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The Effects of Using Ground Cockle Seashells as an Additive for Mortar in Peat Environment

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ABSTRACT Seashells are available abundantly in coastal areas and have the potential to be used as aggregates and replacement for cement in mortar and concrete. They are also applied as mineral additives for mortar or concrete to increase the resistance of these materials in an aggressive environment, especially in constructing structures such as drainage and sewer networks which require good resistance to organic acid attack. This paper discusses the potential addition of ground seashells to improve the performance of mortar used as a drainage lining in an acidic environment such as peatland. The mix was designed using a 4% ground cockle shell (Anadara *granosa*) by cement weight as an additive in two mixes which include Ordinary Portland Cement (OPC) and OPC Cockle Shell (OPCCS). The samples were cured in a water pond for 28 days before they were placed in water and peat water for 120 days after which the compressive strength, porosity, sorptivity, change in weight, and visual characteristics were investigated. The results showed the compressive strength of OPCCS mortar increased by 11.29% after immersion in peat water for 120 days with its porosity and sorptivity decreased by 5.78% and 31.07% due to the refinement of the pores and capillary network in the mortar. Moreover, the weight of the brushed and unbrushed OPCCS mortar in peat water was lesser compared to the OPC due to the increase in CaO content which has the ability to fill the pores and reduce disintegration. The visual examination showed an improvement in the pH of OPCCS mortar due to the ability of the ground cockle shells to neutralize the acidity of the peat water. This study, therefore, shows the use of ground cockle shells as an additive makes it possible to use mortar as a drainage lining because the shells provide excellent resistance to acidic peat environments.

KEYWORDS Acidic Environment; Additive; Cockle Shells; Mortar; Peatland.

1 INTRODUCTION

Seashells or mollusc shells are abundantly available in the Indonesian marine and coastal areas and this was observed in approximately 162,086 tonnes captured as a product of aquaculture for consumption between 2005-2009 (DGCF, 2010). The utilisation of seashell as a value-added product, especially in construction, is observed to be limited despite its large quantity in nature. Meanwhile, the shortage of limestone supply in the past led to the mixture of the quick lime from burnt seashells with clay or hydrated lime and applied as mortar in historic buildings of Santa Catarina-Brazil and Bremen-Germany (Gleize et al. 2019; Marinowitz et al. 2012). Recent studies have also explored its use as aggregates and cement replacement material in mortar and concrete for more viable application (Eo & Yi, 2015; Prusty & Patro, 2015; Ponnada et al. 2016). The shells such as oyster, mussel, cockle, and scallop have been reported to have the ability to replace 20-30% coarse and 20-50% fine aggregates without decreasing the compressive strength of the concrete. Moreover, the use of approximately 20% of seashells powder as an additive in concrete has the ability to increase viscosity, consistency, cement hydration rate, and corresponding strength due to the additional solid content in the mixture (Wang et al. 2019, Wang et al. 2020). The reduction in the strength was, however, reported to be due to the inherent weak shells, surface area, poor bonding between aggregates and cement paste as well as the texture, flaky and a flat shape, and the size of the shells (Richardson & Fuller, 2013; Martinez-Garcia, et al. 2017; Nguyen et al. 2017; Varhen et al. 2017).

Cockle shells or blood clamshells or Anadara *granosa* from South East Asia and other parts of Asian's marine environments are applicable as a cement replacement material. Cockle is a primary mollusc product captured in Indonesia with 38.8% of 236,564 tonnes recovered between 1950-2011 (Kartika, 2014). It contains unreactive calcium carbonate or $CaCO_3$ in the raw state but has aragonite, calcite and carbon to produce quick lime and improve bone mineralisation when burnt (Islam et al. 2011; Paris et al. 2016). According to Ferraz et al. (2018), edible cockle shells (Cerastoderma *edule*) produce more aragonite than calcite even though they are more reactive than other types of shells studied. Kamba et al. (2013) also found cockle shells with more aragonite, in the form of Calcium carbonate, to be dense but unstable. Meanwhile, the ground cockle needs to be used in specific proportion as a cement replacement material to avoid strength reduction due to its ability to become less reactive in high proportion as observed in a study by Lertwattanaruk et al. (2012) which examined the quick setting time and compressive strength reduction by increasing the percentage of cockle shell powder in the mortar. Othman et al. (2013) reported an improvement in the permeation properties, tensile strength and a modulus elasticity of concrete through the use of only 5-10% cockle ash and this is in line with the findings of another research that the optimum ground cockle shells of 4% increased tensile and flexural strength of concrete (Olivia et al. 2015).

Riau Province has one of the largest peatland areas in Indonesia with approximately 3.69 million ha (Ritung et al. 2012). Concrete structures in acidic peat environments are generally prone to the organic acid attack in water and soil and this inhibits, delays, and damages the hydration products of Ordinary Portland Cement (OPC) concrete even at early ages. Therefore, mortar and concrete with high quality, water tightness, corrosion-resistant and which are physically and chemically stable after being exposed in such an environment are required to minimise the deterioration effect of the organic acid attack (Ouslati & Duchesne, 2012). It has, however, been reported that incorporating cementitious materials such as fly ash and palm oil fuel ash has the ability to enhance the cement chemistry and resistance of concrete in the peatland (Olivia et al. 2017; Olivia, et al. 2018). Moreover, silicate material modifies the matrix by increasing Ca/Si ratio in Calcium Silicate Hydrate (C-S-H) but Makhloufi et al. (2014) found the replacement of the cement content with pozzolanic material to have reduced the strength development of the matrix at early ages due to the slow pozzolanic reaction in the short term. Meanwhile, the use of a filler-based Ca material as an additive in mortar and concrete usually provides better resistance and faster strength gain at early ages. Seashells, however, contains more than 90% of calcium carbonate ($CaCO_3$) by weight, which is equivalent to limestone powder used in producing Portland cement, and this makes it a suitable alternative additive (Lertwattanaruk, et al. 2012). A study by Bhatty & Taylor (2006) also reported the inclusion of high Ca content material or metakaolin as additive led to an excellent resistance when exposed in an acidic environment and also improved the properties of concrete at all ages by improving the chemical bonding between cement-aggregates and acting as filler or micro-aggregate in the cement matrix.

This research aims to determine the effect of adding ground cockle shell powder to mortar for drainage lining application in peatland on the compressive strength, porosity, sorptivity, and weight change of the mortar. Previous study showed the addition of calcined oyster shell as an additive to improve early age compressive strength of high-volume slag cement (Naqi et al. 2020) but there are limited studies on the use of material containing Ca such as ground cockle as an additive in concrete or mortar in an acidic environment. Meanwhile, the utilisation of ground cockle shells which are considered biological and natural wastes is beneficial to the strength and durability of concrete in the peatland environment without replacing the cement.

2 MATERIALS AND METHODS

Anadara *granosa* or cockle from Riau Province is a major type of shell consumed in Indonesia and those used in this study were collected as post-consumer products from street vendors. They were cleaned, air-dried, and burnt in a local brick maker stove at a temperature of 500-800^{O}C for three days after which the burnt shells were crushed, ground and sieved to produce fine particles as shown in Figure 1 at the Construction Materials Laboratory, Faculty of Engineering, Universitas Riau. Moreover, Ordinary Portland Cement (OPC) was used as the primary mortar binder and its chemical composition, as well as those of ground cockle shells, are indicated in Table 1. The CaO content of the shells was reported to be 61.09% and this is 6.31% lesser than for OPC while the silicate oxide (SiO_2) content was found to be smaller than the value for OPC by 98.61%. This low SiO_2 content, however, reduces the effectiveness of the shells to function as a cement replacement material. Furthermore, fine aggregates were obtained from Kampar Regency in Riau Province and were observed to have a modulus of fineness at 3.212, water content at 0.608, and specific gravity at 2.61.

The samples including two different mixtures of mortars which were OPC as the control and the OPCCS (OPC Cockle Shell) were prepared, produced, and tested at the Construction Materials Laboratory. The composition of the specimens includes cement:sand:water at 1:2.75:0.55 and this means each with 50x50x50mm size contained 83.34g of cement, 229.1g of sand, and 45.84g of water. A trial specimen was also prepared to determine the optimum amount of ground cockle shell needed at 0, 2, 4, 6, and 8% by cement weight and the most appropriate value was found to be 4%.

The samples were produced by mixing sand, OPC, water, and ground cockle shells according to the SNI 6882:2014 (SNI 6882:2014, 2014) and cast in a 50x50x50mm mortar mould. They were demoulded the next day and cured in a water pond at Structural Engineering Laboratory, Faculty of Engineering for 28 days after which the OPC and OPCCS samples were immersed in water up to 120 days as the control mixes and also in fresh peat water for the same period.

Peat water was obtained from the peat canal in Rimbo Panjang, Kampar Regency, Riau Province and the physical and chemical composition recorded are presented in Table 2. It was observed to have a brownish-red colour like rust, high organic content estimated at 328 mg/l, pH at 3.85 which was considerably lower than the value required for quality drinking water, and very low alkalinity which exposes the mortar to the organic acid attack in the peat environment.

Figure 1. Seashells were cleaned, crushed and ground into fine particles.

Table 1. Chemical composition of Ordinary Portland Cement (OPC) and Cockle Shell (CS)

Oxides (%)	Type of material	
	Ordinary Portland Cement (OPC)*	Cockle Shell (CS)
CaO	65.21	61.09
SiO_2	20.92	0.29
Al_2O_3	5.49	1.72
Fe_2O_3	3.78	0.08

*Salain (2009)

Table 2. Physical and chemical composition of peat water

Parameters	Unit	Drinking water qualities	Results
Colour	TCU	15	550
Turbidity	NTU	5	99.7
pH value	-	6.5-8.5	3.85
Organic content	mg/L	10	328
Alkalinity	mg/L	500	53
Iron (Fe)	mg/L	0.3	0.8
Manganese (Mn)	mg/L	0.1	<0.0248
Sulfate (SO_4^{2-})	mg/L	400	34
Chloride (Cl)	mg/L	250	31

The compressive strength of the samples was measured on the 7th, 28th, 91st, and 120th day according to SNI 03-6825-2002 (2002) while the porosity was evaluated on the 28th, 91st, and 120th day in line with ASTM C642-06 (ASTM C642-06, 2006). Moreover, the sorptivity test was determined by adopting a method from Taywood Engineering (Peek et al. 2007) where 50x110mm specimens were prepared, dried at 105^{O}C to achieve constant mass, and supported with steel rods in a tray of water at 1-2 mm depth. The specimens were also dried and weighed at an increment of 5, 10, 30, 60, 120, 180, and 240 minutes while the change of mortar weight in water and peat water were determined at 1, 3, 7, 14, 21, 42, 56, 84, 91, and 120 days based on ASTM C267 method (ASTM C267, 2006). Some specimens were brushed using a soft brush in line with O'Connell et al. (2012) to obtain the original weight of the mortar without deposit and loose aggregates at the surface after being exposed to the water and peat water. The pH inside split mortar specimens was visually examined at the 7th and 120th days using phenolphthalein solution.

3 RESULTS AND DISCUSSION

3.1 Compressive Strength

Figure 2 shows the compressive strength for both the OPC and OPCCS mortar samples immersed in water between 0 to 120 days and a gradual increase was observed in the OPC (OPC-W) and OPCCS mortar (OPCCS-W). However, the OPCCS-W mix was found to have higher values after 28 days than the OPC-W and this indicates a positive effect of adding 4% ground cockle shells to the mortar. A similar result was reported by Seo et al. (2019) through the use of oyster shells powder as an additive in OPC mortar which provided an extra enhancement during the hydration process which involves adding $Ca(OH)_2$ at the initial reaction stage. This study, therefore, showed the inclusion of 4% ground cockle shells certainly improves the strength development of OPCCS-W.

Figure 3 shows the compressive strength of the OPC-P sample increased by 19.64% on the 28th day due to the early hydration process and also decreased quite significantly by 22.87% on the 120th day. This gradual reduction occurred due to the reaction of the acid ion with the hydration product of Portland cement and water or portlandite ($Ca(OH)_2$). This is in agreement with the findings of Zivica (2006) that the chemical bonding between cement paste becomes weak due to the continuous reaction between portlandite and acid. Olivia et al. (2014) also observed a similar result with the immersion of OPC mortar in peat water for 120 days. This means the gradual attack of the calcium in the cement matrix by the acid ion eventually deteriorated the OPC samples even though the mortar matured fully at 28 days.

A different trend was observed for OPCCS mortar in peat water (OPCCS-P) as observed with the considerable increase in the compressive strength by 11.29% from 28 to 120 days after a high early value was recorded. This indicates the provision of additional CaO by ground cockle shell in the mixture, thereby, leading to an increase in early hydration process and hardening rate of OPCCS-P. The extra enhancement provided showed the shells performed a better role as an additive by improving the resistance ability of mortar compared to the cement material it replaced. According to Lerwattanarurk et al. (2012), the use of ground shells in replacing cement involves mixing a less reactive material with Portland cement and this is not usually beneficial for concrete in the aggressive environment at the early period.

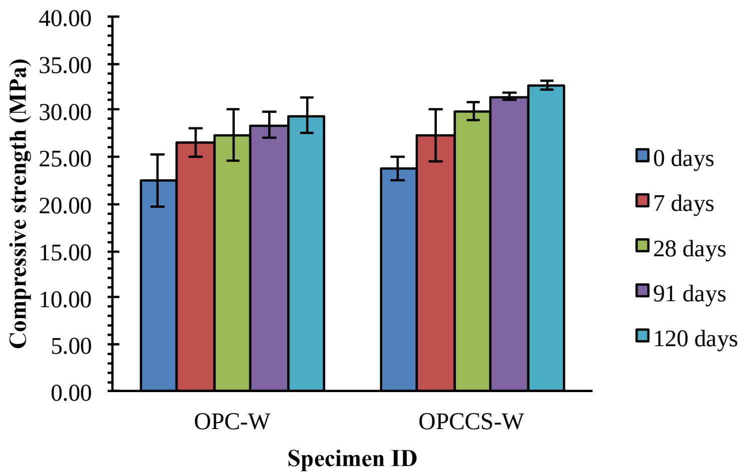

Figure 2. Compressive strength development of OPC and OPCCS mortar in water from 0 to 120 days.

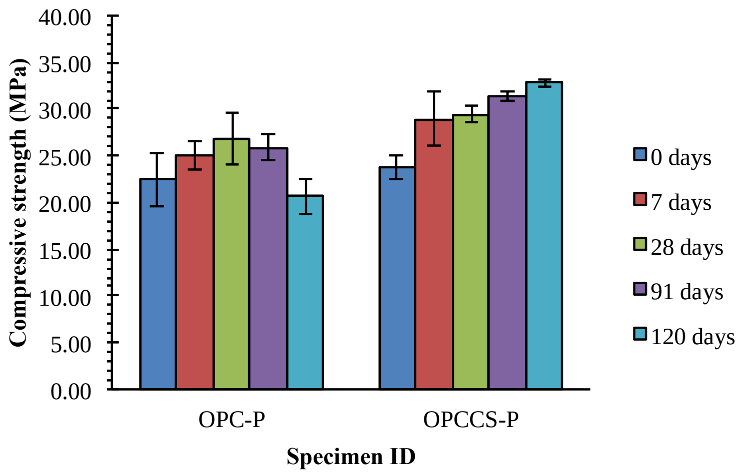

Figure 3. Compressive strength development of OPC and OPCCS mortar in peat water from 0 to 120 days.

3.2 Porosity and Sorptivity

Porosity and sorptivity tests were used as indicators of pores and capillary pores network development in the mortar or concrete. The values obtained for all the samples are presented in Tables 3 and 4 with a gradual porosity reduction in the range of 2.81-5.87% generally observed for both samples from 28 to 120 days. Moreover, the continuous hydration of portlandite or $Ca(OH)_2$ which fills the pore network helps to decrease the pore size and tortuosity of the cement matrix. Meanwhile, the porosity of OPC-P mortar was recorded to have increased quite significantly by 12.14% from 28 to 120 days and this is possibly associated with pores alteration when portlandite was leached due to the acid ion attack. This is in line with the reports of Beddoe & Dorner (2005) that a common phenomenon for cement deterioration due to acid attack is leaching of portlandite, Calcium Silicate Hydrate, and ettringite and this consequently changes the concrete's porosity. Goyal et al. (2009) also found a higher mass loss in Portland cement concrete compared to those with supplementary cementitious materials due to the increment in porosity after the sulfuric acid attack. Moreover, Olivia et al. (2014) confirmed a similar behaviour based on the early deterioration of Portland cement mortar strength in comparison with other mortars which used pozzolanic materials.

OPCCS-P mortar, however, showed a considerable porosity reduction of 6.13% at 120 days and this improvement is possibly due to the ability of ground cockle shells to provide an extra filling to the pores even though the samples were immersed in organic acid peat water. This is supported by the results of Sophia & Sakthieswaran (2019) where a significant porosity reduction was recorded due to the addition of fine shell powder which modified the porous network of plaster by filling the inter-crystalline and interlaminate pores. Another research on the ability of seashells brick to neutralize acidity in water tanks confirmed the porosity improvement even though the material has been immersed in an acid environment for some time (Chiou et al. 2014).

Sorptivity or water absorption rate is mostly influenced by the capillary porosity network of the mortar and its values were observed in Table 4 to have decreased after 28 days for all mixtures, except OPC-P mortar. OPC-W showed a very significant sorptivity reduction by 39.10% while OPCCS-W showed a slight decrease by 8.11% at 120 days. This shows the addition of ground cockle shells improved the capillary porosity of the OPCCS-W during the period of being immersed in water. Furthermore, OPC-P mortar sorptivity showed a similar trend with porosity as observed in a significant increase in its value by 29.89% at 120 days. Based on this evaluation, the water absorption rate of the OPC-P mortar was high because the samples have lost their integrity after being exposed to peat water for up to 120 days.

A noticeable reduction in sorptivity by 31.07% was observed in OPCCS-P mix and this indicates the ground cockle shell added improved the capillary porosity network of concrete as observed with the gradual decrease in the value over time. The material acted as a filler which reacted with Portland cement to significantly improve the tortuosity or fluid flow path in the mortar as also observed in Tayeh et al. (2020). Moreover, a study by Martinez-Garcia et al. (2020) confirmed the use of crushed mussel shells as a sand replacement in a mortar was able to reduce the water path tortuosity due to their surface texture and shape. Therefore, the improvement of sorptivity using ground cockle shells has the ability to increase water tightness for mortar during continuous exposure in peat water.

Table 3. The porosity of OPC and OPCCS mortar in water and peat water at 28, 91 and 120 days

Mixtures	Porosity (%)					
	28 days	SD	91 days	SD	120 days	SD
OPC-W	16.37	1.14	16.04	3.93	15.91	4.04
OPCCS-W	15.00	0.46	14.87	0.65	14.12	1.26
OPC-P	17.46	1.68	18.33	0.38	19.58	3.75
OPCCS-P	16.78	0.64	16.36	1.12	15.81	1.65

Table 4. Sorptivity of OPC and OPCCS mortar in water and peat water at 28, 91 and 120 days

Mixtures	Sorptivity (%)					
	28 days	SD	91 days	SD	120 days	SD
OPC-W	0.2537	0.9992	0.2370	0.9939	0.1825	0.9993
OPCCS-W	0.1825	0.9993	0.1751	0.9905	0.1688	0.9958
OPC-P	0.2736	0.9939	0.3021	0.9982	0.3554	0.9989
OPCCS-P	0.2636	0.9990	0.2339	0.9935	0.2011	0.9943

3.3 Change in Weight

Figure 4 displays the weight change for both brushed and unbrushed mortar specimens after continuous immersion in water. Brushing was used to represent abrasive behaviour associated with flowing water and loss of materials on the surface of the samples. The difference between these specimens during the immersion time was indicated by the increasing chemical reaction at the specimen's surface and the reduction in the amount of residual weight in brushed than unbrushed specimens. Figure 4 shows the weight of OPC-W and OPCCS-W have gradually increased during immersion in water with a weight difference of 21.33% recorded between the brushed and unbrushed samples of OPC-W while OPCCS-W had 26.30%. This means the OPCCS-W samples lost more weight between 45.52- 47.66% even though the trends remained the same and despite the improvement observed at the porosity level due to the addition of ground cockle shell, the surface deterioration of the samples showed the hydrated product of OPCCSS-W was slightly dissolved in the water.

Figure 5 presents the changes in the weight for both OPC-P and OPCCS-P specimens subjected to the peat water and the values for unbrushed OPC-P and OPCCS-P were considerably higher than for brushed samples. The formation of a soft white substance was reported on the surface of the samples and brushing was observed to have removed the loosely acid attack product. Meanwhile, both brushed OPC-P and OPCCS-P recorded a significant weight loss between 11.44 to 47.42% than the unbrushed samples after they were immersed in peat water for four months and the brushed OPC-P mortar seemed to be more prone to the acidic environment. According to Koenig et al. (2017), materials containing high content of Ca tend to have high porosity in the organic acid environment due to erosion and leaching of hydrated Portland cement phases. However, a significant weight loss after the OPCCS-P samples have been brushed shows there was a considerable corrosion product on their surface and the gradual decrease in the weight is attributable to the increase in high pores interconnectivity which allows organic acid movements from the outer to the inner side of the concrete. Meanwhile, the increase in CaO content due to the addition of 4% cockle shell in the mortar was able to improve the porosity of the unbrushed OPCCS-P concrete and stability of the network. Moreover, Chiou et al. (2014) have previously reported that calcining oyster shells at 850-950^{O}C produced CaO while a more reactive CaO was produced at 1050^{O}C with the ability to improve the bricks alkalinity and this probably counterbalanced the acid attack and reduced the leaching process of the mortar through a slow reaction. The acidity reduction around the brick, however, has the ability to neutralize the acidic rain tank.

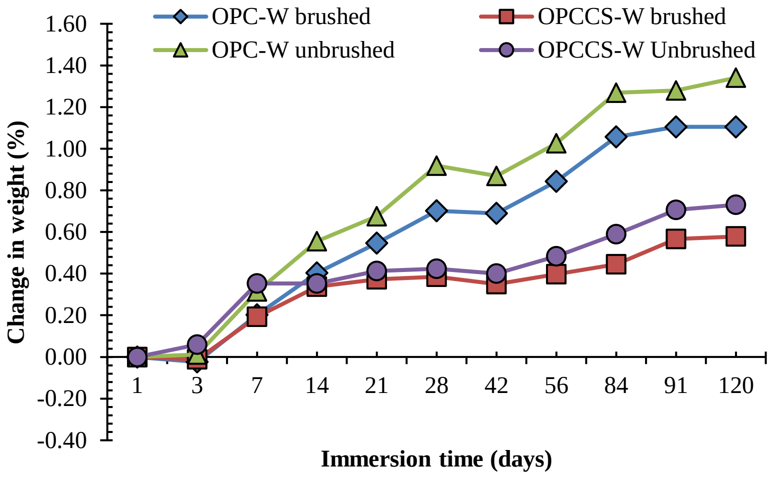

Figure 4. Weight change of brushed and unbrushed OPC and OPCCS mortar in water.

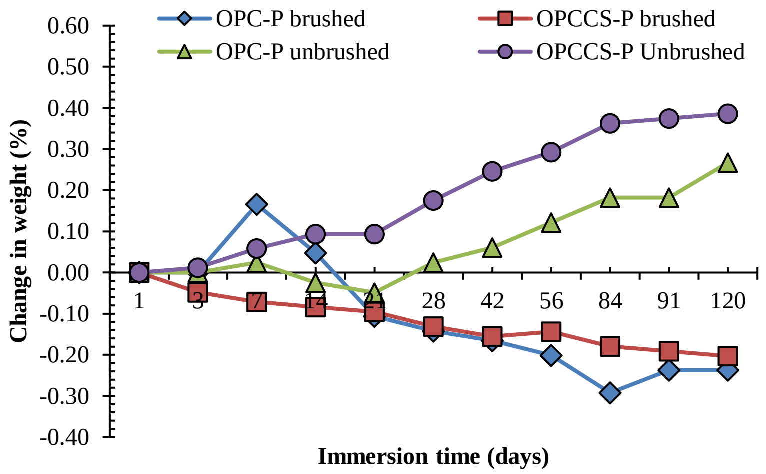

Figure 5. Weight change of brushed and unbrushed OPC and OPCCS mortar in peat water.

3.4 Visual Examination

All the specimens were visually examined at the 7th and 120th days with the pH measured using phenolphthalein solution to determine the mortar colour or alkalinity after immersion in both water and peat water. The OPC mortar specimen sprayed with phenol had solid pink surface colour on the 7th day after immersion in water as shown in Figure 6(a) and a stronger pink colour after 120 days as indicated in Figure 6(b) to show the better alkalinity on both mixtures compared to the 7th day. Meanwhile, there was no significant change in the OPC and OPCCS surfaces when immersed in peat water for seven days as shown in Figure 6(c) since the acid attack was not very strong at the period.

Both specimens did not show intense pink colour in peat water and the tendency of the OPC mortar to be colourless at 120 days as shown in Figure 6(d) due to pH reduction after they were immersed and this indicates alkalinity reduction. In the long term, the specimens experienced a decrease in pH due to the contact with peat water but OPCCS showed a considerable reduction with values lesser than OPC at 120 day and slightly similar to OPCCS immersed in water for 120 days as shown in Figure 6(c). This visual

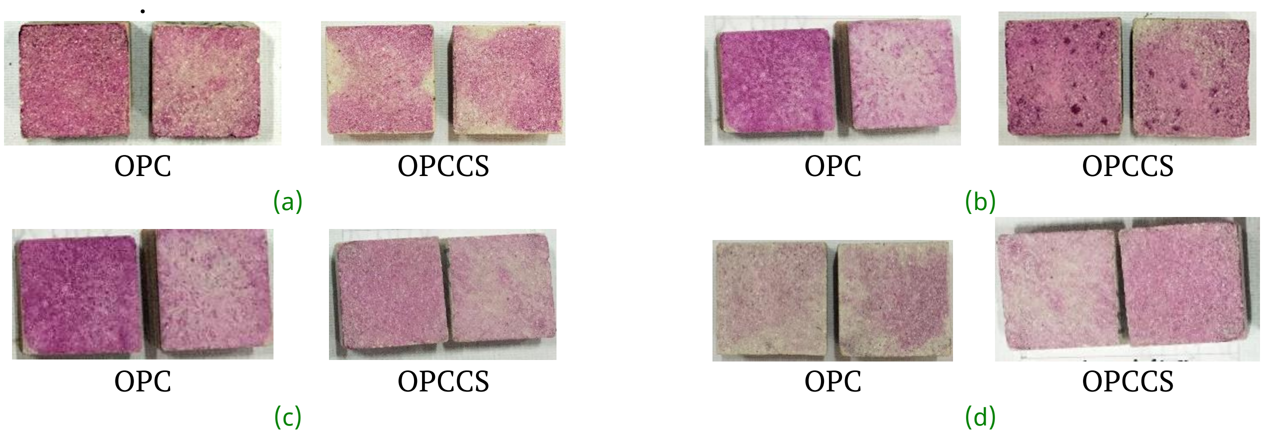

Figure 6. Visual examination of OPC and OPCCS mortar using phenolphthalein solution: (a) in the water at 7 days, (b) in the water at 120 days, (c) in peat water at 7 days, (d) in peat water at 120 days.

examination shows the alkalinity reduction of the OPCCS mortar was lesser than the control mortar (OPC) after prolonged immersion in the peat water.

The OPCCS specimens were observed to have a better resistance in peat water and this is associated with the stability of the CaO in the additive material compared to the rich-Ca cement only and its ability to create and neutralize weak acidic environments like peat. The addition of ground cockle shells also reduces the leaching rate of Ca from mortar, protect its network by increasing the pH of the specimens, and slows down the deterioration of the mortar. Therefore, this study recommends the use of OPCCS mortar due to its high potential as a lining material in the peat environment.

4 CONCLUSIONS

Cockle shells were burnt, grounded, and applied as an additive in mortars in this study as OPCCS and the results showed the compressive strength of OPCCS in peat water increased by 11.29%, porosity decreased by 5.78% and sorptivity reduced significantly by 31.07% after 28 days. Moreover, the weight change of brushed OPCCS in peat water was lesser compared to brushed OPC mortar partly due to the less disintegration of the particles in the OPCCS mortar surface after the specimens were brushed. The visual examination showed the addition of ground cockle shells increased the resistance of mortar in peat water due to the improvement in the pH and this is considered beneficial for the application of the mortar in peat environments. Furthermore, a faster strength gain at the early ages, high filling capability of pores, ability to neutralize acidity, and better long-term resistance to organic acid in peat water were also recorded with the use of the material as an additive than the use of Ordinary Portland Cement in mortar. This research, therefore, demonstrated the use of a ground cockle shell in mortar has the ability to efficiently improve the porosity, sorptivity, and strength of mortar and this means it is suitable as a drainage lining in peat environments.

DISCLAIMER

The authors declare no conflict of interest.

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