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Environmental Life Cycle Assessment of Alkali-activated Material with Different Mix Designs and Self-healing Agents

Jerome Ignatius T. Garces[a], Raymond R. Tan[a], Arnel B. Beltran[a], Jason Maximino C. Ongpeng[b], Michael Angelo B. Promentilla[a,*]

[a] Department of Chemical Engineering, De La Salle University, Manila, Philippines
[b] Department of Civil Engineering, De La Salle University, Manila, Philippines
michael.promentilla@dlsu.edu.ph

Alternative low-carbon cementitious binders such as geopolymers are rapidly garnering scientific interest to replace Ordinary Portland Cement (OPC). Industrial waste by-products such as coal fly ash (CFA) and ground granulated blast furnace slag (GGBS) are usually utilized for this material. Microcapsules can also be added to geopolymers to enhance its self-healing capability as it has potential environmental benefits aside from cost savings. This study thus focuses on the cradle-to-gate LCA of geopolymer concrete containing microcapsules for self-healing. Two different microcapsules were considered: Urea-formaldehyde (UF)/Dicyclopentadiene (DCPD) and Poly(urea-urethane) (PUU)/Alkali-activator (AA). Three different geopolymer precursors were studied: CFA, GGBS and CFA/GGBS. OpenLCA software was used for the calculations and analysis, and the inventory data were obtained from Ecoinvent and OzLCI2019 supplemented by literature data. An impact assessment was carried out using CML 2001. Self-healing geopolymer concrete is better in terms of lower global warming potential but performs worse in other impact categories than conventional OPC concrete. Of the geopolymer concrete assessed, GGBS concrete has the lowest impacts. On the other hand, of the two self-healing microcapsules, UF/DCPD is better for the environment. The primary factor for this difference is the solvent used in microcapsule synthesis. From this result, it has been shown that self-healing GGBS concrete containing UF/DCPD microcapsules has the lowest impacts of the self-healing concrete assessed.

1. Introduction

Ordinary Portland Cement (OPC) is used extensively in the construction sector. However, sustainability issues arise in cement production due to its significant greenhouse gas (GHG) emissions. Li et al. (2015) calculated that about 0.80 kg eq. CO_2 is produced per kg of OPC. Andrew (2019) estimates that cement production accounts for 8 % of the global CO_2 emissions and a major reason for this is the energy-intensiveness of the clinker production. Due to these issues, greener alternative materials, such as alkali-activated materials, particularly geopolymers, are gaining increasing interests to improve the sustainability of the construction sector. Geopolymers are inorganic polymers that can be obtained through the alkali-activation of aluminosilicate rich raw materials (Davidovits, 1979). Among the main properties of this material are its capacity for lower GHG emissions and waste valorization.

Geopolymers can develop cementitious properties and fully replace OPC as a concrete binder. Optimizing the mix design by incorporating different aluminosilicate sources can lead to further improvements in its properties, as shown by Sumabat et al. (2015). Environmental assessments on geopolymer concrete through Life Cycle Assessment (LCA) have also been done to quantity its impacts. Habert et al. (2011) concluded that geopolymers have that capacity for GHG reduction; however, other environmental impacts are worse when compared to OPC. Petrillo et al. (2016) arrived at a similar conclusion, and both studies attribute this to the use of alkali-activators for geopolymerization. A pollution transfer from GHG to other relevant environmental impacts is observed for geopolymers. Most of these studies are focused on coal fly ash (CFA) geopolymers. Replacing CFA with ground granulated blast furnace slag (GGBS) permits the synthesis of geopolymers with improved strengths that can be cured at ambient temperatures and uses lower doses of alkali-activators relative to CFA geopolymers (Rafeet et al., 2017). However, Rafeet et al. (2017) notes that GGBS geopolymers have low

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workability and setting time, leading to practicality issues. Mixing CFA and GGBS can allow for a compromise to resolve these issues and preserve the advantages of pure GGBS geopolymers.

Despite the promises of geopolymer, there are other issues that must be resolved, the chief of which is its proneness to crack propagation (Pan et al., 2011). To counter this, self-healing in geopolymers has been explored. Kan et al. (2019) has concluded that geopolymer can undergo natural self-healing wherein previously unreacted geopolymer precursors present within the material's matrix can react to form more geopolymers which can seal the cracks, similar in nature to autogenic self-healing in OPC concrete. However, autogenic self-healing in conventional concrete has limitations in its healing capability, and a similar behavior can be expected for geopolymers. This can possibly be boosted through the addition of self-healing microcapsules containing healing agents to assist in the self-healing of geopolymers.

To date, only one study on ex-ante LCA for self-healing of geopolymers is available and is limited to CFA geopolymers (Garces et al., 2021). Additionally, the microcapsule considered for self-healing in this study is limited to poly(urea-urethane) (PUU) microcapsules containing alkali-activators (AA). Ex-ante LCA has not been applied for self-healing CFA/GGBS and GGBS geopolymers. This research seeks to address this by considering CFA/GGBS and GGBS geopolymers and an alternative self-healing microcapsules coated with urea-formaldehyde (UF) and containing dicyclopentadiene (DCPD). This study considers the following self-healing microcapsules: (1) UF/DCPD synthesized through in-situ polymerization (Gilford et al., 2014) and (2) PUU/AA synthesized through interfacial polymerization (Beglarigale et al., 2018). The expected healing mechanism for geopolymer containing PUU/AA microcapsules is through the reaction between unreacted precursors and AA. Meanwhile, for UF/DCPD microcapsules, it is through the reaction between DCPD and the Grubbs' catalyst dispersed during geopolymer mixing. This study will provide a baseline upon which future self-healing studies on these methods can be compared to, and the environmental trade-offs that will arise during the usage phase can be examined.

2. Materials and Methods

The proposed self-healing methods are expected to contribute to the overall environmental impacts brought about by the synthesis of the geopolymer concrete. These impacts can be quantified through LCA in accordance with the International Organization for Standardization (ISO) standards for LCA methodology. Due to a lack of data with regards to the self-healing performance of the proposed methods, this study is limited to cradle-to-gate. Calculations were performed using the free software OpenLCA (GreenDelta, 2020) and MS Excel. Inventory data for LCA were obtained from literature data supplemented by databases such as Ecoinvent v3.7 and, the Evah OzLCI2019 available for free with OpenLCA. An impact assessment was performed using CML 2001, developed by the Institute of Environmental Sciences (CML) of Leiden University (Universiteit Leiden, 2015). The impact categories considered are as follows: Acidification Potential (AP), Abiotic Depletion of Fossil Fuels (ADPF), Eutrophication Potential (EP), Climate Change (GWP), Ozone Depletion Potential (ODP), and Photochemical Oxidation Potential (POCP).

2.1 Goal and scope

The scope of the study involves raw material extraction until concrete block production only. 1 m^3 concrete block with 35 MPa 28-day strength was taken as the functional unit for the different mix designs. Initially, three different types of geopolymer concrete were considered: (i) CFA geopolymers (GPC-1), (ii) GGBS geopolymers (GPC-2), and (iii) CFA/GGBS geopolymers (GPC-3). The mix formulations were obtained from Junaid et al. (2015), Serag Faried et al. (2020), and Rafeet et al. (2017). The LCA results for these mixes were compared to that of an OPC concrete (OPCC) of similar strength. Subsequently, two different microcapsule types for self-healing were considered: (a) UF/DCPD microcapsules (SH1) and (b) PUU/AA microcapsules (SH2). The environmental impacts for 1 kg of these microcapsules were compared with each other. Mix designs for self-healing geopolymer concrete were formulated by adding microcapsules to the mix designs of GPC-2 (CFA/GGBS) and GPC-3 (GGBS), equivalent to 3 % by weight of the wet binder (precursor raw material + alkali-activators + water). These were compared to OPCC. This 3 % dosage was considered as it can lead to improved concrete pore structure and acceptable self-healing performance (Wang et al., 2018). Future studies dealing with self-healing performance are recommended to confirm these. To summarize, four different mix designs for concrete blocks were formulated (OPCC, GPC-1, GPC-2, GPC-3). For the self-healing versions of the concrete blocks, four mix designs were considered (SH1GPC-2, SH1GPC-3, SH2GPC-2, SH2GPC-3). Hence, a total of eight mix designs were assessed. These mix formulations are summarized in Table 1. No allocations were allotted for CFA and GGBS and were neglected in the calculations. Additionally, to polymerize DCPD for self-healing, Grubbs' catalyst is necessary. However, no inventory data can be found for this. Thus, it was also neglected in the calculations.

Table 1: Mix designs for the concrete blocks

Component (kg/m^3)	OPCC	GPC-1	GPC-2	GPC-3	SH1GPC-2	SH1GPC-3	SH2GPC-2	SH2GPC-3
GGBS	-	-	354	71	354	71	354	71
CFA	-	354	-	283	-	283	-	283
OPC	354	-	-	-	-	-	-	-
Sodium Silicate (48 %)	-	101	72	83	72	83	72	83
Sodium Hydroxide Pellets	-	14	4	21	4	21	4	21
Water	220	39	101	106	101	106	101	106
Sand	722	741	759	742	759	742	759	742
Gravel	1,111	1,142	1,170	1,144	1,170	1,144	1,170	1,144
UF/DCPD Microcapsules	-	-	-	-	16	17	-	-
PUU/AA Microcapsules	-	-	-	-	-	-	16	17

2.2 Life cycle inventory analysis

Life cycle inventories of the relevant materials in the process are required for the calculation of the environmental impacts of the concrete blocks. Industrial scale-up of the laboratory-scale processes in the study is necessary to assess the environmental impacts accurately. For the microcapsule production, the procedures are in laboratory scales, and scale-up is necessary. The scale-up protocol by Piccinno et al. (2016) was used to generate the scaled-up inputs for industrial microcapsule production. The basis output for this is 500 kg microcapsules per batch. Waste disposal and recycling were not included in this scale-up due to a lack of data. Future works are recommended to study and include these processes for more accurate and representative results. Additionally, some of the relevant reagents lacked inventory data from databases and literature. These were neglected in the LCA calculations. Table 2 summarizes the resulting scaled up material and energy inputs.

Table 2: Material and Energy inputs for industrial-scale UF/DCPD and PUU/AA microcapsule production

UF/DCPD Microcapsule (Gilford et al., 2014)		PU/AA Microcapsule (Beglarigale et al., 2018)	
Input	Amount	Input	Amount
Poly(ethylene-alt-maleic anhydride) (EMA) , kg	7.28	4,4′-Methylenebis(phenyl isocyanate), kg	226.89
Deionized Water, kg	1,213.30	Span 85, kg	45.38
Urea, kg	29.11	Poly(ethylene glycol) dioleate*, kg	22.69
Ammonium Chloride, kg	2.91	Toluene, kg	901.67
Resorcinol*, kg	2.91	Dibutyltindilaurate*, kg	14.97
Dicyclopentadiene, kg	570.62	Deionized Water, kg	170.17
37 % Formaldehyde, kg	73.95	Alkali-activator, kg	170.17
Electricity, kWh	166.19	Electricity, kWh	303.63

*neglected in the LCA calculations.

For the scale-up of the concrete block production, daily output of 5,760 blocks was considered. Industrial machineries were obtained to model the energy requirements for concrete mixing (120 s) and block molding (45 s). Elevated temperature curing is usually necessary for CFA geopolymers. However, the incorporation of GGBS eliminates this in favor of ambient temperature curing (Rafeet et al., 2017). Steam curing at 60 °C for 48 h was assumed for GPC-1 only. The energy requirements for these are summarized in Table 3.

Table 3: Energy requirements for concrete block production per cubic meter of concrete

Process	Energy Requirement (MJ)
Concrete Mixing (Diesel)	4.10
Block Molding (Electricity)	2.05
Steam curing (GPC-1 only)	112.21

The Philippine setting was taken into consideration during the calculations. The 2017 Philippine electricity grid mix was applied for electricity usage. Most of the inventory data required for the LCA can be obtained from the databases. Inventory data for OPC was obtained from Hong and Li (2011). For the alkali-activators, sodium hydroxide and sodium silicate inventories were obtained from Hong et al. (2014) and Fawer et al. (1999). For microcapsule production, the inventory data for the inputs are mostly available in the databases. However, there is a lack of data for Span 85. Its precursor, sorbitol, was used as its representative; consequently, the data for this was obtained from Akmalina (2019). For EMA, a similar problem was encountered. To represent its impacts,

an equivalent mass of maleic anhydride and ethylene at a 1:1 molar ratio was used. For transportation, the concrete block production plant was assumed to be in Manila, Philippines.

3. Results and Discussion

Using CML 2001 methodology, the impact assessment results for one functional unit of the concrete blocks are summarized in Table 4. As can be seen from these results, geopolymers and their self-healing variants have varying environmental impacts brought about by the microcapsules. A bar chart showing the comparative analysis of the environmental impacts of the different geopolymer concrete mix designs (GPC-1, GPC-2, GPC-3) relative to OPCC is shown in Figure 1. The red line signifies the 100 % mark as a guide to visualize whether the impact of the geopolymer concrete at a specific category surpasses or falls behind that of the baseline case.

Table 4: Impact assessment results per functional unit

Component (kg/m^3)	OPCC	GPC-1	GPC-2	GPC-3	SH1GPC-2	SH1GPC-3	SH2GPC-2	SH2GPC-3
ADP, kg SO_2-eq	0.78	1.21	0.94	1.11	1.09	1.27	1.43	1.63
ADPF, MJ	1,892	2,443	1,618	2,127	1,817	2,339	4,558	5,247
EP, kg PO_4-eq	0.265	0.265	0.195	0.243	0.2226	0.273	0.340	0.397
GWP, kg CO_2-eq	333.65	207.51	138.89	178.10	171.0760	212.26	252.06	298.19
ODP, kg CFC-11-eq	$4.19x10^{-6}$	$1.10x10^{-5}$	$7.75x10^{-6}$	$9.50x10^{-6}$	$8.73x10^{-6}$	$1.05x10^{-5}$	$1.74x10^{-5}$	$1.98x10^{-5}$
POCP, kg C_2H_4-eq	0.013	0.043	0.032	0.039	0.036	0.042	0.085	0.095

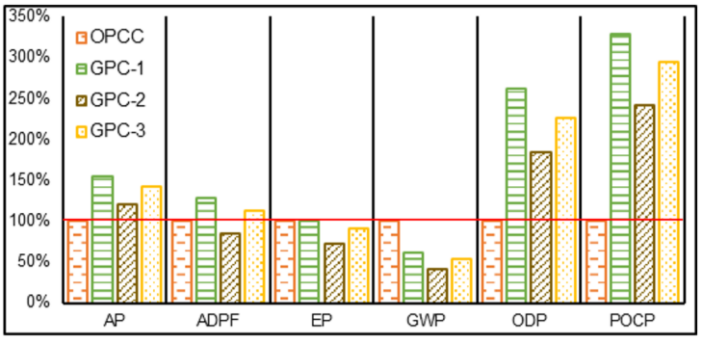

Figure 1: Comparative bar chart between geopolymer concrete and OPC concrete

Consistent with the conclusions from other LCA studies, an environmental burden shift can be observed from GWP to other impact categories. Geopolymer concrete has the capacity for reduced GHG emissions of up to 58 % reduction (GPC-2). GPC-1 has the largest impacts among the three geopolymer types due to the relatively larger alkali-activator doses required for CFA geopolymers. On the other hand, GPC-2 represents the lowest environmental impacts in all categories relative to other geopolymers because of the capacity of GGBS for lower alkali-activator requirements. As expected, GPC-3 is intermediate between GPC-1 and GPC-2 in its impacts since this mix design makes use of a CFA and GGBS mixture. Increasing the proportion of GGBS can lead to further reductions in the environmental impacts and improved compressive strengths. However, this may also lead to lower workability. Optimization with the goal of achieving minimal environmental impacts and maintaining acceptable workability can be performed for future studies. Additionally, the absence of elevated temperature curing contributed to the lower impacts of GPC-2 and GPC-3 to GPC-1.

On the other hand, self-healing versions of GPC-2 and GPC-3 through the two different microcapsules were also compared to OPCC through a bar chart shown in Figure 2. As can be seen from the chart, the mix designs containing SH1 microcapsules have significantly lower environmental impacts compared to those containing SH2 microcapsules. Additionally, it can be seen from the GWP category that despite the addition of self-healing microcapsules, the calculated impacts are still below that for OPCC, testifying to the climate change reduction potential of geopolymers. From Figure 3, SH2 microcapsules significantly exceed the impacts of SH1. This can be attributed to the difference in the reagents and solvents used in the production of the microcapsules. SH1 microcapsules use deionized water as the solvent, while SH2 uses toluene, which is more impactful than the former. Additionally, SH2 requires chemicals such as 4,4'-MDI, which adds further to the impacts brought about by toluene. Comparatively, the most relevant reagent for SH1 is DCPD, but it is still less impactful than toluene or 4,4'-MDI for SH2. Lastly, SH1 production requires lower energy than SH2. SH1GPC-2 has the lowest impacts among the different self-healing mix designs and has the capacity for reduced GHG emissions of up to 24 %

compared to OPCC. Experimentations can confirm the practicality of working with self-healing GGBS concrete and the healing efficiency of the SH1 microcapsules, supplementing the results of this study. The proposed self-healing methods require an initial investment in environmental burdens, which has the capacity to pay off during the usage phase as additional burdens from maintenance and repair are mitigated.

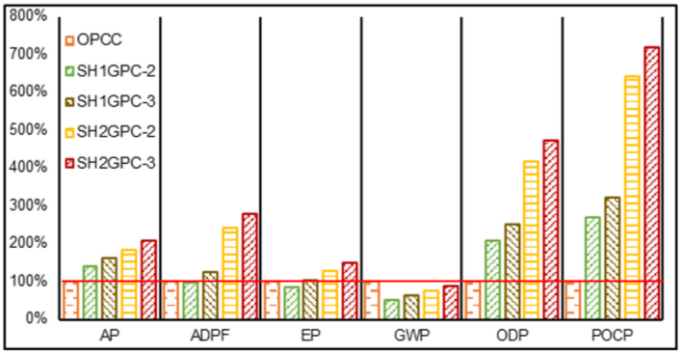

Figure 2: Comparative bar chart between geopolymer concrete and OPC concrete

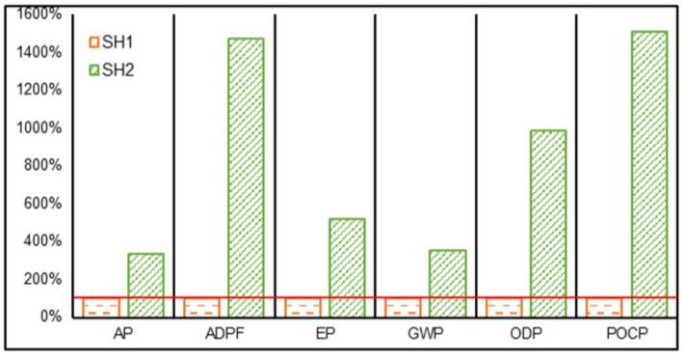

Figure 3: Comparative bar chart between UF/DCPD microcapsules (SH1) and PUU/AA microcapsules (SH2)

4. Conclusions

Ex-ante LCA was applied to assess the environmental impacts of microcapsule-based self-healing geopolymer concrete. Based on the impact assessment results, geopolymer concrete can reduce GHG emissions by up to 58 % compared to conventional OPC concrete. Additionally, among the three mix designs considered, GGBS geopolymer represents the lowest environmental impacts, while CFA geopolymer represents the opposite. On the other hand, CFA/GGBS geopolymer concrete has intermediate environmental impacts. The lower impacts of incorporating GGBS is brought about by the reduction in alkali-activator requirements and the elimination of elevated temperature curing. Optimization of the mix design considering environmental impacts and workability is recommended for future studies.

The addition of self-healing microcapsules to the concrete intensifies the impacts further. Of the two microcapsules considered, lower impacts are brought about by UF/DCPD microcapsules. This is primarily due to the utilization of water as a solvent in its production as opposed to the use of toluene for PUU/AA microcapsules. Ultimately, GGBS geopolymer concrete containing these microcapsules has the lowest environmental impacts of the self-healing concrete considered. Moreover, this self-healing geopolymer concrete has the capacity for reduced GHG emissions of up to 24 % compared to non-self-healing OPC concrete. Despite this, GGBS/CFA self-healing geopolymer concrete still has its merits, especially with regards to the material's workability. Tests on the actual self-healing performance of the materials must be done to determine the actual performance of the material and determine whether the same trends will be found in the cradle-to-grave analysis.

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