

Geopolymer Bricks from Concrete Residue and Palm Oil Fuel Ash: Evaluating Physical-mechanical Properties, Life Cycle Assessment and Economic Feasibility

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Abstract

This study investigated the development of physical and chemical properties in geopolymer brick from palm oil fuel ash by adding concrete residue. Palm oil fuel ash was mixed with concrete residue in five ratios: 100:0, 90:10, 80:20, 70:30 and 60:40 by weight. Ten molar alkaline activator solution was prepared from NaOH and Na₂SiO₃ in the weight ratio of 2:5. The physical and mechanical properties of the bricks were analyzed according to Thai standard TIS 77-2545, and the chemical properties were investigated for brick structure. Life cycle assessment (LCA) using SimaPro 7.3.3 and economical feasibility were calculated. The results showed that calcium oxide in concrete residue reduced the pore volume and increased the formation of calcium silicate hydrate gel and developed the compressive strength. Using 40% concrete residue in geopolymer brick produced the highest compressive strength (12.53 MPa). Geopolymer bricks made from palm oil fuel ash and concrete residue with the optimal ratio 60:40 had low impact on climate change, at 18.49%. The break even volume was 577,428 pieces, and the payback period was 3.88 years.

Keywords: Palm oil fuel ash; Concrete residue; Geopolymer bricks

1. Introduction

Geopolymer, developed by Davidovits (1991), is an inorganic material having amorphous polymetric structures with interconnected Si-O-Al bonds formed by dissolving silica and alumina from raw materials in a highly concentrated alkali hydroxide or silicate solution (Wongpa *et al.*, 2010; Nazari *et al.*, 2011; Ahmari and Zhang, 2013). Geopolymer is increasingly used as a construction material, because of its rapid development of compressive strength (Nazari *et al.*, 2011; Ahmari and Zhang, 2013). Numerous studies have investigated using high silica and alumina sources as raw materials for producing geopolymer.

In Thailand, palm trees for oil constitute the major crop (Saswattecha *et al.*, 2016). After the process of extracting oil from the palm fruit to produce vegetable oil, a large amount of solid waste in different forms is generated, including

shells and fibers (Islam et al., 201). These are usually used to generate heat for boilers in the oil industry (Tangchirapat et al., 2007). After burning, the waste becomes palm oil fuel ash (Tangchirapat et al., 2007). Approximately 100,000 tons of palm oil fuel ash are disposed of in landfills every year, causing environmental problems (Chindaprasirt et al., 2007). Palm oil fuel ash is a pozzolanic material with a high silicon oxide composition (Chindaprasirt et al., 2007). Silicon oxide experiences a pozzolanic reaction with calcium hydroxide to produce calcium silicate hydrates, and this reaction can improve crystallinity in microstructures (Pourakbar et al., 2015). Islam et al. (2014) found that palm oil fuel ash should be used as a binder in low quantities, because geopolymer brick produced from palm oil fuel ash forms a C-S-H gel with low compressive strength and gradual setting (Salih et al., 2014).

Numerous studies use high silica and alumina materials mixed with palm oil fuel ash, for instance, fly ash, rice husk ash and fumed silica, to produce strong binders (Islam *et al.*, 2014; Karim *et al.*, 2013). The geopolymer brick from palm oil fuel ash can improve compressive strength up to 92 MPa over 90 d through the addition of a high-calcium material, ground granulated blast furnace slag. Calcium compounds from slag increases the formation of C-S-H gel as well as the compactness of the gel (Salih *et al.*, 2015).

Concrete is the main material in concrete brick for building construction. It is frequently used in the process of concrete production, generating waste called concrete residue. Concrete residue has less cohesiveness and cannot be recycled to form concrete. Consequently, all produced concrete residue is disposed to landfill (Siriruekratana and Supakata, 2017). The major compositions in concrete residue are calcium and silica, with small amounts of alumina and iron oxide (Siriruekratana and Supakata, 2017). Calcium compounds in concrete residue can improve the compressive strength of geopolymer brick, because calcium compounds increase the calcium silicate hydrate (C-S-H) gel, and this binding gel forms gel networks to make geopolymer brick more durable (Salih et al., 2015). However, adding calcium into the matrix has limitations, because the excess calcium compound uses a high amount of water in the hydration reaction (Lee and Lee, 2013) to increase the formation of C-S-H gel, causing cracks in the brick.

In this study, concrete residue was added as the calcium source in geopolymer brick made from palm oil fuel ash in order to increase its compressive strength. The aim of this study was to investigate the physical and mechanical properties of geopolymer brick produced from palm oil fuel ash and concrete residue to make it compliant with the TIS 77-2545 standard. Life cycle assessment (LCA) and economical feasibility were investigated. The findings of this research will facilitate the development of geopolymer brick using concrete residue and palm oil fuel ash in terms of environmentally and economical friendly.

2. Materials and Methods

2.1 Materials

Palm oil fuel ash was collected from the Thai Eastern Group in Chonburi Province, Thailand, and concrete residue was the waste remaining after cleaning the equipment for concrete production obtained from Tratmunkong Construction Materials Co., Ltd. in Trat Province. It consists of Portland cement type I 12.5%, sand 37.5%, gravel 45.8%, and water 4.2%. The palm oil fuel ash was dried and sieved through 300 μ m (Salih *et al.*, 2015). Then, the ash was passed though sieve No.200 (<75 μ m). The concrete residue was dried and also screened into smaller sizes through sieve No.200 (<75 μ m).

2.2 Alkaline solution

Flakes of 99% pure sodium hydroxide (NaOH) were dissolved in distilled water to prepare a 10 molar sodium hydroxide solution. Sodium silicate solution (Na₂SiO₃) was obtained from industrial-grade sodium silicate in liquid form with 14.25% Na₂O, 31.25% SiO₂ and 54.50% water by weight. For the alkaline solution, the mass ratio of sodium hydroxide to sodium silicate was 2.5, and the solution was prepared 24 h before use (Salih *et al.*, 2015).

2.3 Characterization of palm oil fuel ash and concrete residue

The chemical composition of the palm oil fuel ash and concrete residue were determined using an X-ray fluorescence spectrometer (Bruker model S8 Tiger). The particle size distribution of the ash and the concrete residue were analyzed using laser diffraction spectroscopy (Malvern Mastersizer 3000).

2.4 Preparation of geopolymer bricks

Palm oil fuel ash (PA) and concrete residue (CR) were mixed to form a homogeneous powder in the ratio shown in Table 1. The alkaline solution was mixed with the materials in the ratio of 0.8 materials: alkaline solution by weight for 10 min. The specimens were

Name	Palm oil fuel ash (% by weight)	Concrete residue (% by weight)	
PA100	100	0	
PA90CR10	90	10	
PA80CR20	80	20	
PA70CR30	70	30	
PA60CR40	60	40	

Table 1. The ratio of materials.

poured into $5 \ge 5 \ge 5 \le 3$ acrylic molds in two layers. The molds were vibrated using a vibrating table to compact the specimen for 1 min/layer immediately after mixing and were wrapped with plastic film for 24 h to prevent moisture loss. The specimens were cured at ambient temperature for 24 h.

2.5 Characterization of geopolymer bricks

The methods in Thai Industrial Standard 77-2545 (2003) were used to test the physical properties of the bricks. The microstructure of the bricks was tested by scanning electron microscope (JEOL JSL-6500F). An X-ray diffractometer (D8-Discover) was used for the mineralogical phase identification of the geopolymer brick, and Fourier Transform Infra-Red (Perkin Elmer, Spectrum One) was used to measure the functional group.

2.6 Life Cycle Assessment of geopolymer bricks

There are 4 steps in terms of LCA including goal and scope definition, inventory analysis, impact assessment, and interpretation. In the present study, the life cycle assessment (LCA) methodology was implemented for geopolymer brick from palm oil fuel ash and concrete residue (optimum ratio of 60:40 obtaining the highest compressive strength) in laboratory in term of cradleto-gate using SimaPro 7.3.3 software from National Metal and Materials Technology Center for calculation. Six impact categories were considered: climate change, ozone depletion, human toxicity, terrestrial acidification, fossil depletion, and terrestrial ecotoxicity.

2.6.1 Goal and scope

The goal of this study was to investigate the environmental life cycle impact of geopolymer brick made from concrete residue and palm oil fuel ash. The analysis used in this study follows the phases of LCA, which cover raw material acquisition to geopolymer brick manufacturing, otherwise called "cradle to gate". The functional unit was defined as one piece of facing brick (5 cm \times 5 cm \times 5 cm) compliant with TIS 77-2545.

2.6.2 Life cycle inventory

The life cycle inventory (LCI) data of geopolymer brick was gained from lab-scale experiment as shown in Table 1, with the optimal ratio.

2.6.2.1 Assumptions

(1) Raw material transportation vehicles for the four geopolymer bricks are light commercial vehicles (LCV) using diesel as fuel. Light commercial vehicles (LCV) have at least 4 wheels, with a 3,500 kilogram weight limit, and are used to transport goods and have a maximum of 8 seats for passenger transportation (Thailand Automotive Institute, 2013).

(2) Transportation distance was estimated from industry to work site at the Department of Environmental Science, Chulalongkorn University.

(3) Raw materials were assumed to be industrial waste that does not cause environmental impacts. Thus, environmental impact was calculated from only raw material transportation.

2.6.2.2 Inventory and data calculation

The input data were defined from the material and energy resource information used in the geopolymer brick process. The output data were defined from waste and emitted pollution. The inventory of geopolymer brick is shown in Figure 1.

2.6.2.3 Data calculation

The emission estimation methodology covers exhaust emissions by road vehicles, including ozone precursors (CO, NO_x, non-methane volatile organic compounds, NMVOC), greenhouse gases (CO₂, CH₄, N₂O), and acidifying substances (SO₂, NH₃). Particulate matter, toxic substances (Benzo (a) pyrene, Benzo (k) flouranthene, Benzo (b) flouranthene) and heavy metal (lead) were estimated by the 2006 IPCC guidelines (IPCC, 2006), and the 2016 EEA air emission inventory guidebook (EEA, 2016).

2.6.3 Life cycle impact assessment

The life cycle assessment study was carried out with the SimaPro 7.3.3 software, using ReCiPe Midpoint (H) V1.06, World ReCiPe H method from the National Metal and Materials Technology Center (MTEC). Results were expressed in six impact categories, including climate change, ozone depletion, terrestrial acidification, human toxicity, fossil depletion, and terrestrial ecotoxicity.

2.6.4 Interpretation

In this step, the data were analyzed. The results from the SimaPro 7.3.3 software calculation were collected.



Figure 1. The inventory of geopolymer brick

2.7 Economical Feasibility

An economics analysis of the production of geopolymer bricks from palm oil fuel ash and concrete residue (with an optimal ratio) was evaluated using the following equations. The break even volume (N^*)

 $N * = \frac{F}{P-V}$, (1) (Moles and Terry, 1997) where

N* is the break even volume

F is the fixed cost, Baht

P is the price per unit, Baht/unit

V is the variable cost, Baht

The payback period

Payback period

Payback period $= \frac{N^*}{N}$,(2) (Moles and Terry, 1997) where

N* is the break even volume

N is the productivity yield/year

The data for fixed cost and variable cost are shown in Table 2.

3. Results and Discussion

3.1 Characterization of Palm oil fuel ash and concrete residue

3.1.1Chemical composition of raw materials

Table 3 shows the chemical composition of the oxide form of palm oil fuel ash and concrete residue. The ash consists of a high amount of SiO_2 and CaO at concentrations of

Table 2. Data of fixed	cost and	variable	cost
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46.0% and 7.59%, respectively. Al_2O_3 is found in a low concentration of approximately 0.74%. In the concrete residue, the major components are CaO and SiO₂ at concentrations of 27.4% and 24.7%, respectively.

3.1.2 Particle size distribution

The particle size distributions of the raw materials are shown in Figure 2. The particle sizes of palm oil fuel ash with D_{10} , D_{50} and D_{90} were 8.38 µm, 33.3 µm and 80.4 µm, respectively, which show that the size of palm oil fuel ash particles is approximately 33.3 µm. The particle sizes of concrete residue with D_{10} , D_{50} and D_{90} were 3.60 μ m, 19.8 µm and 76.2 µm, respectively. The specific surface area of the raw materials was 422.1 m²/kg for the palm oil fuel ash and 739.0 m^2/kg for the concrete residue. The results show that concrete residue particle size is smaller than that of palm oil fuel ash (SEM images in Figure 2), and concrete residue has a greater specific surface area than that of palm oil fuel ash.

3.2 Characterization of geopolymer bricks

3.2.1 General appearance

The general appearance of geopolymer brick is shown in Figure 3. All batches of geopolymer bricks had a smooth surface and black color, following the TIS 77-2545 specification.

	Туре	Cost	Source
Industrial	Concrete residue	0 Baht/kg	Tratmunkong Co.,
wastes			Ltd., Trat province
	Palm oil fuel ash	0 Baht/kg	Thai Eastern Group,
			Chonburi province
	Sodium silicate (Na ₂ SiO ₃)	50 Baht/kg	Roongsub Chemical
Chemicals			Ltd.,Part., Bangkok
	Sodium hydroxide (NaOH)	300 Baht/kg	Roongsub Chemical
			Ltd.,Part., Bangkok
	Los Angeles abrasion machine	110,000 Baht/unit	(Cooper
	(Cooper technology Ltd.)		Technology, 2017 :
			online)
	Aggregate vibration screen	120,000 Baht/unit	(Gilson company,
Machines	(Gilson Porta-Screen® PS-3F)		2017 : online)
	Electric concrete pan mixer (JQ350)*	25,000 Baht/unit	(Alibaba, 2017 :
			online)
	Laboshake (C. Gerhardt GmbH & Co. KG)	18,000 Baht/unit	(C. Gerhardt GmbH
			& Co. KG, 2015 :
			online)
	Drying oven (BINDER BD/ED/FD)	46,000 Baht/unit	(Binder, 2017 :
			online)

The weight of geopolymer brick per unit was 0.025 kg.

Chemical compound	Palm oil fuel ash (%)	Concrete residue (%)
SiO ₂	46.0	24.70
Al_2O_3	0.74	6.65
CaO	7.59	27.4
K_2O	-	0.44
MgO	0.11	1.96
Fe_2O_3	2.47	2.88
P_2O_5	4.78	< 0.1
SO3	0.85	1.88
Cl	0.63	0.24
TiO ₂	< 0.1	0.29
Na ₂ O	0.12	0.30

Table 3. Chemical composition of palm oil fuel ash and concrete residue



Figure 2. The particle size distribution and microstructures of palm oil fuel ash and concrete residue



Figure 3. General appearance of geopolymer bricks

3.2.2 Water absorption

The water absorption of the geopolymer bricks is shown in Figure 4. The standard value of water absorption, according to the TIS 77-2545 standard, should be less than 25%. The results showed that the minimum water absorption was 0.25% in PA60CR40. The water absorption decreased when the volume of concrete residue increased. Concrete residue particles are smaller than those of palm oil fuel ash; therefore, the concrete residue particles could fill in pores of the specimen structures, increasing the density of the bricks and decreasing the pore volume in the structure of the brick specimens. The maximum water absorption was 1.8% in PA100. These results show that palm oil fuel ash, with larger particles than concrete residue (Figure 2), forms a binder with more varied porousness and no filler (Figure 6), as compared to the concrete residue, to reduce the pores. The alkaline activator solution also reacts more with the concrete residue than with palm oil fuel ash, because the concrete residue has a higher surface area and an increased geopolymerization reaction rate (Liew *et al.*, 2016).



Figure 4. Water absorption of geopolymer bricks



Figure 5. Compressive strength of geopolymer bricks



Figure 6. The microstructure characterization of geopolymer bricks: (a) PA100 (b) PA90CR10 (c) PA80CR20 (d) PA70CR30 (e) PA60CR40

3.2.3 Compressive strength

The compressive strength of the geopolymer bricks is shown in Figure 5. The standard value of the compressive strength, according to the TIS 77-2545 standard, should be higher than 7 MPa. As shown in Figure 5, the compressive strength of the geopolymer bricks increased when the concrete residue was replaced. The results showed that palm oil fuel ash was the only aluminosilicate source material. The effect of calcium from the concrete residue to develop geopolymerization reaction is described as follows:

Calcium reacts with an alkaline activator solution to accelerate the geopolymerization process. The amorphous phases shift to poorly crystalline phases that may cause water insufficiency in the alkaline mixture and increase the alkalinity. The higher alkalinity can increase the dissolving of aluminosilicate, which results in a higher rate of polycondensation-geopolymerization (Khater, 2012; Salih *et al.*, 2015).

In a high calcium concentration, calcium reacts with silicate and aluminate to form a calcium silicate hydrate gel (C-S-H) (Temuujin *et al.*, 2009).

Aluminate substitutes in the C-S-H gel synthesize to a calcium aluminosilicate hydrate gel (C-A-S-H) by crosslinking between C-S-H chains (Salih *et al.*, 2015). It increases the compactness, reduces the total pore volume and develops the compressive strength.

3.2.4 Microstructures of geopolymer bricks

The microstructures of the geopolymer bricks are shown in Figure 6. In PA100, the structure has various pores, causing high water absorption and low compressive strength. Calcium from the concrete residue has a smaller size than palm oil fuel ash, so it can fill in the spaces to increase the density in the structure found in PA60CR40. The results suggest that calcium increases C-S-H gel formation and changes a rough structure to a smooth structure, causing high compressive strength and low water absorption. It was observed that the addition of concrete residue into the brick produced from palm oil fuel ash decreased the pore volume and supported the increment of the compressive strength with low water absorption (Siriruekratana and Supakata, 2017).



Figure 7. The mineralogical phases of geopolymer bricks

3.2.5 XRD patterns of geopolymer bricks

Figure 7 shows the XRD patterns of the replacement of palm oil fuel ash with concrete residue in the brick specimens. The hump shape from 20° (2θ) to 40° (2θ) was transferred from a smooth flow to a sharper shape when the concrete residue increased. It can be concluded that the additional concrete residue increased the geopolymer gel and the degree of crystallinity (Richardson *et al.*, 1994). The C-S-H gel was observed in all batches of geopolymer bricks, with one peak in PA100 and two peaks in PA90CR10, PA80CR20, PA70CR30 and PA60CR40.

3.2.6 FTIR patterns of geopolymer bricks

Figure 8 illustrates the comparison of the function groups for the palm oil fuel ash, the concrete residue and the geopolymer brick specimens for all batches. The FTIR measured for the raw materials showed a different O-H peak. The concrete residue had a higher band intensity for the O-H groups than the palm oil fuel ash, demonstrating that there were more nonhydrated silicates available in the palm oil fuel ash than in the concrete residue (Salih *et al.*, 2015).

A peak of Si-O bonds was found between 900 and 1000 cm⁻¹ (Yusuf et al., 2014). As can be seen from Figure 9, the FTIR of single and binary mixtures of palm oil fuel ash and concrete residue specified the formation of a new phase assemblage through changes in the shape and wave number of the Si-O and O-H stretching vibration bands (Lee and Deventer, 2003). The geopolymerization reaction decreased the wave number of the Si-O band when the concrete residue increased. The Si-O band wave numbers of raw palm oil fuel ash and PA100 shifted to 1087.28 cm⁻¹ and 1026.97 cm⁻¹, respectively. For the geopolymer bricks produced from palm oil fuel ash and concrete residue, the Si-O bands centered at 1026.91 cm⁻¹ (for PA90CR10), 1014.93 cm⁻¹ (PA80CR20), 1011.47 cm⁻¹ (PA70CR30) and 1019.80 cm⁻¹ (PA60CR40).



Figure 8. FTIR patterns of geopolymer bricks



Figure 9. Distribution of environmental impact of geopolymer brick made from palm oil fuel ash and concrete residue with the optimal ratio of 60:40

3.3 Life Cycle Assessment

As shown in Figure 9, the main cause of climate change is raw material acquisition, at 61.67%, due to fuel consumption, specifically diesel combustion, and air pollutants caused by the transportation of palm oil fuel ash from Eastern Palm Oil Company, Chonburi province, to the work site at the Department of Environmental Science, Chulalongkorn University in Bangkok (118.73 kilometers). The main cause of ozone depletion is the preparing stage, at 50.01%, due to electricity consumption from the grinding and sieving machines and drying oven. The main cause of human toxicity is the preparing stage, at 40.02%, due to electricity consumption from the grinding and sieving machines and drying oven. The main cause of terrestrial acidification is raw material acquisition, at 95.22%, due to fuel consumption, specifically diesel combustion, and air pollutants caused by the transportation of palm oil fuel ash from Eastern Palm Oil Company, Chonburi, to the work site at the Department of Environmental Science, Chulalongkorn University (118.73 km). The main cause of terrestrial ecotoxicity is the preparing stage, at 50.01%, due to electricity consumption from the grinding and sieving machines and drying oven. The main cause of fossil depletion is the preparing stage, at 49.52%, due to electricity consumption from the grinding and sieving machines and drying oven.

Furthermore, when comparing the geopolymer bricks from this study and other brick types from the study of De Souza et al. (2016) related to climate change, the results show that geopolymer bricks made from palm oil fuel ash and concrete residue with the optimal ratio 60:40 had the impact on climate change, at 18.49%, compared to a concrete brick wall, at nearly 70%. Geopolymer brick had a lower impact on climate change than concrete brick. Geopolymer brick can reduce environmental problems in terms of greenhouse gas emissions, especially carbon dioxide (CO_2) and sulfur dioxide (SO_2) , in the atmosphere, which is a source of global warming potential, climate change, and acidification; thus, the geopolymerization technique is an alternative in brick production in terms of environmentally friendly.

3.4 Economical Feasibility

An evaluation of the economical feasibility in terms of the break even volume (with N* indicating the point at which the total cost and total revenue are equal and the payback period, which is the length of time required to recover the cost). The results showed that for the geopolymer bricks made from palm oil fuel ash and concrete residue, the break even volume was 577,428 pieces, indicating the point at which total cost and total revenue are equal. The payback period was 3.88 years (46.5 months).

4. Conclusions

This study investigated the role of calcium from concrete residue in geopolymer bricks from palm oil fuel ash. The study hypothesized that the addition of concrete residue into palm oil fuel ash could increase the compressive strength of geopolymer brick. The results of this study can be summarized as follows:

- Palm oil fuel ash had a high amount of silica and alumina, known as an aluminosilicate source, and concrete residue had a high amount of calcium and silica, which are important elements in forming a binder. These results show that both materials can be used as raw materials for geopolymer brick production.
- Concrete residue can fill in the pores of the structure to enhance the compressive strength and water absorption, because the particle size of concrete residue is smaller than that of palm oil fuel ash.
- The replacement of palm oil fuel ash by concrete residue in the brick increased the compressive strength and decreased the water absorption of all batches. When 40% of the concrete residue was replaced in the geopolymer brick, the compressive strength increased up to 12.53 MPa, and the percent of water absorption was 0.25%.
- Water absorption and compressive strength of all batches of geopolymer bricks satisfied the TIS 77-2545 specification.
- Increasing the calcium by adding concrete residue to the reaction allowed the formation of a C-S-H gel, which caused a higher degree of polymerization. Si O decreased when the concrete residue was increased, because silica was used to form a C-S-H gel with dissolved calcium from the concrete residue. The formation of the C-S-H gel depended on the volume of calcium and silica in reaction.
- Geopolymer bricks made from palm oil fuel ash and concrete residue with the optimal ratio 60:40 had the impact on climate change, at 18.49%.

- The break even volume was 577,428 pieces, and the payback period was 3.88 years (46.5 months).

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