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Carbon dioxide equivalent (CO_2 -e) emissions: A comparison between geopolymer and OPC cement concrete

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HIGHLIGHTS

- ▶ Ordinary Portland cement (OPC) has high embodied energy arising from manufacturing.
- ► Carbon footprint of geopolymers, an alternative binder to OPC, was estimated.
- ► CO₂-e of geopolymer concrete is 9% less than OPC: unlike past studies (26-80%).
- ► Key factors for high CO₂-e of geopolymers: energy expended on alkali activators.
- ▶ Geopolymers need high temperature curing for strength: a further source of CO₂-e

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ABSTRACT

Concrete for construction has traditionally been based on an Ordinary Portland Cement (OPC) binder. Geopolymers, an alternative binder based on fly ash (a fine waste collected from the emissions liberated by coal burning power stations) that is activated by an alkaline activator, have potential to lower the significant carbon footprint of OPC concrete. This paper presents the results of comprehensive carbon footprint estimates for both geopolymer and OPC concrete, including energy expending activities associated with mining and transport of raw materials, manufacturing and concrete construction. Previous studies have shown a wide variation of reported emission estimates: the results of this study are benchmarked with data from those studies.

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ERIALS

1. Introduction

1.1. Background

Concrete is the most widely used construction material in the world, with current consumption of 1 m³ per person per year [1]. Ordinary Portland cement (OPC) has traditionally been used as the binder material in concrete, however OPC has high embodied energy, with carbon dioxide equivalent (CO_2 -e), the measure used to compare the emissions from various greenhouse gases based upon their global warming potential, ranging from 0.66 to 0.82 kg of CO_2 emitted for every kilogram manufactured [2–4]. The contribution of the production of OPC is approximately 5–7% of global anthropogenic CO_2 emissions [4,5]. The key causes of high CO_2 emissions arising from OPC manufacture have been attributed to: (i) calcination of limestone, one of the key ingredients, which

leads to formation and release of CO_2 ; and (ii) high energy consumption during manufacturing, including heating raw materials within a rotating kiln at temperatures greater than 1400 °C [1].

Alternative cements to OPC have been proposed to reduce greenhouse gas emissions. Blended cements, comprising OPC that has been partly substituted by supplementary cementitious materials (SCCs), are used as binders for concrete. Common SCCs include fly ash, a fine waste residue that is collected from the emissions liberated by coal burning power stations, and ground granulated blast furnace slag, a waste by-product from steelmaking. Flower and Sanjayan [6] showed that blended cements reduced CO₂ emissions by 13–22%, although this estimate can vary depending on local conditions at the source of raw materials, binder quantity and amount of OPC replacement, type of manufacturing facilities, climate, energy sources, and transportation distances.

An alternative cementitious binder, termed "geopolymer", comprising of an alkali-activated fly ash, has been considered as a substitute for OPC. Geopolymers were first described by Davidovits [7] as inorganic materials rich in silicon (Si) and Aluminium (Al) that

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react with alkaline activators to become cementitious. Alkaline activators used for geopolymers are usually a combination of a hydroxyl, usually sodium hydroxide (NaOH) or potassium hydroxide (KOH), and a glassy silicate, consisting of sodium silicate or potassium silicate [7–12], with NaOH and sodium silicate being the most common due to cost and availability. To achieve comparable strength to OPC concrete, it is necessary to provide geopolymer concrete with elevated temperature curing between 40 and 80 °C for at least 6 h [8,9,12]. Further background to geopolymers is provided in a state-of-the-art paper by Duxson et al. [8].

The range of reported CO_2 -e values for geopolymer concrete compared with OPC is considerable, with estimates as high as 80% less than OPC [13,17] to 26–45% lower than OPC concrete [14–16,18–19]. The differences in CO_2 -e arise from: (i) whether the mining, processing, and transport of raw materials have been considered; and (ii) whether the significant energy expended during manufacturing of the alkaline activators is included in the estimates. A further important factor regarding estimation of CO_2 -e is the consideration of energy expended during elevated temperature curing of geopolymers, a necessary requirement for reasonable strength development that has not been considered in past studies.

1.2. Aims

Geopolymers, when substituted for OPC as a binder in concrete, have potential to lower CO_2 emissions. This paper presents the results of a comprehensive analysis of CO_2 -e per unit of activity during the sourcing and manufacturing of raw materials, concrete production, and construction activities related to the production of 1 m³ of concrete. The results for geopolymer concrete are contrasted with comparable concrete composed of OPC binder. Previous studies have shown a wide variation of reported emission estimates: the results of this study are benchmarked with data from those studies.

1.3. Research significance

Concrete for construction has traditionally been based on an OPC binder. Geopolymers, when substituted for OPC, have potential to lower the significant carbon footprint of OPC concrete, however the few past research studies that have been conducted report a wide range of outcomes. This paper fills a knowledge gap by presenting the results of comprehensive CO₂-e estimates for both geopolymer and OPC concrete, including energy expending activities associated with mining and transport of raw materials, manufacturing and concrete construction.

2. Materials and methods

2.1. Functional unit and system boundaries

The Functional Unit, defined as CO₂-e emitted (kg CO₂-e/kg) due to the activities necessary to construct 1m³ of concrete, is the unit constant that was used in this study to compare different concrete made with OPC and geopolymer binders. The System Boundaries for the study are summarized in Fig. 1. The activities summarized in Fig. 1 relate to a point of CO₂ emission (e.g. energy use) associated with the mining, processing and manufacturing of raw materials, concrete production, and construction phases for construction of 1 m³ of concrete. Fig. 1 also summarizes the key energy types expended with each activity. The activities associated with points of emission release for the geopolymer alkali activators are summarised in Fig. 2.

2.2. Methodology

Calculation of CO_2 -e was based on the collective contributions of CO_2 , CH_4 , NO_2 , and synthetic gases evolved during each activity, taking into account the energy content of the fuel, the global warming gas types produced, and the respective gas global warming potential (GWP), when the fuel is fully combusted.

$$CO_2-e = Q \times EC \times GWP$$

where Q is the quantity of fuel combusted to undertake a particular activity (kg); EC is energy content of the specific fuel type (s) utilized to undertake the activity (J/kg); GWP is total global warming potential of the specific fuel type, comprised of the sum of the emissions of individual global warming gases comprising carbon dioxide, methane. nitrous dioxide, and synthetic gases (kg CO₂-e/l).

(1)

The EC and GWP factors used in this study, summarized in Table 1, were based on the 2012 Australian National Greenhouse Accounts (NGAs) Factors [20]. Eq. (1) was implemented using the emission factors shown in Table 1. This framework reflects international guidelines governing the estimation of national greenhouse gas inventories, including the EU Guidelines for the Monitoring and Reporting of Greenhouse Gas Emissions [21] and the US EPA Mandatory Greenhouse Gas Reporting Rule [22].

To estimate CO₂-e arising from a particular activity, the quantity and type of fuel was identified by reference to audited records obtained from the relevant manufacturers, suppliers, and contractors (for example, operation hours by particular machinery and receipts of fuel consumed). Estimates of CO₂ were made for each activity and aggregated to estimate the carbon footprint raised by 1 m^3 of concrete.

2.3. Assumptions

The following assumptions were made in the analysis:

- (a) The calculations were based on the activities associated with production of 1 m³ of Grade 40 concrete (i.e. compressive strength of 40 MPa) comprised of locally available materials, manufacturing, and construction methods in the Melbourne Metropolitan area. Common concrete mixtures were analysed in the calculations and are summarised in Table 2.
- (b) Sodium hydroxide, a commonly utilized alkali activator for geopolymers, is produced in Australia through the chlorine–alkali (chlor-alkali) process, which produces both sodium hydroxide and chlorine, as outlined in Fig. 2. Consultation with manufacturers indicated that salt brine is extracted from a nearby salt lake, transported to the factory, cleaned and prepared before undergoing electrolysis, followed by cooling to produce sodium hydroxide. The major energy expended in the process occurs in the electrolytic cell, which has a large electricity requirement despite being extremely efficient (95% according to the manufacturer, Coogee Chemicals, who utilize a membrane cell).
- (c) Sodium silicates, with weight ratio SiO₂/Na₂O of less than 2.4, are utilized for geopolymer concrete mixes, and manufactured through melting of silica sand and sodium carbonate (Fig. 2). Manufacturers confirmed that the energy expending processes for sand sourcing are the dredging, washing, drying and classifying, as well as delivery in bulk by a pneumatic tanker. For the raw material, sodium carbonate, energy is expended via the Solvay manufacturing process (i.e. ammonia reacts with CO₂ (derived from calcined limestone) which is then introduced to brine which reacts to create sodium carbonate). Sand and soda ash are then mixed and melted; a process that expends significant energy due to the high temperatures (approximately 1400 °C) and pressures needed.
- (d) A further consideration is that OPC and geopolymers have a documented ability to chemically react with airborne carbon dioxide, a process referred to as "carbonation" [23–27]. During the service life of a built concrete structure, gaseous CO₂ penetrates by diffusion through unsaturated concrete pores and reacts with alkaline solutes, leading to progressive carbonation with increasing depth. The literature reports a range of factors that influence both binders to carbonate, including the constituents of the cementitious binder, diffusivity of concrete to CO₂, compaction and curing, and exposure environment (temperature, relative humidity, and air concentration of CO₂). In terms this study, the depth of carbonation during the service life of a structure is not significant toward off-setting CO₂-e because the exposed surface area (to airborne CO₂) relative to the overall volume of concrete is very low [27] therefore the CO₂-e offset due to carbonation has not been considered in this analysis.

3. Results and discussion

3.1. Manufacture of alkali activators

3.1.1. Sodium hydroxide

Sodium hydroxide is produced concurrently with chlorine through the chlor-alkali process, the processing of salt water by electrolysis. Australian production is dominated by two organisations, Coogee Chemicals and Orica, and from discussions and our audits of their manufacturing outputs, the products NaOH and Cl₂ are almost equally produced, by mass ratio of 1.18:1 (this compares similarly with [14] based on production in the USA



Fig. 1. CO₂ emissions system diagram for production of 1 m³ concrete.



Fig. 2. CO₂ emissions system diagram for production of alkali activators.

and Europe). Following audits of energy use by the Authors, the electricity use by Australian manufacturers in their membrane cell is 2800 kW h per tonne of chlorine. This data was utilized to estimate the CO₂-e emission results per kg of NaOH, as summarized in Table 3. The estimate of 1.915 kg CO₂-e per kg NaOH is considerably greater than previously reported [17].

3.1.2. Sodium silicate

Sodium silicate is produced in Australia by three manufacturers. The method of production was confirmed as melting silica sand and sodium carbonate, although none of the manufacturers were willing to disclose any specific information about energy usage or emissions from the process. Our estimates of energy

Table 1	
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			CO 111 / 1	c	1.00	c 1.	
Emission factors	utilised to	estimate	CO_2 liberated	tor	different	fuel t	ypes.

Energy source	Emission factor ^{a,b} EC × GWP	Unit
Diesel	2.68	kg CO ₂ -e/L
Electricity ^c	1.35	kg CO ₂ -e/kW h
Liquid Petroleum Gas (LPG)	1.54	kg CO ₂ -e/L
Explosives ^d	0.44	kg CO ₂ -e/kg product

^a Includes the net effects of E_{CO_2} , E_{CH_4} , and E_{N2O} , taking into account the relevant oxidation factors and energy content of the fuel (i.e. – EC × GWP).

^b Source: Ref. [19].

^c Calculated as indirect emissions for the consumption of purchased electricity. Appropriate for Melbourne, Australia, and may vary elsewhere due to differences in energy or fuel production methods.

^d ANFO Ammonium nitrate and fuel oil slurry.

Table 2

Mixture proportions for 40 MPa concrete.

Material	Concrete mixture proportions (kg/m ³)	
	Geopolymer [30]	100% OPC [31]
Coarse aggregates	1202	1242
Fine sand	647	781
Fly ash	408	
Sodium hydroxide	41 (16 M)	
Sodium silicate (Na ₂ O = 14.7%, SiO ₂ = 29.4%, and water = 55.9%)	103	
OPC		328
Superplasticizer	6	_
Free water ^a	26	190
Curing	Steam curing at 60 °C for 24 h	Moist curing

^a Allowance was made for free water contained in the alkali activators and aggregates (to saturated surface dry condition).

Table 3

Emissions from NaOH manufacture.

	Emission	Unit
Electricity used in cell per tonne NaOH	1.285	kW h
Emissions from cell	1.581	kg CO ₂ -e
Proportion of cell electricity used in process	115.8%	%
Overall emissions from electricity	1.830	kg CO ₂ -e
Percentage of cell electricity used in fuel	25.3%	
Energy used on fuel	0.325	kW h
Overall emissions from fuel	0.086	kg CO ₂ -e
Total Emissions	1.915	kg CO ₂ -e

expenditures were based an extensive inventory of the energy consumed for the production of sodium silicate, including transport and reported by Fawer et al. [28]. The results of CO₂-e estimates using NGA factors [20] are shown in Table 4. The total emission estimate of 1.514 kg CO₂-e per kg sodium silicate is significantly higher the reported value of 1.0 [17]. Fawer et al. [28] did not include the energy expended during extraction of raw materials (e.g. sand dredging, washing, drying and classifying) and the actual CO₂-e is likely to be higher than the estimate of 1.514 CO₂-e.

3.2. Manufacture of OPC

The estimation of CO_2 -e due to cement manufacturing is complicated due to the chemical liberation of CO_2 due to decomposition of limestone during calcination, limestone source variability, and also the use of calorific wastes in cement kilns which provide energy as a substitute fuel. Within Australia, the most recently reported emission factor for cement production is 0.82 kg CO_2 -e/kg [27]. This estimate includes the mining of raw materials, cement manufacturing, and all transport associated, including the conveyance of cement to concrete batching plants. However, this estimate does not include the compensation calculation of CO_2 -e/kg due to the energy saved from waste fuel utilisation during cement manufacturing and therefore it is a conservative estimate.

Table 4Estimates of emissions arising due to sodium silicate manufacture [28].

	Emissions arising from energy expended during manufacturing		
	Energy flow (MJ/1000 kg)	Emissions (kg CO ₂ -e/kg)	
Electricity	3118	1.065	
Coal	296	0.027	
Oil (heavy)	9	0.001	
Oil (average/light)	456	0.033	
Diesel oil	144	0.010	
Gas	1270	0.076	
Others	78	0.009	
Total	5371	1.222	
	Emissions caused by transport		
	Air emissions (kg/1000 kg)	Emissions (kg CO ₂ -e/kg)	
Carbon dioxide (CO_2)	288.7	0.289	
Methane (CH ₄)	0.128	0.003	
	Total	0.292	
	Grand total (kg CO ₂ -e/kg)	1.514	

3.3. Manufacturing of fly ash

Fly ash is a waste by-product arising from coal-burning power stations and therefore some studies have considered the raw material to contribute zero CO_2 -e [14]. However energy expenditure occurs during fly ash capture, milling and grinding, drying, and transport [29] and an emission factor of 0.027 kg CO_2 -e/kg for fly ash has been calculated. Compared with OPC and the alkali activators, fly ash has a significantly lower emission factor.

3.4. Manufacturing of aggregates

The fine and coarse aggregates are assumed to be locally quarried basalt and river sand. Inventory data collected from coarse and

Table 5

emission factors manufacture and placement of concrete [27].

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	Activity	Emission factor	Unit
	Concrete batching	0.0033	kg CO ₂ -e/m ³
	Concrete transport	0.0094	kg CO ₂ -e/m ³
	On site placement activities	0.009	kg CO ₂ -e/m ³
	Concrete transport On site placement activities	0.0094 0.009	kg CO_2 -e/m ³ kg CO_2 -e/m ³

fine aggregate quarries within Victoria (Australia) has been used for estimating the emission factors of 0.0408 kg CO_2 -e/kg for coarse aggregate and 0.0139 kg CO_2 -e/kg for fine aggregate. Despite the inclusion the activities of quarrying and crushing, transport of raw materials to the concrete manufacturing premises, the emission factor for the aggregates is very low when compared with OPC and the alkali activators.

3.5. Concrete construction

The contribution to emissions during the concrete production and construction of 1 m^3 of concrete has been based on the mixing and transport of concrete from a Melbourne metropolitan concrete plant to the construction site of a bridge (within 10 km distance). The case examines construction of a crosshead beam that is part of a two-span concrete bridge. Energy expenditure occurs due to manufacture and transport of concrete, provision of temporary structural support and access during construction, pumping and concrete placement, finishing, and curing of the concrete. Theequivalent emission factors estimated for each of the concrete construction activities are summarized in Table 5.

3.6. Elevated temperature curing

In addition, to achieve comparable strength to OPC concrete, the geopolymer mixture required elevated temperature at 60–80 °C for 24 h [8,9,12]. Our estimates of energy expended due to elevated temperature curing were based on audits of natural gas usage by four local precast concrete manufacturers for elevated temperature curing of concrete (approximately 16 h overnight) at an average temperature of 50 °C, which was extrapolated to 24 h for our calculations (plus approximately 9 h gradual heating/cooling time). The calculated emission for elevated temperature curing was 39.97 kg CO_2 -e/m³. It is important to note that previous estimates of

CO₂-e geopolymer binders [13,15–17] do not include elevated temperature curing in the calculations.

3.7. Comparison between OPC and geopolymer concrete

The contribution to CO_2 -e from each of the activities, from sourcing raw materials to the manufacture and construction of 1 m³ of concrete, is summarised in Fig. 3. Portland cement was by far the most significant contributor to emissions; contributing 76.4% of CO_2 -e for OPC concrete. However, the alkali activators expend significant energy during manufacture and the contribution of the geopolymer binder (fly ash + sodium silicate + sodium hydroxide) is 201 kg CO_2 -e/m³ compared with OPC 269 kg CO_2 -e/ m³ (25.2%). The total emissions from the OPC and geopolymer concrete comparison mixes used in this report were estimated as 354 kg CO_2 -e/m³ and 320 kg CO_2 -e/m³ respectively, showing 9% difference.

A further consideration would be to compare geopolymer concrete with CO_2 -e arising from blended fly ash or slag cements that comprise partial replacement of OPC. Although we have not conducted calculations on specific blended cement concrete mixtures, 19–29% reductions in CO_2 -e are feasible [6,27] and would provide lower CO_2 -e than geopolymer binders.

3.8. Comparison with earlier studies

Differences between studies arise due to proximity, availability and composition of raw materials; energy/fuel types; concrete mixture compositions, and manufacturing processes for the alkali activators. Nevertheless, even allowing for these differences, our findings were that the CO₂-e of geopolymer concrete was approximately 9% less than comparable concrete containing 100% OPC binder. This contrasts with earlier studies: the range of estimates of CO₂-e due to substitution of OPC with a geopolymer binder ranging from 80% [13,17] to 26–45% [14–16,18,19] lower than OPC concrete. The key factors that have led to the differences in reported results include:

(i) Transportation emissions were considered by Stengel et al. [14] but not considered in [15] and unclear in [13,17]. Transportation emissions are significant and explain differences in the past estimates of CO₂-e.



Fig. 3. Summary of CO₂-e for Grade 40 concrete mixtures with OPC and geopolymer binders.

- (ii) Expenditure of significant energy during sourcing of raw materials and manufacturing of the sodium silicate and sodium hydroxide alkali activators for the geopolymer concrete was not included in previous estimates.
- (iii) Elevated temperature curing contributed 12.4% CO₂-e for the geopolymer concrete, compared with negligible emissions for OPC concrete that was cured at ambient temperature. The contribution was not considered in [13,15–17].

The quantities of alkaline activator required to manufacture geopolymer concrete are considerable, totalling 103 kg/m³ and 41 kg/m³ sodium silicate and sodium hydroxide for the production of 1 m³ of concrete, respectively. The manufacture of the activators is energy intensive, requiring: (i) sodium silicate from the melting of silica sand and soda ash at approximately 1400 °C; and (ii) sodium hydroxide which is processed by electrolysis of salt water. The contributions to CO₂-e by the alkaline activators were considered in [13,17], although only based on personal communications with the Supplier rather than an audit of energy expended for each activity associated with manufacturing and transport. It is also unclear whether [13,17] included mining, treatment and transport of raw materials for manufacture of alkali activators. Previous studies [14–16] are based on data from European manufacturing of the activators and therefore comparison between the studies is made difficult by the different sources of energy and also the types of production when compared with Australian conditions for the manufacturing of the alkali activators; sodium silicate and sodium hydroxide.

4. Conclusions

This study quantified the carbon dioxide equivalent emissions (CO_2-e) generated by all the activities necessary to obtain raw materials, concrete manufacturing, and construction of one cubic metre of concrete in metropolitan Melbourne. The CO_2 -e footprint generated by concretes comprising geopolymer binders and 100% OPC concrete were compared. The CO_2 footprint of geopolymer concrete was approximately 9% less than comparable concrete containing 100% OPC binder: much less than predictions by earlier studies. The key factors that led to the higher than expected emissions for geopolymer concrete included the inclusion of mining, treatment and transport of raw materials for manufacture of alkali activators for geopolymers, expenditure of significant energy during manufacture of alkali activators, and the need for elevated temperature curing of geopolymer concrete to achieve reasonable strength.

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