



Research article

Bio-waste-based lightweight cement blocks with antibacterial performance

Latda Chandeng^{a,b}, Khotamy Saphongxay^{a,b}, Gasidit Panomsuwan^{a,b}, Chakrapan Tuakta^c,
Patcharaporn Siwayaprahm^d, Oratai Jongprateep^{a,b,*}

^a Department of Materials Engineering, Faculty of Engineering, Kasetsart University, Bangkok 10900, Thailand

^b ICE-Matter Consortium, ASEAN University Network/Southeast Asia Engineering Education Development Network (AUN/SEED-Net), Bangkok 10330, Thailand

^c Department of Civil Engineering, Faculty of Engineering, Kasetsart University, Bangkok 10900, Thailand

^d Department of Microbiology, Faculty of Science, Kasetsart University, Bangkok 10900, Thailand

Article Info

Article history:

Received 11 April 2021

Revised 3 August 2021

Accepted 20 December 2021

Available online 9 February 2022

Keywords:

Antibacterial,

Bio-waste,

Cement-like material,

Lightweight cement blocks

Abstract

Importance of the work: Utilization of bio-waste-based construction materials is an effective route to create a more sustainable ecosystem.

Objectives: This research investigated fabricating lightweight cement blocks prepared from bio-wastes. Additionally, the bio-waste-based cement blocks were tailored to include antibacterial properties.

Materials & Methods: The synthetic cement-like material was prepared using a solution combustion technique, with cockleshells and rice husk ash as the key raw materials. Rice husk ash (20–60 wt%) was added to the cement-like material prior to casting. The density, compressive strength and antibacterial performance of the cement blocks were evaluated.

Results: It was found that increased rice husk ash suppressed the compressive strength. Nevertheless, the average density (1.004 g/cm³) and average compressive strength (3.65 MPa) of all cement blocks were in the acceptable ranges for industrial requirements based on recognized standards, specifically Thai Industrial Standards Institute Type C12 (TISI 2601-2556). Enhancement of the compressive strength by more than 10% could be achieved when jute fibers were added. A colony count method was used to determine antimicrobial performance of the lightweight cement blocks. Addition of the powder obtained from the cement blocks into the Petri plates containing Gram-positive *Staphylococcus aureus* and Gram-positive *Bacillus cereus* indicated successful reductions of more than 99% of *Staphylococcus aureus* and *Bacillus cereus* colonies.

Main finding: Cement-like material, which was successfully synthesized from bio-waste, was used in fabrication of jute fiber-reinforced eco-friendly lightweight cement blocks. In addition to acceptable density and compressive strength, the cement blocks demonstrated antibacterial performance.

* Corresponding author.

E-mail address: fengotj@ku.ac.th (O. Jongprateep)

online 2452-316X print 2468-1458/Copyright © 2021. This is an open access article under the CC BY-NC-ND license (<http://creativecommons.org/licenses/by-nc-nd/4.0/>), production and hosting by Kasetsart University of Research and Development Institute on behalf of Kasetsart University.

<https://doi.org/10.34044/j.anres.2021.56.1.05>

Introduction

Elevated demand in cement production, which contributes 35% of greenhouse gas emissions has resulted in an emerging trend globally for the utilization of environmental-friendly construction materials produced using sustainable and resource-efficient processes (Chakrabarty and Lekhwani, 2016; Maddalena et al., 2018). Bio-waste and organic material residues, such as rice husk ash (RHA), bagasse ash, jute fiber, coconut husk, and cotton stalk and shells, have potential as raw materials in the production of green building materials (Madurwar et al., 2012; Kumar et al., 2016). Abundant waste streams, including the annual production of 230 million t of rice husk ash (RHA) and over 10 million t of aquaculture waste have led to environmental problems and pollution (Ansari and Chen, 1991; Yao et al., 2014; Soltani et al., 2015; Martinez-Garcia et al., 2017; Mo et al., 2018; Mosaberpanah et al., 2020). Hence, the reutilization of such waste reduces the detrimental effects associated with land-filling and alleviates pollution problems (Pappu et al., 2007; Madurwar et al., 2012).

It has been generally established that the main constituents of ordinary Portland cement (OPC) include Alite (C_3S , $3CaO \cdot SiO_2$), Belite (C_2S , $2CaO \cdot SiO_2$), Celite (C_3A , $3CaO \cdot Al_2O_3$), and Felite (C_4AF , $4CaO \cdot Al_2O_3 \cdot Fe_2O_3$). Raw materials containing high contents of silicon and calcium are desired to produce synthetic cement-like material with a chemical composition similar to OPC. Numerous studies have reported that RHA contains over 90 wt% of silica, implying that RHA is a suitable source of silica for cement production (Nehdi et al., 2003; Gastaldini et al., 2007; Korotkova et al., 2016). Shells provide another alternative waste material containing a high calcium content, including cockleshells, mussel shells, oyster shells and hen eggshells (Hart, 2020). It has been reported that cockleshells have similar properties to limestone, containing more than 95 wt% calcium oxide (Boey et al., 2012; Jongprateep et al., 2014; Nordin et al., 2015). Cement-like material can be synthesized from wastes by using robust and eco-friendly solution combustion techniques (Jongprateep et al., 2014).

In addition to a high silica content, RHA can also function as a filler in construction materials. RHA is generally recognized as a pozzolanic material, which can react with calcium hydroxide $Ca(OH)_2$ to form the calcium silicate hydrate (C-S-H) phase that is responsible for enhancing the compressive strength and durability of concrete (Chandrasekhar et al., 2006; Kumar et al., 2016). To further enhance the mechanical properties of concrete or cement blocks, natural fibers can be used as

reinforcement, as the addition of 1.5 weight per volume percent (w/v%) of jute fiber improved the strength by 16% (Islam and Ahmed, 2018; Raval and Patel, 2018; Kesikidou and Stefanidou, 2019).

Along with improvements in the processing and usage of eco-friendly cement-like material, additional cement block characteristics are desired that improve hygiene and sanitation. The long-term use of construction materials generally contributes to the accumulation of bacteria and fungi, which can cause be harmful to human health. The addition of effective antibacterial agents into the construction materials can help to eliminate bio-organisms and to reduce health problems associated with allergy and respiratory track diseases. The addition of titanium dioxide, a renowned photocatalyst which exhibits prominent oxidation processes to sterilize and decompose organic impurity in environment, can be a route to produce construction materials with antibacterial properties (Abdel-Messih et al., 2013; Sangchay and Kaewjang, 2016).

The current study investigated the fabrication of bio-waste-based lightweight cement blocks with antimicrobial properties, using synthetic cement-like material. The properties of density and compressive strength of the cement blocks were examined to confirm satisfactory usage for practical application.

Materials and Methods

Cement-like materials

Synthetic Alite (C_3S , $3CaO \cdot SiO_2$) and Belite (C_2S , $2CaO \cdot SiO_2$) were prepared using the solution combustion technique based on calcium oxide (CaO) from cockleshells and silica (SiO_2) from rice husk ash (RHA; DhebKaset Industrial Co., Ltd). Glycine ($C_2H_5NO_2$, Daejung) was used as the combustion fuel during the synthesis process. The molar ratio of the raw materials and fuel (cockleshells:RHA:glycine) used in the preparation of the combusting solution was 0.848:0.392:0.494.

To produce Celite (C_3A), cockleshells, aluminum nitrate nonahydrate ($Al(NO_3)_3 \cdot 9H_2O$; Daejung) and glycine ($C_2H_5NO_2$; Daejung) were mixed at the molar ratio of cockleshells:aluminum nitrate:glycine equal to 0.717:2.774:0.554.

Felite (C_4AF) was synthesized by preparing an aqueous solution consisting of cockleshells, aluminum nitrate nonahydrate ($Al[NO_3]_3 \cdot 9H_2O$; Daejung), ferric oxide red 95% (Fe_2O_3 ; Lobachemie), and glycine ($C_2H_5NO_2$, Daejung) with the molar ratio of 0.808:2.357:0.503:0.234, respectively.

The aqueous solutions containing the raw materials were diluted in deionized water to achieve 0.1 M concentration and subsequently heated at 400°C. Combustion reactions were generally initiated within 2 hr. The powders obtained from the combustion reactions were collected, ball-milled and calcined at 900°C for 3 hr.

To obtain the cement-like material with a chemical composition similar to ordinary Portland cement (OPC), Alite (C₃S) and Belite (C₂S), Celite (C₃A) and Felite (C₄AF) obtained by solution combustion were mixed at a weight ratio of C₃S:C₃A:C₄AF equal to 0.75:0.13:0.09. To control the hardening rate of the cement paste during the casting, 3 wt% of gypsum was subsequently added to the mixture of C₃S, C₂S, C₃A and C₄AF.

Fabrication of lightweight cement blocks

Lightweight cement blocks were cast in 2.5 cm × 2.5 cm × 2.5 cm acrylic molds. The main components of the cement block were mixed using the cement-like material (20–40 wt%), OPC (20–40 wt%) and RHA (20–60 wt%). Anatase-phase titanium dioxide (TiO₂) and aluminum powder with 1 wt% content were added to the mixture to enhance the antibacterial properties and to create a porous structure within the cement blocks, respectively.

To improve the mechanical properties of the cement blocks, 1.5 (w/v%) of jute fibers were added as reinforcement. All components were mixed and cast at a water-to-binder ratio of 0.9:1. The cement blocks were cured for 7 d prior to autoclaving for 6 hr. The varying compositions of the lightweight cement blocks are listed in Table 1.

Characterization

Examination of the chemical composition of the cement-like material was conducted using an X-ray diffractometer (X'Pert; Philips), measuring over 2θ ranging from 10 to 70°. The microstructures of raw materials (OPC, cement-like material, RHA) were examined using a scanning electron microscope (SU3500; Hitachi). The Image J software was used in the determination of particles sizes. The specific gravity values of the OPC, cement-like material, RHA and OPC/RHA/cement-like material mixtures were determined using a pycnometric technique, according to Equation 1:

$$\text{Specific gravity} = \frac{m_2 - m_1}{(m_2 - m_1) - (m_3 - m_4)} \quad (1)$$

where m_1 is the mass of the empty pycnometer, m_2 is the mass of the pycnometer with solid powder, m_3 is the mass of the pycnometer and cement filled with water and m_4 is the mass of the pycnometer filled with water.

Table 1 Compositions of lightweight cement blocks with and without jute fiber

No.	Component of lightweight cement block	Percentage of component by weight	Addition			Water-to-binder ratio
			TiO ₂ (Anatase)	Al (Powder)	Jute fiber (w/v%)	
1	Cement	40 wt%	1 wt%	1 wt%	0	0.9
	Cement-like	40 wt%			1.5	
	Rice husk ash	20 wt%				
2	Cement	35 wt%	1 wt%	1 wt%	0	0.9
	Cement-like	35 wt%			1.5	
	Rice husk ash	30 wt%				
3	Cement	30 wt%	1 wt%	1 wt%	0	0.9
	Cement-like	30 wt%			1.5	
	Rice husk ash	40 wt%				
4	Cement	25 wt%	1 wt%	1 wt%	0	0.9
	Cement-like	25 wt%			1.5	
	Rice husk ash	50 wt%				
5	Cement	20 wt%	1 wt%	1 wt%	0	0.9
	Cement-like	20 wt%			1.5	
	Rice husk ash	60 wt%				

w/v% = weight per volume percent

The tests related to setting time of cementitious materials were conducted following the ASTM C191 test procedure (ASTM C191-08, 2008). The cementitious mixtures used in the determination of the setting time were mixture No.1 and mixture No.5 that contained RHA for 20 wt% and 60 wt%, respectively.

The compressive strength of the cement blocks (after curing for 7 days in water and then autoclaving for 6 hr) was determined using a universal testing machine (H50KS; Hounsfield).

Determination of antibacterial performance

The antibacterial performance of the lightweight cement blocks was evaluated. The fiber-reinforced and unreinforced lightweight cement blocks with TiO_2 were prepared in powder form for the test. The control experiment used the cement-blocks without the addition of TiO_2 .

Single colonies of *Staphylococcus aureus* strain (ATCC6538) and *Bacillus cereus* strain (BCC 6386) were separately prepared by streaking on culture medium agar plates containing tryptic soy agar (TSA) and incubating the agar plates overnight at 37°C. Subsequently, each prepared colony was mixed with tryptic soy broth and shaken overnight at 37°C. Saline was added to the incubated bacterial strains to obtain the diluted bacterial suspension of 1.510^8 CFU/mL (0.5 McFarland turbidity standard). To prepare cement powders for the test, the cement powders (0.5 g) were mixed with TSA in another culture plate, which was subsequently heated at 121°C for 15 min. Then, 0.1 mL of the diluted bacterial suspension was painted on the surface of the TSA medium containing cement powder and incubated overnight at 37°C. The number of bacterial colonies were counted and used in further calculations.

Results and Discussion

Characteristics of raw materials

A scanning electron microscope was used to examine the morphology and particle sizes of the raw materials, as shown in Fig. 1. Image analysis of the micrographs revealed irregular shaped OPC and RHA particles with average sizes (\pm SD) of $6.01 \pm 4.53 \mu\text{m}$ and $4.44 \pm 2.90 \mu\text{m}$, respectively. The particles sizes obtained from this study were finer compared to those obtained by other studies, as shown in Table 2. Very fine particles were evident in the cement-like material; however, the particles clustered into larger agglomerates with an average size of $7.47 \pm 5.45 \mu\text{m}$.

The average specific gravity values of OPC and RHA were 3.23 and 2.07, respectively. The results obtained from this study were in the range comparable to those obtained by other researchers, as shown in Table 2.

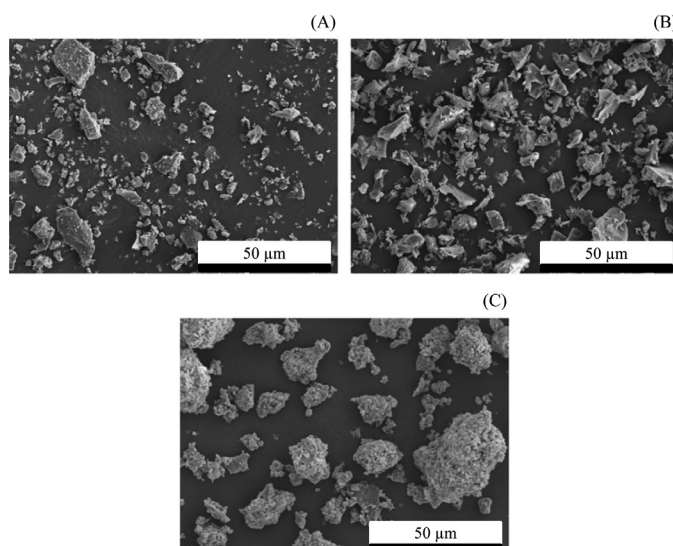


Fig. 1 Morphology of raw materials (A) ordinary Portland cement, (B) rice husk ash; (C) cement-like material

Table 2 Specific gravity and particles size of raw materials

Raw Material	Property	References			
		Deepa et al. (2013)	Habeeb and Fayyadh (2009)	Chao-Lung et al. (2011)	This work (mean \pm SD)
Cement	Specific gravity	2.93	2.94	3.15	3.23 ± 0.02
	Particle size (μm)	-	22.1	-	6.01 ± 4.53
Rice husk ash	Specific gravity	2.04	2.11	2.06	2.07 ± 0.02
	Particle size (μm)	8–10	11.5–31.3	-	4.44 ± 2.90
Cement-like	Specific gravity	-	-	-	2.42 ± 0.01
	Particle size (μm)	-	-	-	7.47 ± 5.45

Characteristics of cement-like materials

It is generally accepted that the main constituents of OPC include Alite, Belite, Celite and Felite (Enríquez et al., 2020; Plank, 2020; Nari and Praneeth, 2021). To obtain cement-like material with a chemical composition similar to OPC, the synthesized Alite, Belite, Celite and Felite were mixed together. Phase identification of the synthetic cement-like material was examined using X-ray diffraction (XRD). As shown in Fig. 2, the XRD pattern of the synthesized cement-like powder indicated prominent peaks corresponding to Alite (Ca_3SiO_5 , JCPDS card no. 00-031-0301), Belite (Ca_2SiO_4 , JCPDS card no. 00-029-0369), Celite ($\text{Ca}_3\text{Al}_2\text{O}_6$, JCPDS card no. 00-008-0005) and Felite ($\text{Ca}_4\text{Al}_2\text{Fe}_2\text{O}_{10}$, JCPDS card no. 00-011-0124). Low intensity peaks were also observed representing calcium oxide (CaO, JCPDS card no. 01-077-2010), as the presence of calcium oxide in OPC is common (Amin et al., 2008 Rukzon and Chindaprasirt, 2008). Additionally, the results agreed with studies by Amin et al. (2008) and Jongprateep et al. (2014).

Characteristics of cementitious mixtures

The cementitious mixtures consisting of various contents of OPC, RHA and cement-like material were tested for their specific gravity and setting time. The average values of specific gravity of all mixtures ranged from 2.35 to 2.61, as shown in Table 3. As the RHA content increased, the specific gravity decreased due to the low specific gravity of the RHA.

The setting behavior were tested of the cementitious mixtures with compositions of 40 wt% OPC + 20 wt% RHA + 40 wt% cement-like material and 20 wt% OPC + 60 wt% RHA + 40 wt% cement-like material at a water-to-binder ratio of 0.9. There was a prolonged initial setting time of the cementitious mixtures with increasing RHA content. The mixture with 20 wt% RHA had a setting time of 7 hr and 30 min, whereas the setting time of the mixture containing 60 wt% RHA was 13 hr and 45 min.

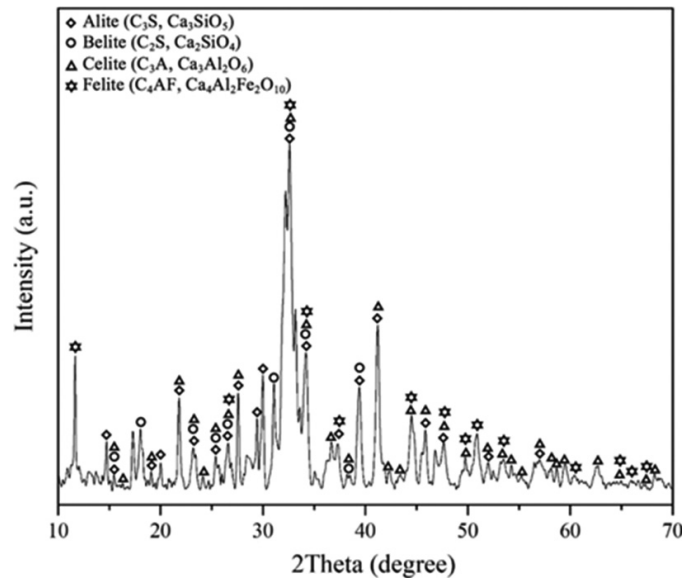


Fig. 2 XRD pattern of synthetic cement-like material

Table 3 Specific gravity values of cementitious mixtures

Material	Component and percentage by weight		Specific gravity (mean \pm SD)
Mixture No.1	Cement	40 wt%	2.61 \pm 0.01
	Cement-like	40 wt%	
	Rice husk ash	20 wt%	
Mixture No.2	Cement	35 wt%	2.58 \pm 0.01
	Cement-like	35 wt%	
	Rice husk ash	30 wt%	
Mixture No.3	Cement	30 wt%	2.53 \pm 0.01
	Cement-like	30 wt%	
	Rice husk ash	40 wt%	
Mixture No.4	Cement	25 wt%	2.38 \pm 0.02
	Cement-like	25 wt%	
	Rice husk ash	50 wt%	
Mixture No.5	Cement	20 wt%	2.35 \pm 0.01
	Cement-like	20 wt%	
	Rice husk ash	60 wt%	

A higher water content is generally required for fine-particle cementitious materials to achieve proper workability, since fine particles sizes lead to enhanced surface area that requires a greater water content (Givi et al., 2010; Fapohunda et al., 2017). With the high water content, the setting time of cement mixture increased (Stefanou and Larsinos, 1981).

In the current study, the cementitious mixtures contained RHA and cement-like material with very fine particle sizes. Therefore, a high water-to-cement ratio was required to achieve workability. The higher the water content, the longer the setting time. In addition, RHA might have contributed to a low rate of hydration, which consequently resulted in a slow initial setting rate. A longer initial setting time was evident in the cementitious mixture containing a great amount of RHA. A similar trend (increasing the RHA content prolonging the initial setting time) was also observed by El-Dakrouy and Gasser (2008) and Dabai et al. (2009).

Characteristics and properties of lightweight cement blocks

The fiber-reinforced lightweight cement blocks were prepared by casting the mixture containing synthetic cement-like material, OPC, RHA, aluminum, titanium dioxide and jute fibers into 2.5 cm × 2.5 cm × 2.5 cm cubes. To ensure that the cement blocks were suitable for practical usage, their density and compressive strength were evaluated and compared with industrial standards.

Bulk density of eco-friendly lightweight cement blocks

The effects of jute fiber and RHA addition on the density of the lightweight cement blocks are shown in Fig. 3. For the cement blocks containing natural jute fiber at 1.5 w/v%, the density values were in the range 0.91–1.14 g/cm³. Without the jute fiber, the bulk density was slightly lower in the range 0.90–1.10 g/cm³. Regardless of whether any jute fiber had been added, the density values of the cement blocks were within the standard range for Type C12 set by the Thai Industrial Standards Institute (TISI 2601-2556, TIS 2601, 2003), which requires a density lower than 1.20 g/cm³ for lightweight concrete. Fig. 3 also reveals that the density was less with incremental RHA content. This could be attributed to the lower specific gravity of the RHA (2.07) compared to that of OPC (3.23). The effects of RHA addition on the weight reduction of the concrete or cement blocks have been observed generally (Singh, 2018).

Compressive strength of eco-friendly lightweight cement blocks

The effects of jute fiber and RHA addition on the compressive strength of the lightweight cement blocks are demonstrated in Fig. 4. The lightweight cement blocks without jute fiber addition had compressive strengths in the range 3.24–4.35 MPa. With the jute fiber reinforcement, the compressive strength of the cement blocks was enhanced by more than 10%, with the values in the range 3.30–4.90 MPa. Both these compressive strength ranges were within the specified range of not lower than 2.50 MPa standard set by the Thai Industrial Standards Institute (TISI 2601-2556, TIS 2601, 2003) for Type C12 lightweight concrete blocks and American Concrete Institute (ACI 213, ACI Committee 213, 2003).

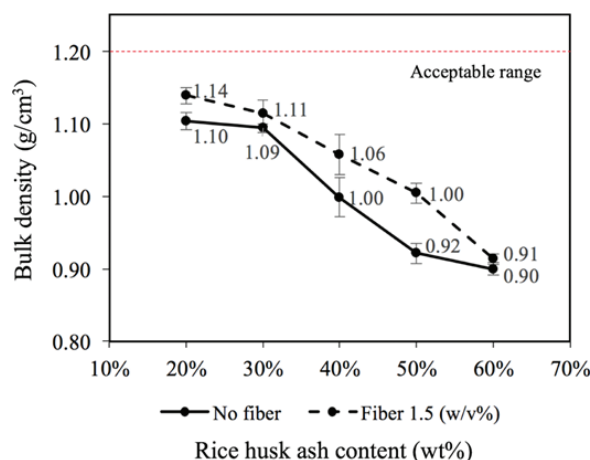


Fig. 3 Relationship between bulk density and rice husk ash content with or without 1.5% weight per volume (w/v%) of jute fiber, where error bars indicate mean ± SD

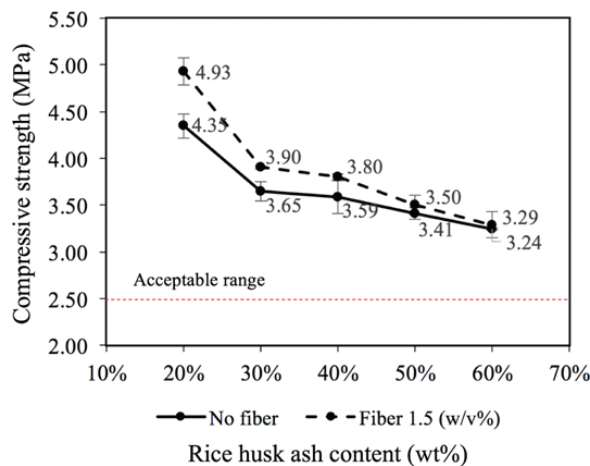


Fig. 4 Relationship between rice husk ash content and addition of jute fiber at 1.5% weight per volume (w/v%) on compressive strength, where error bars indicate mean ± SD

In addition to enhancing the compressive strength, the jute fibers substantially improved the mechanical properties of the cement blocks in terms of the development of specimen integrity at maximum compressive strength. Reduction of flaking, surface peeling and spalling in the materials was clearly evident, as shown in Fig. 5. According to Kundu and Chakraborty (2018) and Kesikidou and Stefanidou (2019), disintegration of mortar without fiber reinforcement was commonly observed when the maximum load was applied. Using 1 cm long fibers randomly and uniformly dispersed in the cement matrix enhanced integral adhesion within the lightweight cement blocks due to the crack-bridging capacity of the jute fibers. As shown in Fig. 6, jute fibers can function as bridges, joining the cement matrix and maintaining the integrity of the materials.

In addition to jute fiber addition, the RHA content significantly influenced the compressive stress of the cement blocks. The compressive strength of the cement blocks containing 20 wt% RHA was superior to those containing 60 wt% RHA. In general, the appropriate content of RHA did not have an adverse effect on the compressive strength. This was attributed to the high silica content in the RHA, which is capable of initiating pozzolanic reactions in the cement, as shown in the following chemical reactions (2) and (3):

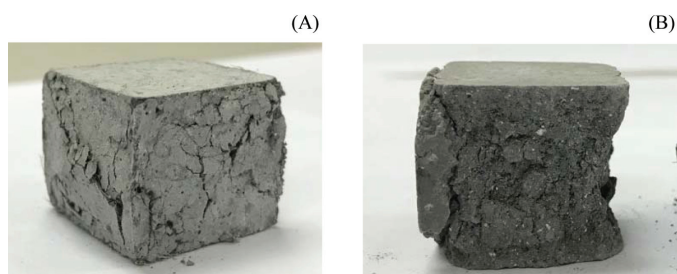
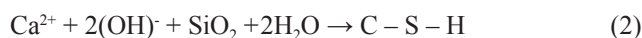


Fig. 5 Photograph of cement block subjected to applied maximum load: (A) with 1.5% weight per volume of jute fiber; (B) without jute fiber

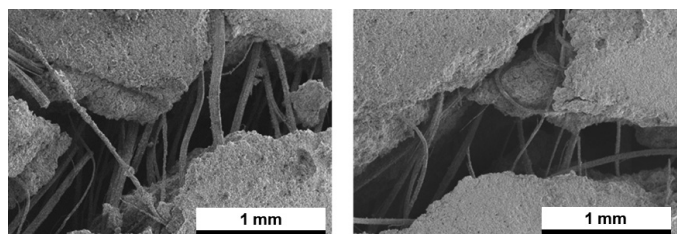
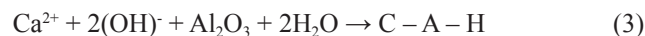


Fig. 6 Scanning electron micrographs showing crack bridging in fiber-reinforced lightweight cement blocks under maximum compressive stress



However, an excessively high RHA content could result in diminished compressive strength due to insufficient cement content. It is generally accepted that cementitious materials also function as binder (Ahsan and Hossain, 2018; Ali et al., 2021). With a considerably high RHA content, low connectivity of the cement matrix was evident, resulting in diminished strength.

Antibacterial property of lightweight cement block

The lightweight cement blocks containing RHA of 20 wt%, 30 wt%, 40 wt%, 50 wt% and 60 wt% with and without jute fiber reinforcement were tested for their antibacterial performance. All the cement blocks (except for the control) contained 1 wt% of anatase phase TiO_2 with an average particle size smaller than 20 nm (Jongprateep et al., 2018). Two types of bacteria were used for the colony count test (Gram-positive *Staphylococcus aureus* and Gram-positive *Bacillus cereus*).

TiO_2 is used ubiquitously as a photocatalyst in applications, such as sensors and solar cells (Tang et al., 1995; Karunagaran et al., 2007; Subramaniam et al., 2017; Akila et al., 2019). Wastewater treatment and disinfection are among prominent applications of TiO_2 . Fig. 7 illustrates some key mechanisms related to antibacterial activities that are attributed to the inhibition of cell wall synthesis, the inhibition of nucleic acid (RNA/DNA) synthesis, the inhibition of vital enzyme synthesis and damage to the membrane and cell wall, as well as disruption of microbial respiration (Erdem et al., 2015). It has been reported that significant reductions occur in microbial activities related to DNA, protein and enzyme synthesis in bacteria in contact with TiO_2 (Liou and Chang, 2012; Erdem et al., 2015).

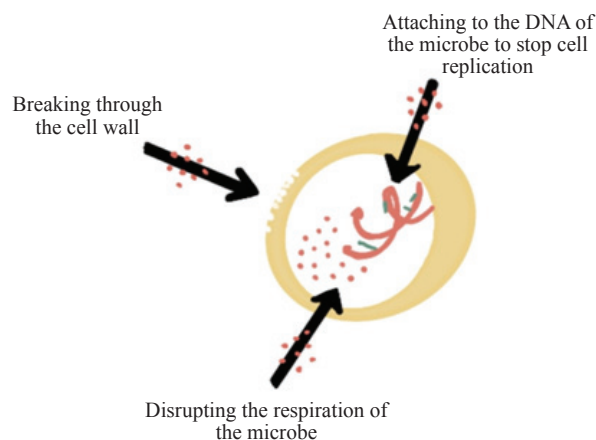


Fig. 7 Schematic representation of some key mechanisms related to bacterial inactivation

The photocatalytic activities of the TiO_2 are credited as one of the key attributes to antimicrobial performance, as exposure of TiO_2 to ultraviolet radiation leads to the generation of light-induced electrons and holes, which react with oxygen and water and lead to the production of reactive oxidizing species (ROS), as shown in the schematic in Fig. 8 (Herrmann, 1999). The ROS effectively degrade inorganic substances and efficiently inhibit bacterial growth by destroying the proteins of the outer layer cells of bacteria (Azizi-Lalabadi et al., 2019).

In addition to damaging the bacterial outer layer cells, nanoparticulate metal oxides are reported to prevent bacterial duplication and to inhibit the synthesis of vital metabolic enzymes in bacteria, leading to microbial inactivation (Holt and Bard, 2005; Heinlaan et al., 2008; Azizi-Lalabadi et al., 2019).

S. aureus and *B. cereus* are Gram-positive pathogenic bacteria. *S. aureus* generally causes skin infection and food poisoning in humans and in some severe cases, multiple exotoxins may occur, leading to critical effects such as shock, organ failure and death (Dinges et al., 2000; Bottone, 2010). It has been reported that diarrheal syndrome and chronic skin infection can be triggered by *B. cereus* (Prod'hom and Bille, 2010; Sangchay and Kaewjang, 2016).

Effect of TiO_2 on *Staphylococcus aureus*

The effects of the TiO_2 -added lightweight cement blocks on suppressing the growth of *S. aureus* were evaluated based on the colony counting method. The powders were tested of lightweight cement blocks containing rice husk ash (20 wt%, 30 wt%, 40 wt%, 50 wt% and 60 wt%) and 1 wt% of TiO_2 (anatase), with and without jute fiber reinforcement.

A macroscopic image of *S. aureus* in a Petri plate without the addition of cement powder (Fig. 9A) reveals large quantities of *S. aureus* colonies, whereas the microscopic image of *S. aureus* (Fig. 9B) presents a round-shaped bacterium.

For the control plate, *S. aureus* was cultured in the presence of cement powder without the addition of TiO_2 . A macroscopic image of *S. aureus* in the control Petri plate (Fig. 10) still shows a number of *S. aureus* colonies. Nevertheless, the numbers of Gram-positive *S. aureus* colonies were reduced.

As powder from the TiO_2 -added lightweight cement blocks was added, it was apparent that Gram-positive *S. aureus* colonies were reduced to a substantial degree. No bacterial colony was observed in all the formulations tested, as shown in Figs. 11 and 12.

Effect of TiO_2 on *Bacillus cereus*

In the absence of cement powder, the Petri plates containing *B. cereus* (Fig. 13A) revealed bacterial colonies with sizes slightly larger than those for *S. aureus*. The microscopic image of *B. cereus* (Fig. 13B) shows a rod-shaped bacterium.

The results regarding the effects of TiO_2 -added lightweight cement blocks on the formation of *B. cereus* colonies are demonstrated in Figs. 14 and 15. Regardless of whether the jute fibers were used, all lightweight cement blocks containing TiO_2 exhibited excellent antimicrobial performance. No Gram-positive *B. cereus* colonies were observed in the presence of powder from the lightweight cement block added with TiO_2 .

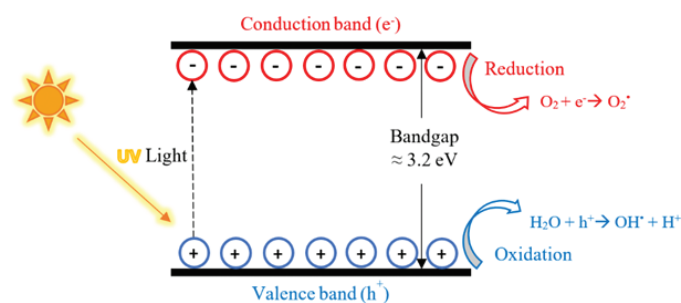


Fig. 8 Schematic representation of photocatalytic activities

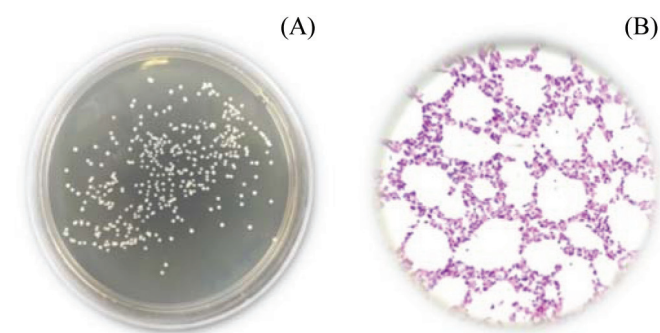


Fig. 9 (A) Macroscopic image of *Staphylococcus aureus*; (B) microscopic image of *Staphylococcus aureus* without added cement powder

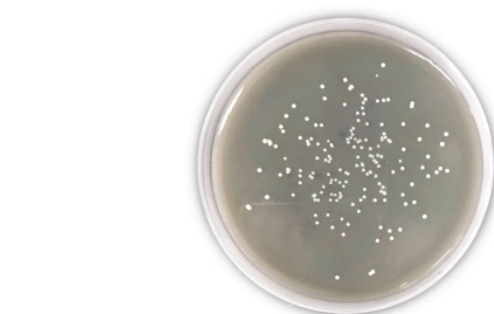


Fig. 10 Macroscopic image of *Staphylococcus aureus* in presence of cement powder without TiO_2 addition

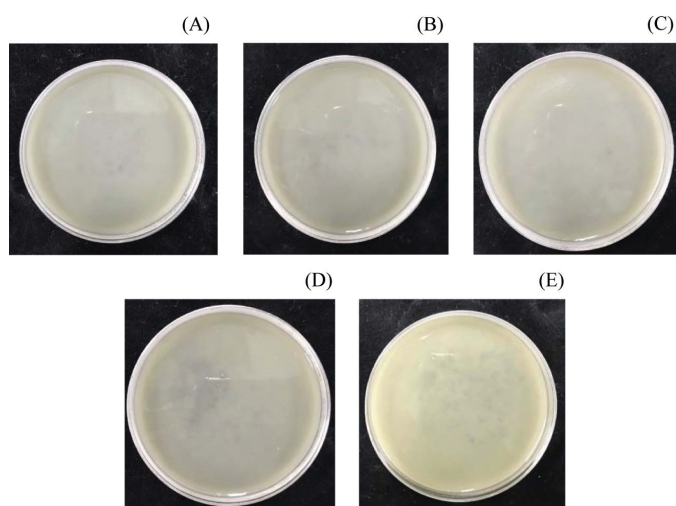


Fig. 11 Antibacterial performance tested using *Staphylococcus aureus* of lightweight cement blocks containing 1 wt% of TiO_2 and rice husk ash at: (A) 20 wt%; (B) 30 wt%; (C) 40 wt%; (D) 50 wt%; (E) 60 wt%

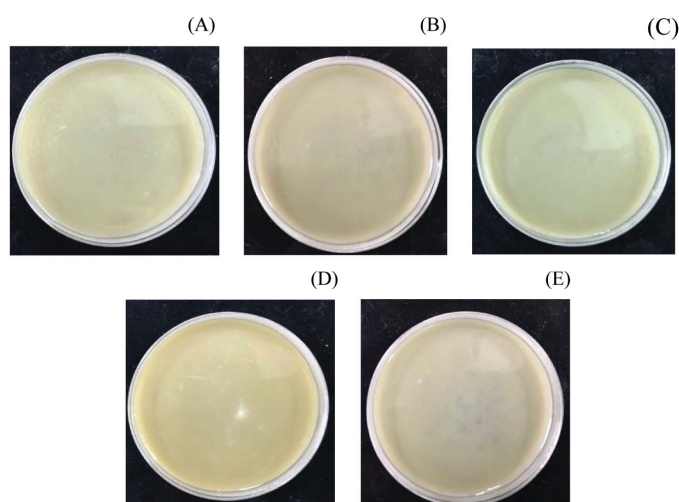


Fig. 12 Antibacterial performance tested using *Staphylococcus aureus* of jute fiber-reinforced lightweight cement blocks containing with 1wt% of TiO_2 and rice husk ash at: (A) 20 wt%; (B) 30 wt%; (C) 40 wt%; (D) 50 wt%; (E) 60 wt%

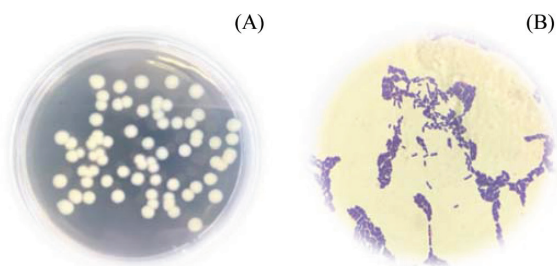


Fig. 13 Macroscopic image: (A) *Bacillus cereus* control plate; (B) microscopic image of *Bacillus cereus* using an optical microscope in absence of cement powder

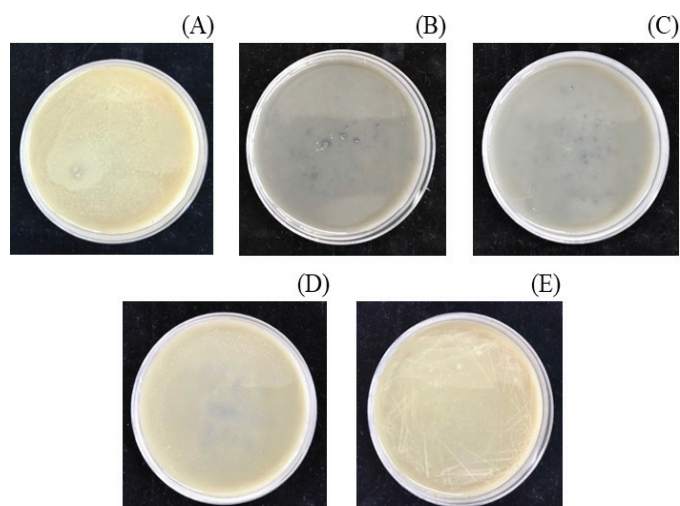


Fig. 14 Antibacterial performance tested using *Bacillus cereus* of lightweight cement blocks containing with 1wt% TiO_2 and rice husk ash at: (A) 20 wt%; (B) 30 wt%; (C) 40 wt%; (D) 50 wt%; (E) 60 wt%

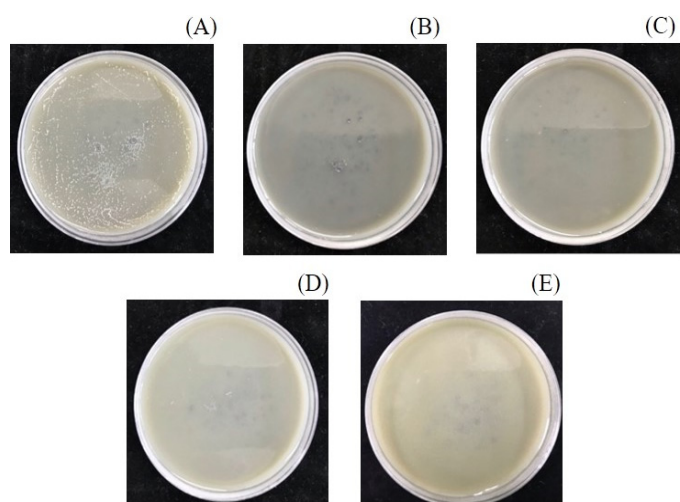


Fig. 15 Antibacterial performance tested using *Bacillus cereus* of jute fiber-reinforced lightweight cement blocks containing with rice husk ash at: (A) 20 wt%; (B) 30 wt%; (C) 40 wt%; (D) 50 wt%; (E) 60 wt%

Conclusion

This study demonstrated the potential utilization of bio-waste in the fabrication of lightweight cement blocks. Cockleshells and RHA were used as the main raw materials in the production of cement-like material consisting of Alite ($3\text{CaO}\cdot\text{SiO}_2$), Belite ($2\text{CaO}\cdot\text{SiO}_2$), Celite ($3\text{CaO}\cdot\text{Al}_2\text{O}_3$), and Felite ($4\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot\text{Fe}_2\text{O}_3$). In addition, rice husk ash was utilized as filler in the lightweight cement blocks. The lightweight cement blocks containing rice husk ash

(20–60 wt%), synthetic cement-like material (20–40 wt%), OPC (20–40 wt%) and TiO₂ (1 wt%) had compressive strengths and bulk densities in the acceptable ranges based on the Thai Industrial Standards Institute Type C12 (TISI 2601-2556) and the American Concrete Institute (ACI 213, 2001). The lowest content of rice husk ash (20 wt%) produced the maximum compressive strength and density of 4.90 MPa and 1.14 g/cm³, respectively. Further strength enhancement, as well as elimination of material disintegration at high applied load, were achieved when jute fibers were used as reinforcement. The antimicrobial performance of all the lightweight cement blocks was confirmed by the total elimination of *Staphylococcus aureus* and *Bacillus cereus*.

Conflict of Interest

The authors declare that there are no conflicts of interest.

Acknowledgements

The ASEAN University Network/Southeast Asia Engineering Education Development Network (AUN/SEED-Net) and ICE-Matter Consortium provided financial support. The Department of Materials Engineering, Faculty of Engineering, Kasetsart University, Bangkok, Thailand provided support related to characterization facilities. DhebKaset Industrial Co., Ltd provided the rice husk ash.

References

- Abdel-Messih, M.F., Ahmed, M.A., El-Sayed, A.S. 2013. Photocatalytic decolorization of rhodamine B dye using novel mesoporous SnO₂-TiO₂ nano mixed oxides prepared by sol-gel method. *J. Photochem. Photobiol.* 260: 1–8. doi.org/10.1016/j.jphotochem.2013.03.011
- ACI Committee 213. 2003. Guide for structural lightweight-aggregate concrete (ACI 213R-03). In: *ACI Manual of Concrete Practice*, American Concrete Institute. Farmington Hill, MI, USA, pp. 213R-1–213R-38.
- Ahsan, M.B., Hossain, Z. 2018. Supplemental use of rice husk ash (RHA) as a cementitious material in concrete industry. *Constr. Build. Mater.* 178: 1–9. doi.org/10.1016/j.conbuildmat.2018.05.101
- Akila, Y., Muthukumarasamy, N., Velauthapillai, D. 2019. TiO₂-based dye-sensitized solar cells. In: *Nanomaterials for Solar Cell Applications*. Elsevier. Amsterdam, the Netherlands, pp. 127–144.
- Ali, T., Saand, A., Bangwar, D.K., Buller, A.S., Ahmed, Z. 2021. Mechanical and durability properties of aerated concrete incorporating rice husk ash (RHA) as partial replacement of cement. *Crystals* 11: 604. doi.org/10.3390/cryst11060604
- Amin, M.M., Jamaludin, S.B., Pa, F.C., Chuen, K.K. 2008. Effects of magnesium sulfate attack on Ordinary Portland Cement (OPC) mortars. *Port. Electrochim. Acta* 26: 235–242
- Ansari, F., Chen, Q.Y. 1991. Fiber-optic refractive-index sensor for use in fresh concrete. *Appl. Opt.* 30: 4056–4059. doi.org/10.1364/AO.30.004056
- ASTM C191-08. 2008. Standard Test Methods for Time of Setting of Hydraulic Cement by Vicat Needle. ASTM International, West Conshohocken, PA, USA. doi.org/10.1520/C0191-08
- Azizi-Lalabadi, M., Ehsani, A., Divband, B., Alizadeh-Sani, M. 2019. Antimicrobial activity of titanium dioxide and zinc oxide nanoparticles supported in 4A zeolite and evaluation the morphological characteristic. *Sci. Rep.* 9: 17439. doi.org/10.1038/s41598-019-54025-0
- Boey, P.L., Ganesan, S., Maniam, G.P., Khairuddean, M. 2012. Catalysts derived from waste sources in the production of biodiesel using waste cooking oil. *Catal. Today*. 190: 117–121. doi.org/10.1016/j.cattod.2011.11.027
- Bottone, E.J. 2010. *Bacillus cereus*, a volatile human pathogen. *Clin. Microbiol. Rev.* 23: 382–398. doi.org/10.1128/CMR.00073-09
- Chakrabarty, M., Lekhwani, N. 2016. Green building materials market-growth, trends and opportunity: South Asian perspective. *Int. J. Environ. Sci. Dev.* 7: 278–284. doi.org/10.7763/IJESD.2016.V7.784
- Chandrasekhar, S., Pramada, P.N., Majeed, J. 2006. Effect of calcination temperature and heating rate on the optical properties and reactivity of rice husk ash. *J. Mater. Sci.* 41: 7926–7933. doi.org/10.1007/s10853-006-0859-0
- Chao-Lung, H., Anh-Tuan, B.L., Chun-Tsun, C. 2011. Effect of rice husk ash on the strength and durability characteristics of concrete. *Constr. Build. Mater.* 25: 3768–3772. doi.org/10.1016/j.conbuildmat.2011.04.009
- Enríquez, M.K., Tobón, J.I., Ramírez, J.H. 2020. Use of industrial wastes for the synthesis of belite clinker. *Mater. Construcc.* 70: 226. doi.org/10.3989/mc.2020.14219
- Dabai, M.U., Muhammad, C., Bagudo, B.U., Musa, A. 2009. Studies on the effect of rice husk ash as cement admixture. *Nig. J. Basic Appl. Sci.* 17: 252–256. doi.org/10.4314/njbas.v17i2.49917
- Deepa, N.G., Sivaraman, K., Job, T. 2013. Mechanical properties of rice husk ash (RHA)-high strength concrete. *Am. J. Eng. Res.* 3: 14–19.
- Dinges, M.M., Orwin, P.M., Schlievert, P.M. 2000. Exotoxins of *Staphylococcus aureus*. *Clin. Microbiol. Rev.* 13: 16–34. doi.org/10.1128/CMR.13.1.16
- El-Dakroury, A., Gasser, M.S. 2008. Rice husk ash (RHA) as cement admixture for immobilization of liquid radioactive waste at different temperatures. *J. Nucl. Mater.* 381: 271–277. doi.org/10.1016/j.jnucmat.2008.08.026

- Erdem, A., Metzler, D., Cha, D.K., Huang, C.P. 2015. The short-term toxic effects of TiO₂ nanoparticles toward bacteria through viability, cellular respiration, and lipid peroxidation. *Environ. Sci. Pollut. Res.* 22: 17917–17924. doi.org/10.1007/s11356-015-5018-1
- Fapohunda, C., Akinbile, B., Shittu, A. 2017. Structure and properties of mortar and concrete with rice husk ash as partial replacement of ordinary Portland cement-A review. *Int. J. Sustain. Built. Environ.* 6: 675–692. doi.org/10.1016/j.ijsbe.2017.07.004
- Gastaldini, A.L.G., Isaia, G.C., Gomes, N.S., Sperb, J.E.K. 2007. Chloride penetration and carbonation in concrete with rice husk ash and chemical activators. *Cem. Concr. Compos.* 29: 176–180. doi.org/10.1016/j.cemconcomp.2006.11.010
- Givi, A.N., Rashid, S.A., Aziz, F.N.A., Salleh, M.A.M. 2010. Contribution of rice husk ash to the properties of mortar and concrete: A review. *J. Am. Sci.* 6: 157–165.
- Habeeb, G.A., Fayyadh, M.M. 2009. Rice husk ash concrete: The effect of RHA average particle size on mechanical properties and drying shrinkage. *Aust. J. Basic Appl. Sci.* 3: 1616–1622.
- Hart, A. 2020. Mini-review of waste shell-derived materials' applications. *Waste Manag. Res.* 38: 514–527. doi.org/10.1177/0734242X19897812
- Heinlaan, M., Ivask, A., Blinova, I., Dubourguier, H.C., Kahru, A. 2008. Toxicity of nanosized and bulk ZnO, CuO and TiO₂ to bacteria *Vibrio fischeri* and crustaceans *Daphnia magna* and *Thamnocephalus platyurus*. *Chemosphere* 71: 1308–1316. doi.org/10.1016/j.chemosphere.2007.11.047
- Herrmann, J.M. 1999. Heterogeneous photocatalysis: Fundamentals and applications to the removal of various types of aqueous pollutants. *Catal. Today.* 53: 115–129. doi.org/10.1016/S0920-5861(99)00107-8
- Holt, K.B., Bard, A.J. 2005. Interaction of silver (I) ions with the respiratory chain of *Escherichia coli*: An electrochemical and scanning electrochemical microscopy study of the antimicrobial mechanism of micromolar Ag⁺. *Biochemistry* 44: 13214–13223. doi.org/10.1021/bi0508542
- Islam, M.S., Ahmed, S.J.U. 2018. Influence of jute fiber on concrete properties. *Constr. Build Mater.* 189: 768–776. doi.org/10.1016/j.conbuildmat.2018.09.048
- Jongprateep, O., Laomarakot, P., Sirinunwatana, K. 2014. Composition and microstructure of cement-like materials synthesized by solution combustion technique. *Adv. Mat. Res.* 1044–1045: 16–22. doi.org/10.4028/www.scientific.net/AMR.1044-1045.16
- Jongprateep, O., Meesombad K., Techapiesanchaenokij, R., Surawathanawises, K. 2018. Chemical composition, microstructure, bandgap energy and electrocatalytic activities of TiO₂ and Ag-doped TiO₂ powder synthesized by solution combustion technique. *Ceram. Int.* 44: 228–232. doi.org/10.1016/j.ceramint.2018.08.108
- Karunakaran, B., Uthirakumar, P., Chung, S.J., Velumani, S., Suh, E.K. 2007. TiO₂ thin film gas sensor for monitoring ammonia. *Mater. Charact.* 58: 680–684. doi.org/10.1016/j.matchar.2006.11.007
- Kesikidou, F., Stefanidou, M. 2019. Natural fiber-reinforced mortars. *J. Build. Eng.* 25: 100786. doi.org/10.1016/j.jobbe.2019.100786
- Korotkova, T.G., Ksandopulo, S.J., Donenko, A.P., Bushumov, S.A., Danilchenko, A.S. 2016. Physical properties and chemical composition of the rice husk and dust. *Orient. J. Chem.* 32: 3213–3219. doi.org/10.13005/ojc/320644
- Kumar, A., Tomar, A.K., Gupta, S.K. 2016. Replacement of cement in concrete with rice husk ash. *SSRG Int. J. Civ. Eng.* 3: 127–129.
- Kundu, S., Chakraborty, S. 2018. Effectiveness of the surface modified jute fibre as fibre reinforcement in controlling the physical and mechanical properties of concrete paver blocks. *Constr. Build Mater.* 191: 554–563. doi.org/10.1016/j.conbuildmat.2018.10.045
- Liou, J.W., Chang, H.H. 2012. Bactericidal effects and mechanisms of visible light-responsive titanium dioxide photocatalysts on pathogenic bacteria. *Arch. Immunol. Ther. Exp.* 60: 267–275. doi.org/10.1007/s00005-012-0178-x
- Maddalena, R., Roberts, J.J., Hamilton, A. 2018. Can Portland cement be replaced by low-carbon alternative materials? A study on the thermal properties and carbon emissions of innovative cements. *J. Clean. Prod.* 186: 933–942. doi.org/10.1016/j.jclepro.2018.02.138
- Madurwar, M.V., Ralegaonkar, R.V., Mandavgane, S.A. 2012. Application of agro-waste for sustainable construction materials: A review. *Constr. Build Mater.* 38: 872–878. doi.org/10.1016/j.conbuildmat.2012.09.011
- Martinez-Garcia, C., Gonzalez-Fonteboa, B., Martinez-Abella, F., Carro-Lopez, D. 2017. Performance of mussel shell as aggregate in plain concrete. *Constr. Build Mater.* 139: 570–583. doi.org/10.1016/j.conbuildmat.2016.09.091
- Mo, K.H., Alengaram, U.J., Jumaat, M.Z., Lee, M.Z., Goh, W.I., Yuen, C.W. 2018. Recycling of seashell waste in concrete: A review. *Constr. Build Mater.* 162: 751–764. doi.org/10.1016/j.conbuildmat.2017.12.009
- Mosaberpanah, M.A., Umar, S.A. 2020. Utilizing rice husk ash as supplement to cementitious materials on performance of ultra high performance concrete: A review. *Mater. Today. Sustain.* 7–8: 100030. doi.org/10.1016/j.mtsust.2019.100030
- Nari, V., Praneeth, P.H. 2021. A comparative study on the thermal behaviour of PPC and OPC cement. *Mater. TodayProc.* 39: 1588–1593. doi.org/10.1016/j.matpr.2020.05.708
- Nehdi, M., Duquette, J., El Damatty, A. 2003. Performance of rice husk ash produced using a new technology as a mineral admixture in concrete. *Cement Concrete Res.* 33: 1203–1210. doi.org/10.1016/S0008-8846(03)00038-3
- Nordin, N., Hamzah, Z., Hashim, O., Kasim, F.H., Abdullah, R. 2015. Effect of temperature in calcination process of seashells. *J. Anal. Sci.* 19: 65–70
- Pappu, A., Saxena, M., Asolekar, S. 2007. Solid wastes generation in India and their recycling potential in building materials. *Build. Environ.* 42: 2311–2320. doi.org/10.1016/j.buildenv.2006.04.015
- Plank, J. 2020. On the correct chemical nomenclature of C₃S, tricalcium oxy silicate. *Cem. Concr. Res.* 130: 105957. doi.org/10.1016/j.cemconres.2019.105957

- Prod'hom, G., Bille, J. 2010. Aerobic gram-positive bacilli. In: Cohen, J., Opal, S.M., Powderly, W.G. (Eds.). *Infectious Diseases*, 3rd ed. London, UK, pp. 1660–1675.
- Raval, G., Patel, U. 2018. Impacts of adding jute fibers to concrete. *Int. J. Adv. Eng. Res. Dev.* 5: 1–8.
- Rukzon, S., Chindapasirt, P. 2008. Use of waste ash from various by-product materials in increasing the durability of mortar. *Songklanakarin J. Sci. Technol.* 30: 485–489.
- Sangchay, W., Kaewjang, S. 2016. SnO₂-doped TiO₂ nanostructured thin films with antibacterial properties. *AIP Conf. Proc.* 1775: 030023. doi.org/10.1063/1.4965143
- Singh, B. 2018. Rice husk ash. In: *Waste and Supplementary Cementitious Materials in Concrete*. Woodhead Publishing, Indian Institute of Technology Roorkee. Roorkee, India, pp. 417–460.
- Soltani, N., Bahrami, A., Peach-Canul, M.I., González, L.A. 2015. Review on the physicochemical treatments of rice husk for production of advanced materials. *Chem. Eng. J.* 264: 899–935. doi.org/10.1016/j.cej.2014.11.056
- Stefanou, G.D., Larsinos, C.H. 1981. Influence of mixing water on the setting time of concrete. *Int. J. Cem. Compos. Lightweight Concr.* 3: 45–48. doi.org/10.1016/0262-5075(81)90022-1
- Subramaniam, M.R., Kumaresan, D., Jothi S., McGettrick, J.D., Watson, T. 2017. Reduced graphene oxide wrapped hierarchical TiO₂ nanorod composites for improved charge collection efficiency and carrier lifetime in dye sensitized solar cells. *Appl. Surf. Sci.* 428: 439–447. doi.org/10.1016/j.apsusc.2017.09.142
- Tang, H., Prasad, K., Sanjinés, R., Lévy, F. 1995. TiO₂ anatase thin films as gas sensors. *Sens. Actuators B Chem.* 26: 71–75. doi.org/10.1016/0925-4005(94)01559-Z
- TIS 2601. 2013. *Cellular Lightweight Concrete Blocks Using Preformed Foam*. Thai Industrial Standard Institute. Bangkok, Thailand.
- Yao, Z., Xia, M., Li, H., Chen, T., Ye, Y., Zheng, H. 2014. Bivalve shell: Not an abundant useless waste but a functional and versatile biomaterial. *Crit. Rev. Environ. Sci. Technol.* 44: 2502–2530. doi.org/10.1080/10643389.2013.8297