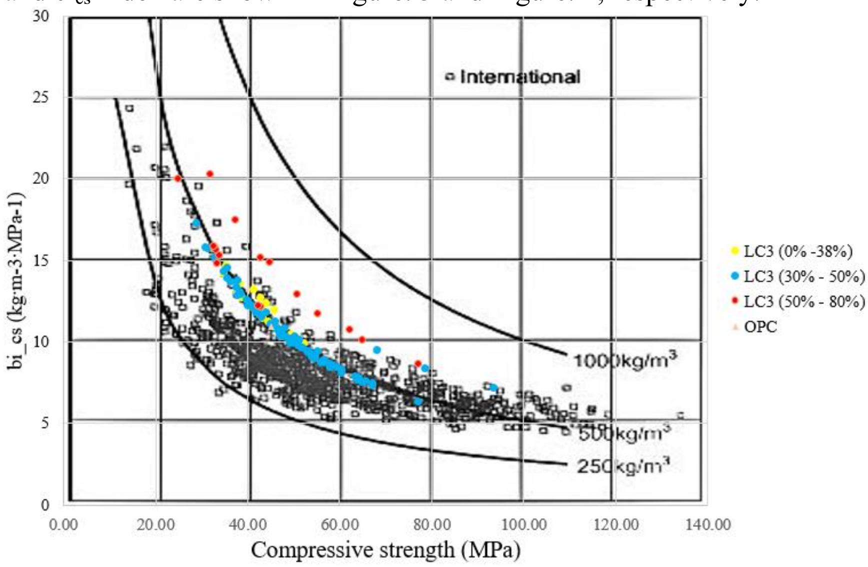

Figure 3. Binder intensity index for various LC3 system. International studies adapted from Damineli, Kemeid [12].

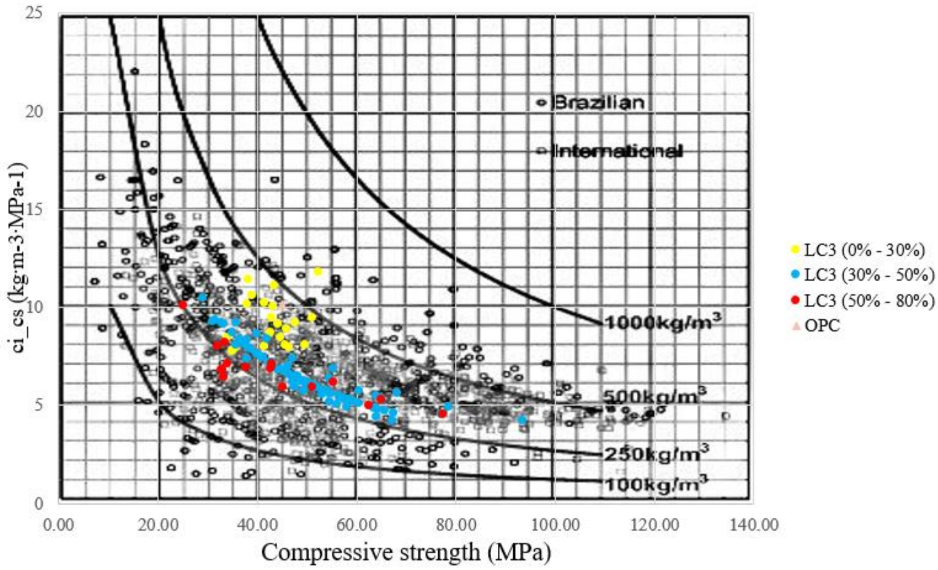

Figure 4. CO_2 intensity index for various LC3 systems. International and Brazilian studies adapted from Damineli, Kemeid [12].

Both indicators show a decreasing trend as compressive increases. The solid black line in Figure. 3 and Figure. 4 indicates the average binder intensity index and CO_2 intensity index relative to total binder content. It can be observed that the bi_{cs} value for all LC3 systems falls roughly along the binder content of 500 kg·m^{-3}, which is consistent with the binder content of the collected database (range from 488.89 kg·m^{-3} to 666.67 kg·m^{-3}). However, the majority of the ci_{cs} value of LC3 system are located close to the 250 kg·m^{-3} binder content. The lower ci_{cs} value of the LC3 system indicates a high efficiency against global warming.

Figure. 5 displays the average indicator value for the LC3 system at various percentages of cement clinker substitution, as well as the reference OPC system. The use of a combined analysis of bi_{cs} and ci_{cs} provides a more comprehensive approach to assessing the cement use efficiency for different clinker factors.

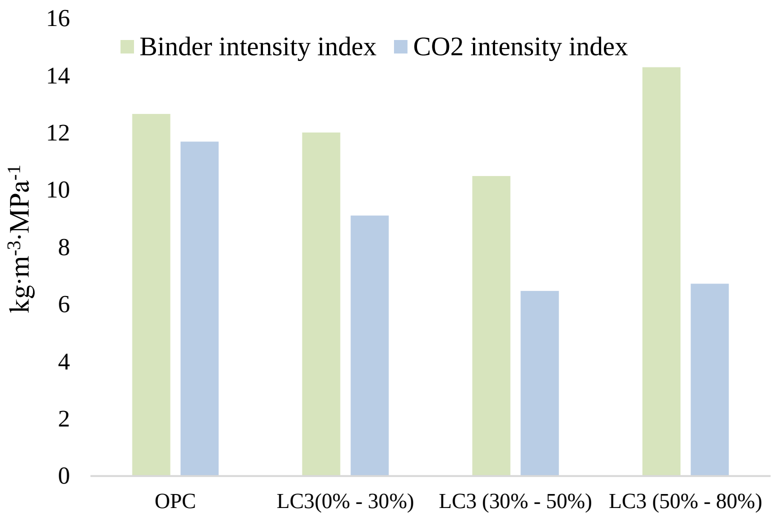

Figure 5. Combined analysis of binder intensity index and CO_2 intensity index.

For LC3 system with more than 50% cement clinker substitution, the highest average bi_{cs} value can be observed, which can be explained by the gradually decreasing trend in compressive strength and increasing trend in the amount of binder material for the high substitution LC3 system. As calcined clay is regarded as a pozzolanic material, the aluminate phase from calcined clay can react with calcium hydroxide and limestone to produce carbo aluminate hydrates [26, 27]. The low OPC content in LC3 (50% - 80%) system results in a low calcium hydroxide content for the pozzolanic reaction, eventually lowering the strength development. In addition, the focus on clinker substitution as the major means for achieving a sustainable cement blender, which leads the LC3 system with high level clinker substitution with high bi_{cs} but low ci_{cs}. The results shown in Figure.5 reveal that the LC3 (30% - 50%) contribute the lowest bi_{cs} and ci_{cs} value, which indicates the highest efficiency of this binder design.

4 Conclusions

The present study was developed to evaluate the environmental impact through the life cycle of LC3 cement production. The following conclusions should be highlighted:

- Dependent on the level of clinker substitution, the LC3 system shows a significant reduction of carbon emissions from 15% to 38% compared to the reference OPC cement.
- The results of the binder intensity index and CO_2 intensity index suggested that LC3 systems with 30% to 50% clinker substitution contribute the most in terms of the eco-efficiency of cement use.

Overall, the use of LC3 cement as a sustainable and eco-efficient alternative to OPC cement shows promise, but further evaluation is required, including the mechanical performance and economic analysis, to understand its potential benefits and drawbacks fully.

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