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COMPRESSIVE STRENGTH OF MORTARS PRODUCED BY PARTIAL REPLACEMENT OF CEMENT WITH SEASHELLS AND SUGARCANE BAGASSE ASH

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Abstract: Cement production is regarded as one of the major contributors for global emissions. Mortars are cement composites used widely as construction. The investigated the suitability of three seashells as replacement of cement in mortars and its possible synergy with sugarcane bagasse ash. Mortars with partial replacement of cement Portland by seashell and sugarcane bagasse ash (SBA) were produced. Three types of Seashells were used, *Polynices mammilla* (PMS), *Ennucula uruguayensis* (EUS) and *Anadara granosa* (AGS). The shells were collected, on the beach of Costa do Sol in Maputo City, washed, dried and calcined at 800 ºC to 1100 ºC for 2 to 3 hours. Sugarcane bagasse was collected on a sugar mill company, namely Açucareira de Xinavane, and was calcined at 500 ºC to 900 ºC for 2 to 3 hours to produce SBA. After grinding, shells flour and SBA were used to replace 32.5N and 42.5N cement at 5, 10 and 15% by weight, either separately or in combination. Using 42.5N cement results indicate a replacement limit at of 5% with seashell flours, while with 32.5N cement the replacement limit was at 5%. Mortars of 42.5N cement with 5% of *P*MS showed a higher compressive strength (45.85 MPa) than the control mix (43.85MPa). For 32.5N cement the highest compressive strength was achieved with 5% replacements using *AGS* and *P*MS. Mortars of 42.5N cement with 5% seashell+3% SBA presented the highest compressive strength compared to other replacements, after 28 days curing, although lower than the control mortar. The association of seashells powders and sugarcane ash can be used to replace cement up to 8%.

Keywords: seashells, sugarcane bagasse ash, cement, mortars

INTRODUCTION

Mortar is a workable paste used in construction material in the world. According ASTM C270-10, (2010) mortars are classified into four types namely M (2500 MPa), S (1800 MPa), N (750 MPa), and O (350 MPa) based on different field of application. To produce a high strength, superior durability and hard cement mortar normally the mortars are mixed with fine aggregate in proportion by volume of 1:3 (NP EN 196-1, 2017). Cement production involves the emission of huge amount of greenhouse gases such as -CO_2 into the atmosphere (Damtoft et al., 2008; Lippiatt et al., 2020; Mohamad et al., 2022). During the production of each ton of cement about 325 kg CO_2 are produced only due to the burning of fuel in the furnace, around 525 kg of CO_2 are emitted per ton of clinker during the decarbonation of limestone, and 50 kg produced due to the consumption of electricity. CO_2 is the main contributor to global warming (Özbay et al., 2016;TERI, 2017). In recent years, researchers are looking into new methods to increase the strength of cement masses, by substitution of cement and aggregates with alternative materials, normally waste materials like ashes from rice husk, sugarcane bagasse, bamboo fibre, wood and corn cob or seashell powder and ferronickel slags (Athira et al., 2020; Bamigboye et al., 2021; Bouasria et al., 2021; Câmara et al., 2016; Marizane et al., 2020; Panda et al., 2020; Rollakanti et al., 2020). These waste products can enhance both the durability and the strength properties of the cement masses when they are used as pozzolanic materials in high performance concrete and mortars (Khalil et al., 2020; Liu et al., 2022). Mozambique currently has four (4) sugarcane industrial units namely; Açucareira de Maragra, Açucareira de Xinavane, Açucareira de Moçambique e Companhia de Sena, which accounts for a total plantation area of more than 40 hectares (Atanassov et al., 2017; Sutton, 2014). These companies produce between 21,478 to 49,848 tonnes of sugarcane bagasse ever three months (Ferrão, 2019). Traditionally, the bagasse (waste material resulting from the sugarcane mill process) is used to produce energy, providing steam for the manufacturing process and for electrical power generation (Gesto-Energia, 2014). However, the bagasse ash from the combustion process is simply discarded. Sugarcane bagasse ash (SBA) is basically composed by amorphous alumina, amorphous silica or partially crystalline (vital ingredients of the pozzolanic reaction with calcium hydroxide). SBA can be utilized as a pozzolanic material. Studies demonstrated that it presents potential to be used as mineral addition for partial replacement of cement in concrete and mortars. Different researchers showed that concrete and mortars with substitution of SBA up to 10% has a compressive strength greater than the control mix (Berenguer et al., 2016, 2018; Khalil et al., 2020; Murugesan et al, 2020; Salimo et al., 2021).

Seashells wastes are by-product of the aquaculture industry. A variety of shellfishes are consumed as food and the shells are discarded because these have little or no commercial value. An alternative solution for discarded seashell wastes is to use as partial replacement of cement in mortar and concrete, contributing therefore to the protection of the environment and preservation of natural resources. When materials are re-used, costs are also saved and which range from not having to landfill or dispose waste materials and not having to source new materials (Eziefula et al., 2018; Liu et al., 2022; Mo et al., 2018). Seashells are very easily found throughout the sea shore or coastal areas (Rollakanti et al., 2020). The 16 million tonnes of molluscs worldwide production represents about 22% of the total global aquaculture production. Shells of bivalves and gastropods of mollusc shellfishes are the main seashells used as aggregate in cement masses. Bivalve molluscs are very common among marine shellfish species, around 87% of molluscan aquacultures are bivalve molluscs; 33.0% clams (including ark shells and cockles), 31.3% oysters, 12.1% mussels and 10.9% pectens and scallops (Eziefula et al., 2018). Mollusc is an important food source for local populations in Mozambique, mainly families living along the coastal areas. According to the Minister of Fishing of Mozambique from 2009 to 2016 Mozambique exported over 100 tonnes of dried shells. However, in Mozambique the invertebrate fauna is still little known and most of the researches of shells were accomplished in Cabo Delgado, Nampula and Maputo Province (Collin et al., 2008; Mualeque, 2014; Nassongole et al., 2019). The chemical composition of shells is >90% calcium carbonate ($CaCO_3$) by weight and this composition is similar to limestone powder or dust-like stone powder from grinding limestone to produce Portland cement. The crystal structures of green seashells are largely composed of aragonite and calcite, which have higher strengths and densities than limestone powder (Bamigboye et al., 2020; Janković et al., 2020; Lertwattanaruk et al., 2012; Olivia et al., 2017; Soltanzadeh et al., 2018; Tayeh et al., 2020; Wan Mohammad et al., 2017; Wang et al., 2019).

Rollakanti et al. (2020)obtained beneficial effect after replacing up to 15% cement with a mixture of seashell and wood ash (1:1 ratio). Olivia et al. (2015) replaced 2, 4, 6 and 8% by weight of cement with ground cockle seashell, the optimum compressive strength was achieved with 4% replacement, although smaller than the control cement masse (37 MPa). According to Lertwattanaruk et al. (2012) the observed lower compressive strength can be attributed to the lower CaO content of the shells, compared to cement, which cause a reduction on hydration reaction. Seo et al. (2019) calcined oyster shell powder as an expansive additive in cement mortar. In this research oyster was calcined at 1000 ^{o}C for 3 hours. The compressive strength of 0%, 3%, 6%, 9% and 12% at 56 days were 53.7 MPa, 57.5 MPa, 52.3 MPa, 51.0 MPa and 42.0 MPa, respectively. These results suggest that the amount of incorporated calcined shell powder should be limited to < 6% considering the workability and the mechanical properties of cement mortar. Wang & Liu (2020) replaced cement by seashell powder of 0%, 5%, 10%, 20%, and 40%. The results showed that the compressive strength of 5 % addition was even comparable to the control mortar. Tayeh et al. (2020) also showed that the compressive strength of the 5% replacement is slightly higher than the control cement at 28 and 90 days of age.

The main purpose of the present research is to investigate the suitability of 3 types of seashells collected in Maputo City on Costa do Sol beach and sugarcane bagasse ash from Xinavane sugar factory, as replacement material in the production of mortar suitable to reduce cement production and to enhance the strength properties of the cement masses. *Anadara Granosa*, *Ennucula Uruguayensis* and *Polynices Mammilla* are the most popular shellfish consumed in Mozambique.

MATERIALS AND METHODS

MATERIALS

The cement used in this study is a CEM II/ BL 42.5N and CEM II/ BL 32.5N made by Cimentos Dugongo (Mozambique). In this research we used sand recommended by NP EN 196-1, 2017 standards, collected at the Corumana dam, located in the District of Moamba – Maputo Province. The sand was washed manually, dried and then grinded to produce six classes. The three types of seashells (*Anadara Granosa Shell* - AGS, *Ennucula Uruguayensis Shell* – EUS and *Polynices Mammilla Shell* - PMS) were collected in Maputo City, Costa do Sol beach. The seashells were cleaned (Fig. 1), dried in electric furnace for 24 hours at temperature of 100 ± 5 ^{o}C, and calcined an electric furnace for 3 h at 1100 ^{o}C and finally crushed into a fine powder (<0.08 mm) (Fig. 2).

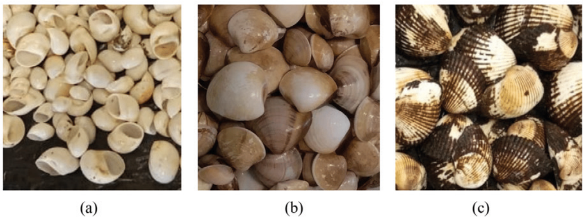

Figure 1: Seashells used after washing. (a) Polinyces mammilla shell; (b) Ennucula uruguayensis shell, and (c) Anadara granosa shell

Figure 2: Flour produced after burning seashell at 1100 ^{o}C and milling.

The sugarcane bagasse ash (SBA) used was collected in Xinavane sugar factory. First the ashes were dried at 25 ^{o}C for one week and in electric furnace for 12 hours at 100 ± 5 ^{o}C and grinding until <0.08 mm, finally SBA were calcined at 900 ^{o}C for 2h in muffle at the rate of 10 °C/min.

The termogravimetric analyses –TGA of seashell powders were conducted in SDT-Q-600, model TGA/DSC Instrument. Samples between 23 mg to 83 mg were placed aluminium tray by heating rate of 10 ^{o}C /min in nitrogen atmosphere up to 1100 ^{o}C.

And the chemical composition of cements, seashells and sugarcane bagasse ash (SBA) was obtained using X-ray fluorescence, Instrument EDX-700, atmosphere of air, collimator: 10 mm.

COMPOSITION OF STUDIED MORTARS

Mortars were prepared by mixing water, cement and sand on fixed ratio of 1:2:3 by weight, according to NP EN 196-1, (2017) standards. The control mix consisted of cement (450 g), water (225 ml) and fine aggregates (1350 g). In this investigation the replacement was categorized in three groups (Table 1): (a) in the first group, the Portland cement 42.5N was replaced by seashell powders with 0%, 5%, 10% and 15%; (b) in the second, the cement 42.5N was partially replaced by association of seashell powders (SP) and SBA with 5%SP+3%SBA, 5%SP+5%SBA, 5%SP+10%SBA, 10%SP+3%SBA and 10%SP+5%SBA; (c) in the third the Portland cement 32.5N was replaced by seashells powders with 0%, 5% and 10%. The mortars were moulded in 40 mm x 40 mm x 160 mm specimens (Fig. 3). The specimens were stored in water under 20 ± 1,0 °C for 7 and 28 days for curing. After the curing the specimens were assessed for the compressive strength in accordance with NP EN 196-1 (2017).

Figure 3: Specimens preparation of 40 x 40 x 160 mm³

RESULTS AND DISCUSSION

MATERIAL CHARACTERIZATION

Termogravimetric analyses –TGA

The TGA (Fig. 4) showed two stages: (i) the first weight loss was observed between 200 °C and 680 °C and occurred 1.87%, 1.29% and 1.4% weight loss for *anadara granosa shell* (AGS), *polynices mammilla* shell (PMS) and *ennucula uruguayensis Shell* (EUS), respectively, due to evaporation of water, and oxidation and removal of organic material from the seashell (Bamigboye et al., 2020; Laskar et al., 2018; Peceño et al., 2021; Suarez-Riera et al., 2021). (ii) The second weight loss is between 640 to 840 °C (43.34%) for AGS, 680 to 835.19 °C (43.06%) for PMS and 43.93% between 640 and 782.5 °C for EUS. The second mass loss is due to the decomposition of carbonates ($CaCO_3$ and $MgCO_3$) to CaO or MgO to CaO with the release of CO_2 (Mo et al., 2018; Peceño et al., 2021; Safi et al., 2015; Wang et al., 2019)in particular seashells in a self-compacting mortars (SCMs.

X-ray fluorescence

Table 2 show the chemical composition of cements, seashells and SBA. SBA was found to have 79.84% of SiO_2 + Al_2O_3 + Fe_2O_3 and 51.78% for SiO_2. The total amount of SiO_2 + Al_2O_3 + Fe_2O_3 is more than 70%, then the ashes can be considered as a pozzolanic material and could contribute to the strength development process in mortars (Akbar et al., 2021; Quedou et al., 2021).

The chemical composition of seashells was mainly CaO and ranged from 93.83% to 99.17%, in agreement other results presented in literature (Hart, 2020; Mo et al., 2018; Seo et al., 2019; Tayeh et al., 2020)eggshells, snail shells, etc.. This suggest that seashells powder can be used as a cement replacement in construction.

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Group	Mortar mix proportions	Mix Identity
References	Cement 42.5N (100%) Cement 32.5N (100%)	CM CM1
Group 1 (mass substitution of cement 42.5N by seashells)	Cement (95%) + PMS (5%) Cement (95%) + EUS (5%) Cement (95%) + AGS (5%) Cement (95%) + SBA (5%) Cement (90%) + PMS (10%) Cement (90%) + EUS (10%) Cement (90%) + AGS (10%) Cement (90%) + SBA (10%) Cement (85%) + PMS (15%) Cement (85%) + EUS (15%) Cement (85%) + AGS (15%) Cement (85%) + SBA (15%)	PMS (5%) EUS (5%) AGS (5%) SBA (5%) PMS (10%) EUS (10%) AGS (10%) SBA (10%) PMS (15%) EUS (15%) AGS (15%) SBA (15%)
Group 2 (mass substitution of cement 42.5N by seashells and sugarcane bagasse ash)	Cement (92%) + PMS (5%) + SBA (3%) Cement (92%) + EUS (5%) + SBA (3%) Cement (92%) + AGS (5%) + SBA (3%) Cement (90%) + PMS (5%) + SBA (5%) Cement (90%) + EUS (5%) + SBA (5%) Cement (90%) + AGS (5%) + SBA (5%) Cement (85%) + PMS (5%) + SBA (10%) Cement (85%) + EUS (5%) + SBA (10%) Cement (85%) + AGS (5%) + SBA (10%) Cement (87%) + PMS (10%) + SBA (3%) Cement (87%) + EUS (10%) + SBA (3%) Cement (87%) + AGS (10%) + SBA (3%) Cement (85%) + PMS (10%) + SBA (5%) Cement (85%) + EUS (10%) +SBA (5%) Cement (85%) + AGS (10%) + SBA (5%)	PM5C3 EU5C3 AG5C3 PM5C5 EU5C5 AG5C5 PM5C10 EU5C10 AG5C10 PMS10C3 EUS10C3 AGS10C3 PMS10C5 EUS10C5 AGS10C5
Group 3 (mass substitution of cement 32.5N by seashells)	Cement (95%) + PMS (5%) Cement (95%) + EUS (5%) Cement (95%) + AGS (5%) Cement (90%) + PMS (10%) Cement (90%) + EUS (10%) Cement (90%) + AGS (10%)	PMS5 EUS5 AGS5 PMS10 EUS10 AGS10

Table 1: Mix proportions of mortar and identity

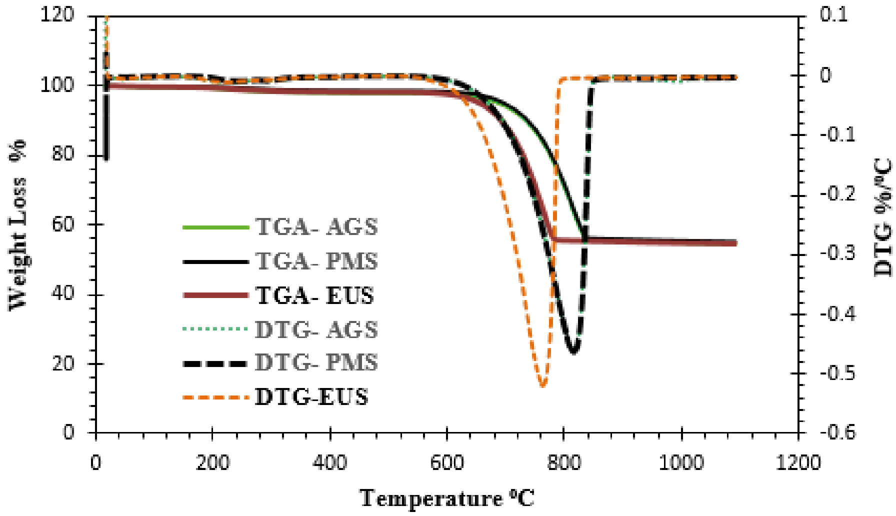

Figure 4: TG/DTG and first derivative of seashells

Oxide	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3	K_2O	TiO_2	SrO
Cement 42.5N	60.81	22.19	7.81	4.84	2.54	1.07	0.49	-
Cement 32.5N	67.37	22.19	0.67	4.71	3.21	1.11	0.48	-
SBA	3.95	51.78	21.34	6.72	0.01	9.87	0.40	-
PMS	98.92	-	-	0.04	-	0.57	-	0.32
EUS	99.23	-	-	0.01	-	-	-	0.27
AGS	98.99	-	-	0.11	-	0.64	-	0.24

Table 2: Chemical composition of cements, seashells and sugarcane bagasse ash (SBA)

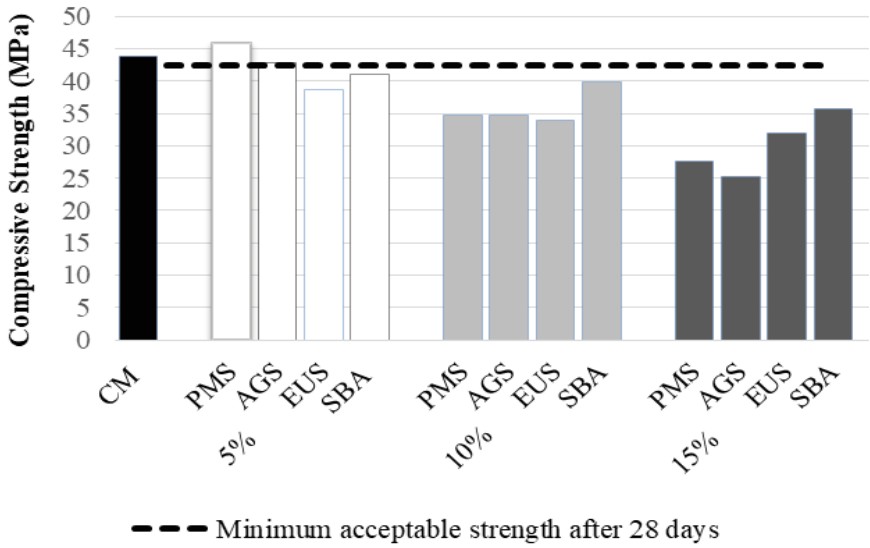

Figure 5: Compressive strength at 28 days of the mixtures for group 1

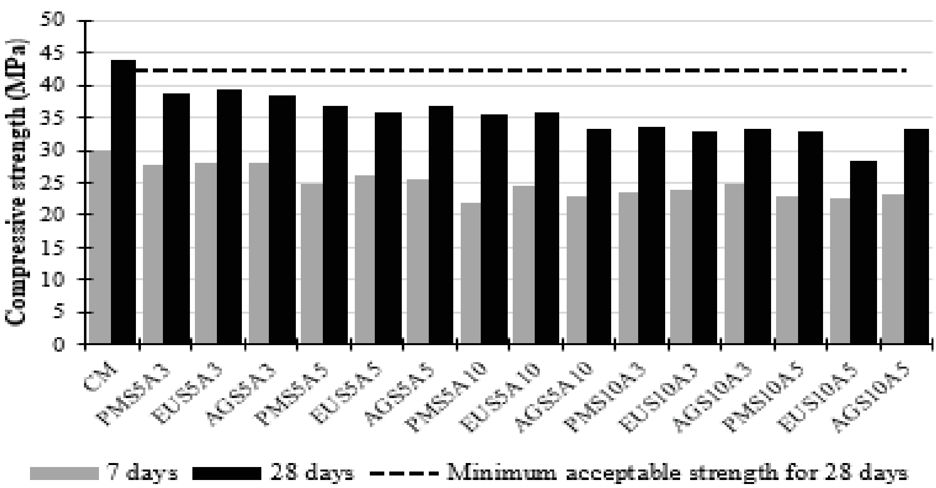

Figure 6: Compressive strength at 7 and 28 days of the mixtures for group 2

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COMPRESSIVE STRENGTH OF MORTARS

The compressive strength of mortars of group 1 at 28 days is given in Figure 5. The obtained results illustrate that the 5% replacement of cement 42.5N by seashells resulted in higher compressive strength than the other replacement (10 and 15%). The mortars with 5% of PMS showed slightly higher than 43.85 MPa for the control mix (CM). The compressive strength was 45.85 MPa for mix with 5% PMS and 42.82 MPa for 5% AGS. The 5% mixture with AGS resulted in little lower compressive strength compared to the CM, but is within recommended range by the Mozambican standard for cements 42.5 MPa. Mortars with 10% of PMS, AGS and EUS showed compressive strength in the order of 34.63, 34.62 and 33.95 MPa, respectively. In the substitutions of 15%, EUS presented the highest value (31.93 MPa) compared to 15% PMS (27.64 MPa) and 15% AGS (25.16 MPa). The compressive strength of mortars produced by SBA 5%, 10% and 15% are 41.03 MPa, 39.82 MPa and 35.61 MPa, respectively.

Figure 6 show that the compressive strength at 7 and 28 days of the mixes for group 2. The compressive strengths of the mortars produced by association of seashells powders and SBA were a little lower than that of CM and of 42.5 MPa for a minimum acceptable strength for cement 42.5N according to NP EN 197 - 1, (2001) standards. The compressive strength of mortars with association of 5% seashells flour and 3% SBA was slightly higher than of the other substitutions at 7 and 28 days of curing age. From the Figure 6, the compressive strength at 28 days are 38.44, 38.85 and 39.46 MPa for AGS5A3, PMS5A3 and EUS5A3, respectively. After 28 days the substitutions of 10% (5% seashell + 5% SBA) showed 36.96, 36.91 and 35.9 MPa for AGS5A5, PMS5A5 and EUS5A5. Mortars produced by AGS10A3, PMS10A3 and EUS10A3 provided 33.34, 33.54 and 32.79 MPa for 13% cement replacement (10% seashell + 3% SBA). The compressive strength of 15% cement partial replacement (10% seashell + 5% SBA) was 33.34 MPa for AGS5A10, 32.92 MPa for PMS5A10 and 28.32 MPa for EUS5A10, respectively.

The cement 32.5N was replaced by 5 and 10% of seashells powders, and Figure. 7 show the compressive strength at 7 and 28 days. The best compressive strength is observed for 5% PMS, after 7 and 28 days were found 26.35 MPa and 34.55 MPa, respectively. At 7 days all the mixes show higher compressive strength than 16 MPa for CM. The compressive strengths of mortars increase with the age of all the mixes. And according to Figure. 7, all seashells can replace cement 32.5 N up to 5%.

The reduction in compressive strength found in this research is in line with the results of other studies where cement was replaced by other types of seashells (Baran et al., 2020; Lertwattanaruk et al., 2012; Nguyen et al., 2013; Rollakanti et al., 2020; Suarez-Riera et al., 2021; Wang & Liu, 2020)waste seashells from seafood and aquaculture industry were recycled into powder form and reused in cementitious construction materials. This work aimed to use this biologically renewable waste material as a possible alternative to the nonrenewable limestone mineral used conventionally with Portland cement. The study explored roles of seashell powder as both a partial replacement and an additional additive to cement through evaluating the rheological and physico-mechanical properties of early-age cement paste. The cyclic rheological measurement including model analysis (Bingham, Casson, and Herschel-Bulkley models. In general, it was observed here that increasing the percentage of replacement up to 10% by seashells and by association of seashell and SBA resulted in the reduction of the compressive strength of the mortars. The cause of reduction in compressive strength

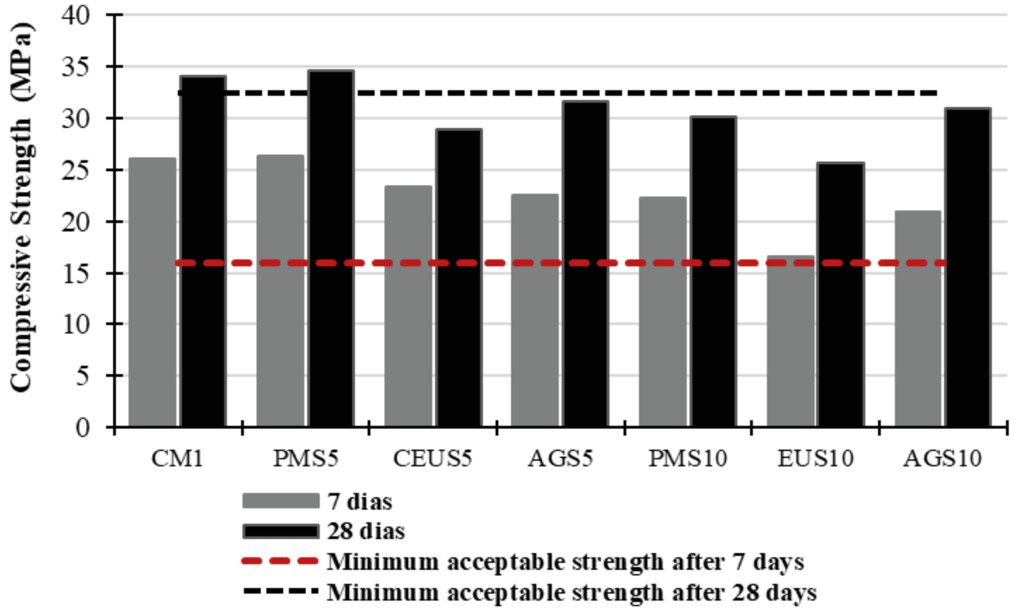

Figure 7: Compressive strength at 7 and 28 days of the mixtures for group 3

with higher percentages of replacement is probably due to the presence of CaO in seashell powder which could react with Al_2O_3 and gypsum, and this reaction reduces the early hydration of alite (Lertwattanaruk et al., 2012; Tayeh et al., 2020)10, 15 and 20% by weight. These mixes are tested and compared with a SC0 that has 0% seashell ash powder (SC0. According to Stel'makh et al. (2022) it has been found that the optimal replacement by the shell powder is in the amount of 6%. The compressive strength of the mortar replaced of 5% cement 32.5N and 42.5N by seashells shows the satisfactory compressive strength class (>32.5 MPa and >42.5 MPa for 28 days) recommended by NP EN 197-1, 2012 and NP EN 197 - 1, 2001 standards (Baran et al., 2020; Lertwattanaruk et al., 2012; Olivia et al., 2015; Seo et al., 2019; Tayeh et al., 2020; Wan Mohammad et al., 2017; Wang & Liu, 2020) the shell is a regionally important waste. The biomass is consumed as a fuel in some homes, workplaces, and public buildings with the help of specific caldrons. In the study, the use of ash was researched in the production of blended cement. For this purpose, blended cement mixtures containing hazelnut shell ash at the ratios of 0, 5, 10, 15, 20, 25 and 30% by weight of CEM I 42.5R ordinary Portland cement were prepared. The binary cementitious systems were assessed in terms of chemical, physical and mechanical properties. Hazelnut shell ash introduction increased the water demand up to 59% for standard consistency and reduced setting time up to 96% according to the ordinary Portland cement. Compressive strength results were remarkably reduced by an increase in the amount of ash. With over 5% of hazelnut shell ash introduction, the 28-d compressive strength value was found unsatisfactory (<42.5 MPa. The reduction of compressive strength in mortar is largely attributed to the higher water absorption of seashell in cement mass, and consequently a non-optimal hydration of the cement and this causes a heterogeneous matrix (Nguyen et al., 2013; Suarez-Riera et al., 2021). The compressive strength of mortar can potentially be improved if there would be a synergic and coordinated adjustment on the water content (w/b ratio) and the dosage of seashell powder when is used to replace cement (Wang & Liu, 2020).

CONCLUSIONS

This research shows that: (a) the main chemical composition of seashells powders was calcium carbonate, in the range of 98.92% (PMS), 99.23% (EUS) and 98.99% (AGS); (b) the termogravimetric analysis (TGA) of seashell flour show two stages of weight loss, and the total weight loss was 42.21% (AGS), 44.35% (PMS) and 45.33% (EUS) between 200 ºC and 840 ºC; (c) the three types of seashells showed that the compressive strength of the mortar produced by 5% of AGS, PMS and EUS were adequate to replace cement 42.5 N in mortars, and all seashells can replace cement 32.5N up to 5%; (d) the compressive strength of the mortars replaced by EUS decreased compared with the control mortar; (e) the replacement of cement 42.2N by association of seashells and SBA led to lower compressive strengths in all mixtures after 28 days; (f) the compressive strength of mortar led to decrease when the percentage of flour seashells was increased from 8% to 15%; (g) Also the sugarcane bagasse ashes can replace cement up to 5%; (h) Although the substitutions of cement by association of seashells powders and sugarcane bagasse ash presented a little lower compressive strength than of the control mix, but the association of seashells powders and sugarcane ash can be used to replace cement up to 8%, the values of compressive strength found are acceptable.

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