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Development of ternary blend cement-free binder material for construction

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ABSTRACT

This study aims to develop new ternary blend (TB) cement-free binder materials as an alternative to conventional cement by using Paper Sludge Ash (PSA) waste as the base material. During this research, the combined application of mechanical activation (grinding) and chemical activation by blending with pozzolanic: (silica fume; SF) and (rice husk ash; RHA), and high al-kalinity: (poultry litter fly ash; PLFA) and (cement kiln dust; CKD) materials were investigated. The research plan included four stages in which PSA was activated and replaced with the above-mentioned materials until reaching a ternary blend binder with the best performance (depending on the mortar compressive strength). Thereafter, the performance of this ternary blend binder was compared with the conventional cement by conducting compressive strength (at the ages of 3, 7 and 28 days) and Scanning electron microscopy (SEM) tests. The findings indicated that a cement-free binder material was developed by using a combination of 60% PSA that was blended with 20% RHA and 20% PLFA. The new binder has shown higher compressive strength than the conventional cement by about 12% after 28 days of curing.

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Cement-free binder; construction; ternary blend; paper sludge ash; rice husk ash

1. Introduction

For the construction sector, cement is considered as one of the crucial ingredients that is utilized for different applications for hundreds of years such as concrete production, dams, pavements and buildings. Cement has the advantages of set and hardened quickly that make it suitable to be used in construction projects that require fast pace. Cement can be used along with other construction materials such as steel or/and timber to form composite materials that would provide desirable properties that led to the construction of complex structures worldwide. However, cement as the main binder used in concrete manufacture has several negative environmental impacts associated with its manufacturing. In addition to that, the cost of its production is continuously increasing due to the rise in the cost of production of energy. Carbon dioxide (CO_2) is one of the main greenhouse gases produced during the production of cement along with other toxic gases such as sulphur dioxides (SO_2) (Singh). The existence of toxic gases in

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cement can also be poisonous in the great amount. Nearly 1000 kg of CO_2 is emissioned into the atmosphere with each one tonne of cement produced, that is approximately 7% of the total CO_2 emissions in the atmosphere (Aprianti, 2017; Hamad et al., 2021).

Currently the world is facing other issues due to the accumulation of by-product and waste materials in huge quantities (Nguyen et al., 2021). Some of these issues are the potential contamination of groundwater and virgin lands (H. M. Jafer et al., 2018). The disposal of such materials is considered a costly process. Extensive research has been conducted all around the world to use such materials to completely or partially replacing the cement in different applications. Alternative materials were applied in the construction industry, involving pulverized fuel ash (PFA), ground granulated blast furnace slag (GGBS), palm oil fly ash (POFA), silica fume (SF), and, rice husk ash (RHA), and other admixtures (Aprianti et al., 2015; Hawileh et al., 2017; Hossain & Mol, 2011; Mehta & Ashish, 2019; Nayel et al., 2018; Sasui et al., 2020). Some scholars used these alternatives to minimize the use of cement in different applications, while others utilise them to improve the properties and characteristics of concrete.

Paper sludge ash (PSA) waste has chemical compositions that is similar to that of the cement (Sadique et al., 2019; Shubbar et al., 2020). Therefore, it could be used by itself to fully replace the cement and produce cement-free binder. Different activation techniques are usually required to enhance the performance of such materials (Wang et al., 2007). These techniques could be mechanical activation by the mean of grinding and/or chemical activation by blending with other powder materials or chemical solutions (H. M. Jafer et al., 2018; Wang et al., 2007). The first chemical activation method could be by blending the PSA with pozzolanic materials that has high silica content such as SF and RHA to produce cementitious products that mimic the hydration process of cement (H. Jafer et al., 2018). The other activation method is by blending the PSA with high alkalinity materials involving Poultry fly ash (PLFA) and cement kiln dust (CKD) to enhance the reaction as higher cementitious products could be produced in high alkalinity environment (H. Jafer et al., 2018).

Recently various studies were conducted to produce ternary blends as an alternative to Ordinary Portland cement (OPC) or binary blended cementitious materials (OPC-GGBFS and/or OPC-PFA cements) (Chindaprasirt & Rukzon, 2008; Khalil & Anwar, 2015; Shubbar et al., 2018; Younes et al., 2018). The main idea behind developing ternary blends is that during the production of such blends there will be an increased particle packing, thus nucleation could happen during such blends in a homogeneous way that would lead to the production of hydration products with a very strong microstructure (Khalil & Anwar, 2015; Sadique et al., 2012; Shubbar et al., 2020). This in turns would lead to the production of binders with higher strength and enhanced durability (Khalil & Anwar, 2015; Sadique et al., 2012; Shubbar et al., 2020).

According to the literature, a very limited number of studies have dealt with the combined use of (PAS + RHA + PLFA) in the production of an environmentally friendly, completely cement-free binder. Therefore, this research aims to develop a ternary blend (TB) cement-free binder materials by using PSA waste as the base material and through the combined application of mechanical activation (grinding) and chemical activation by blending with pozzolanic and high alkalinity materials.

2. Materials and methodology

2.1. Materials

2.1.1. Binder materials

During this research, five materials were utilised to completely substitute the cement in various percentages. These materials were Silica fume (SF), Cement kiln dust (CKD), Rice Husk Ash (RHA), Poultry fly ash (PLFA) and Paper sludge ash (PSA). In addition to these materials, flue gas desulphurisation (FGD) gypsum was utilized as a crushing aid. The cement utilized in this research was CEM-II/A/L 32.5 R, which conforms to the BS EN 197-1 (BSI, BS EN 197-1: 2011, 2011). Both the control cement and the CKD were obtained from CEMEX Quality Department, Warwickshire, UK. The PSA and PLFA were supplied by Aylesford Newsprint Ltd, Kent, UK and Fibrowatt Thetford power station, UK, respectively, while the SF was obtained from Elkem Materials Company, Zuid-Holland, Netherlands. The RHA has been supplied from NK Enterprises Company, Jhar-suguda, India, the colour of RHA was black, and its carbon content was 2.11% (measured by the loss on ignition test). The chemical compositions of all the used materials are presented in Table 1. Figure 1 presents the diffraction patterns of the raw materials using the X-ray diffraction (XRD) analysis. The major crystal peaks for the cement were calcite ($CaCO_3$), alite (Ca_3O_5Si) and

Table 1. Chemical compositions of the binder materials.

	Cement	PSA	PLFA	CKD	RHA	SF	FGD
Na_2O %	1.534		–	0.23		0.8	
CaO %	62.389	65.03	9.91	57.23	0.49	–	35.90
SiO_2 %	26.638	24.58	14.38	16.52	90.20	96.89	14.3
Al_2O_3 %	2.436	2.15	–	4.2	4.0	0.5	–
MgO %	1.573	2.60	0.6	0.8	0.61	0.51	0.54
Fe_2O_3 %	1.744	–	0.01	3.8	0.18	0.13	–
SO_3 %	1.887	0.36	–	4.31	–	–	34.64
K_2O %	0.723	0.27	21.33	6.72	1.36	0.63	–

belite (Ca_2O_4Si). The major crystal peaks for the PSA were calcite ($CaCO_3$), lime (CaO), mayenite ($Ca_{12}Al_{14}O_{33}$) and merwinite (Ca_3Mg [SiO_4]). The major crystal peaks of the PLFA in addition to the calcite ($CaCO_3$) and belite (Ca_2O_4Si) there were potassium aluminium phosphate (K_3Al_2 [PO_4]$_3$), perovskite ($CaTiO_3$) and sylvite (KCl). The dominant crystal peaks for the CKD were calcite ($CaCO_3$) and lime (CaO) with minor peaks for the quartz (SiO_2). On the other hand, both RHA and SF showed an amorphous nature. Figure 2 presents the morphology of the raw materials using the Scanning electron microscopy (SEM).

2.1.2. Sand

The fine aggregate utilized in this investigation was construction sand that confirms to the BS EN 196-1 (British Standard Institution, 2008).

2.1.3. Water

For all the mixtures, tap water of the Liverpool City provided by the United Utilities was used.

2.2. Research plan

This study was conducted with the aim of using PSA as base material to produce cement-free binder. This research investigated different paths to activate PSA. Therefore, the plan followed within this research consist of four stages. In the first stage, the effect of using 100% PSA as a binder material was investigated then the influence of the mechanical activation of PSA by grinding with and without FGD were investigated to get the best compressive strength. In the second stage, the effect of replacing PSA partially with 20% of either high alkalinity materials (PLFA and CKD) or pozzolanic materials (RHA and SF) was investigated. Based on the findings from the second stage, the third stage was conducted by using a combination of 60% PSA, 20% high alkalinity material and 20% pozzolanic material to develop ternary binder (TB). In all three stages compressive strength of mortar samples were conducted at the age of 3, 7 and 28 days. In the fourth stage a direct comparison of the performance of the TB against control cement was conducted, this included compressive strength testing at the age of 3, 7 and 28 days along with scanning electron microscopy (SEM) analysis.

2.3. Programme of testing

2.3.1. Compressive strength

For all the aforementioned stages, compressive strength was the test used for evaluating the performance of the binder in the form of mortar. The compressive strength test has been performed at the ages of 3, 7 and 28 days. For every blend, two samples with the size of $40 \times 40 \times 160$ mm have been produced and at the time of testing each sample was broken into two samples and the average of 4 readings was considered as the final value according to BS EN 196-1 (British Standard Institution, 2008).

2.3.2. Scanning electron microscopy (SEM) analysis

The SEM analysis was employed in this research to evaluate the hydration degree and detect the elemental composition of the developed TB binder at the age of 28 days and compare it with the conventional cement. For testing the SEM, EDX Oxford Inca x-act sensor with an accelerating voltage of 5–20 kV was utilised. Before conducting SEM testing, samples were gold-coated to improve the visibility.

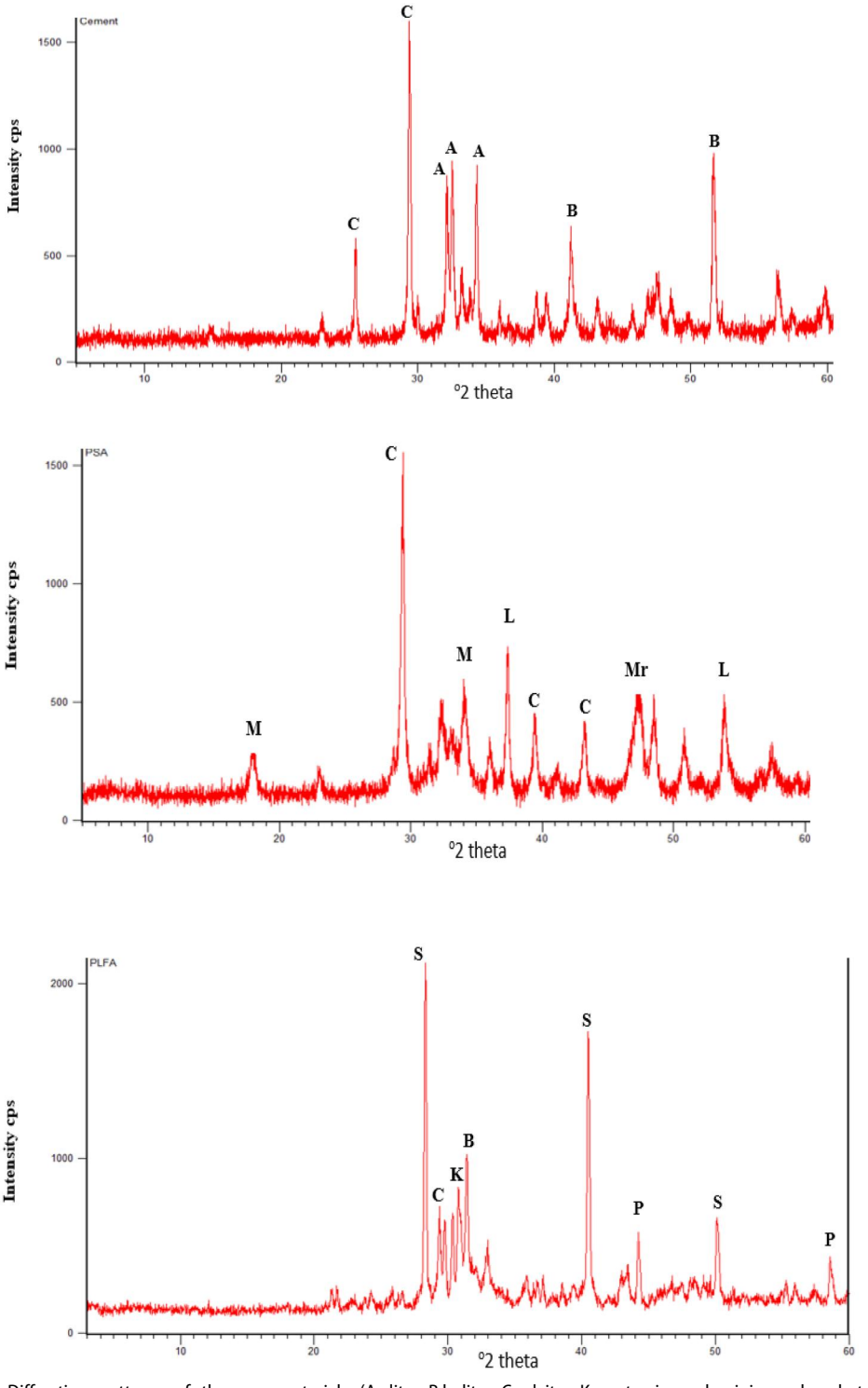

Figure 1. Diffraction patterns of the raw materials (A:alite, B:belite, C:calcite, K: potassium aluminium phosphate, L:lime, M: mayenite, Mr: merwinite, P:perovskite, Q:quartz, S:sylvite).

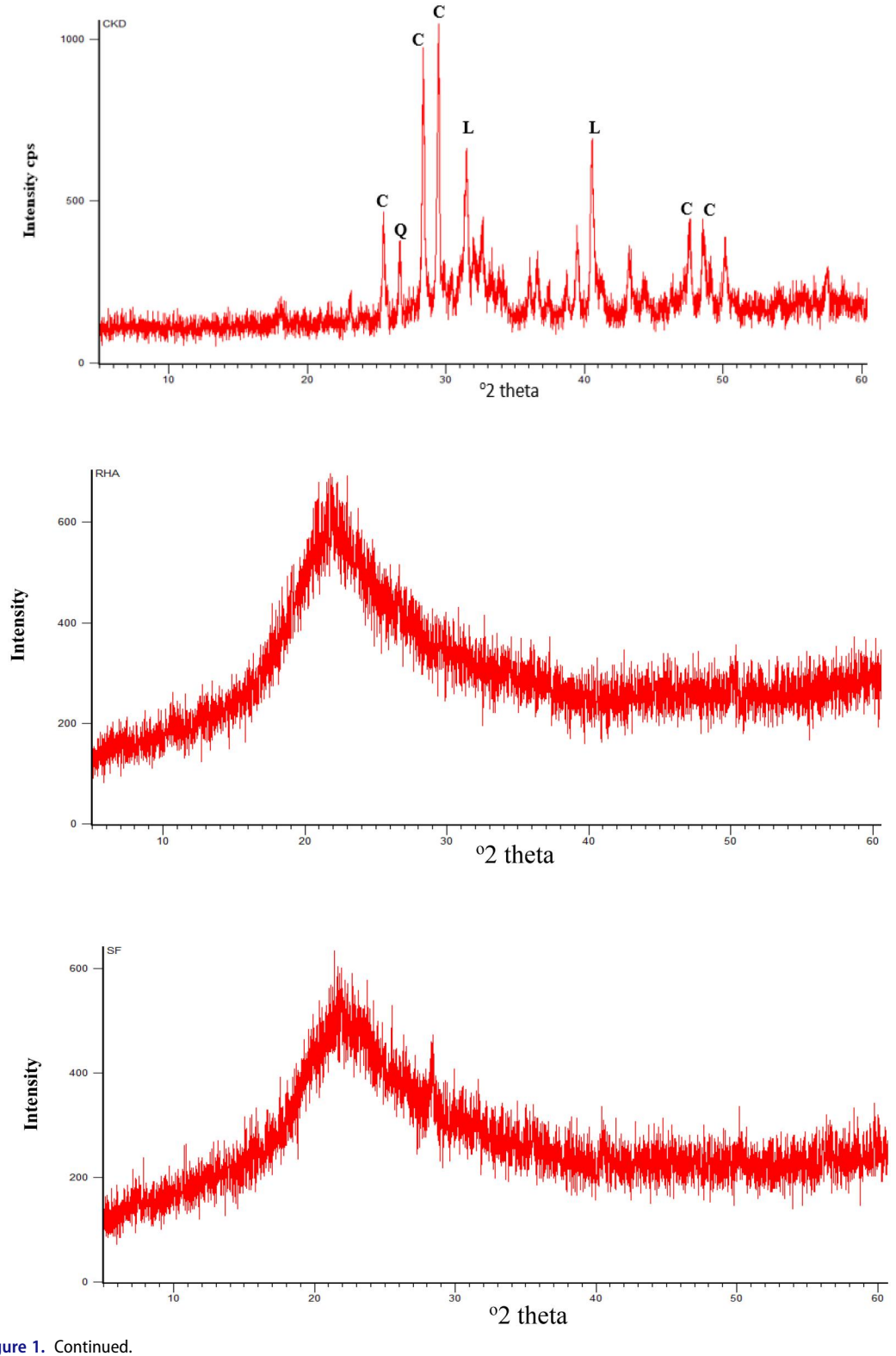

Figure 1. Continued.

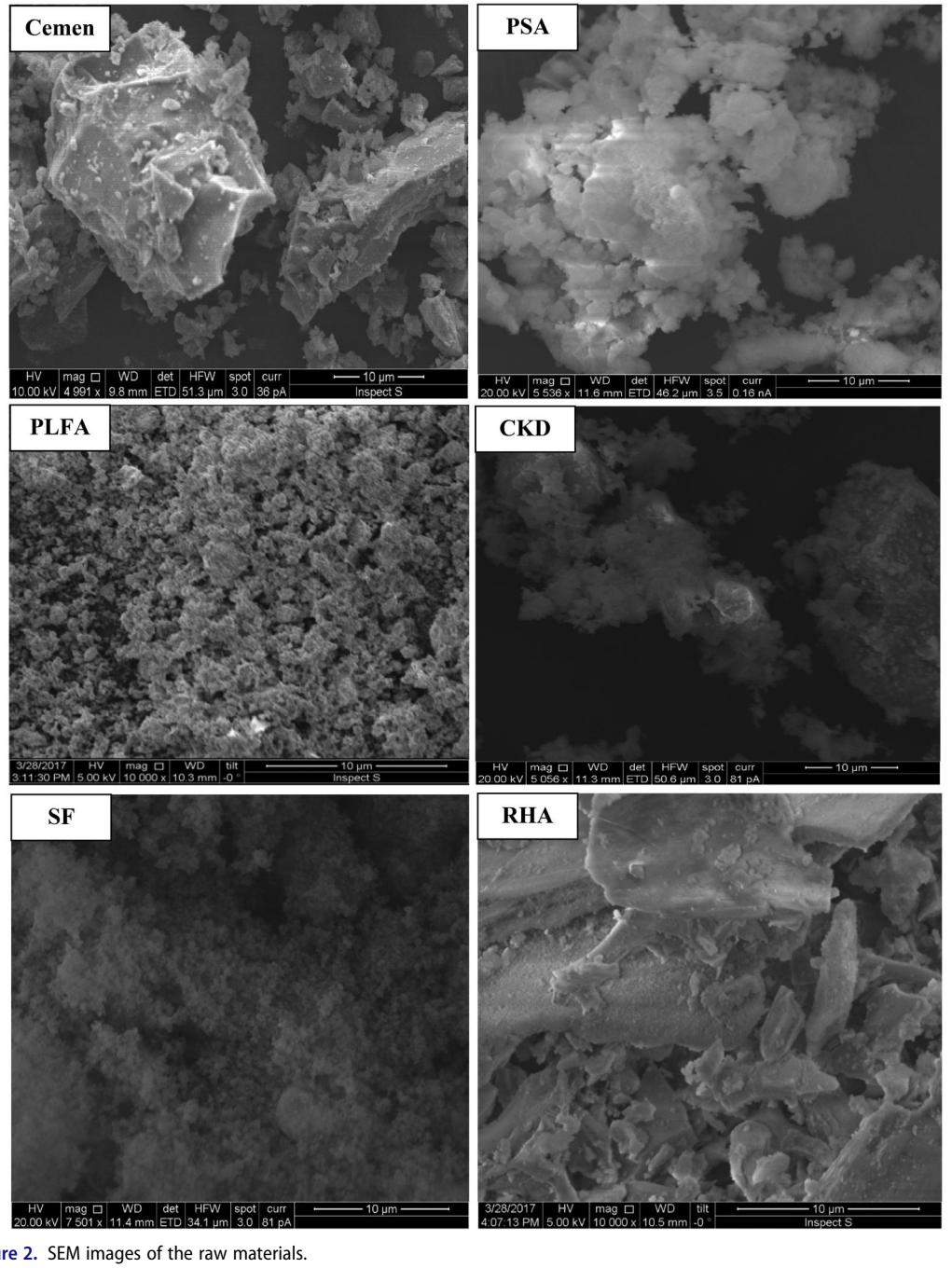

Figure 2. SEM images of the raw materials.

3. Results and discussion

3.1. Mechanical activation of PSA

In this section, the influence of grinding as a mechanical activation for the PSA was investigated by measuring the compressive strength of mortars made with 100% PSA as a binder after 3, 7 and 28 curing days. The technique of activation was chosen as grinding increases the finesses (higher SSA) and reduce the particle size of the of the raw materials (Jafer, 2017; Sadique et al., 2013) that can significantly

improve the compressive strength as reported by (Celik et al., 2008). The period of grinding of the PSA was set as 15 min taking into consideration sustainability and to avoid agglomeration of the PSA particles that would negatively affect the efficiency of grinding as claimed by (Sadique et al., 2012). Additionally, this section also investigated the influence of utilizing FGD gypsum to assist the grinding process of PSA as FGD gypsum controls the agglomeration and decreased the particles' strength (H. M. Jafer et al., 2018). FGD gypsum was added with a amount of 5% of the weight of the PSA as suggested by H. Jafer et al. (2018) and Sadique et al. (2013). In this section, three types of mortar samples were investigated. Table 2 presents details of the mixing proportions of the three samples. For all samples, the sand/binder (S/B) proportion was fixed as 2.25 and the water to binder (W/B) ratio was 0.60. Table 3 presents the influence of mechanical activation (grinding) on the physical characteristics of the PSA. It can be seen from Table 3 that grinding of the PSA resulted in the production of finer particles with median size (D_{50}) decreased from 28.21 μm for Untreated PSA to 5.33 and 4.23 μm for Ground PSA without FGD gypsum and Ground PSA with 5% FGD gypsum, respectively. Similar explanations were stated previously by Sadique and Al-Nageim (2012). Additionally, Table 3 shows that the SSA of the PSA was increased as a result of the grinding and the presence of the FGD gypsum resulted in higher SSA. The high SSA and finer particles obtained in the presence of FGD gypsum were attributed to the high fineness of FGD. A similar observation was obtained by Sadique et al. (2013).

Figure 3 presents the results of the compressive strength improvement of the three mixtures. It could be detected from Figure 3 that grinding the PSA resulted in better compressive strength at all selected

Table 2. Influences of PSA grinding on development of compressive strength.

Unary blends				
Untreated PSA	Ground PSA without FGD gypsum	Ground PSA with 5% FGD gypsum	Binder: sand ratio	Water/binder ratio
100%			1:2.25	0.6
	100%		1:2.25	0.6
		100%	1:2.25	0.6

Table 3. Influence of mechanical activation on the physical characteristics of the PSA.

	Density (g/cm^3)	D_{50} (μm)	BET SSA (m^2/g)
Untreated PSA	2.82	28.21	3.22
FGD gypsum	2.62	10.03	12.82
Ground PSA without FGD gypsum	2.88	5.33	3.71
Ground PSA with 5% FGD gypsum	2.88	4.23	4.68

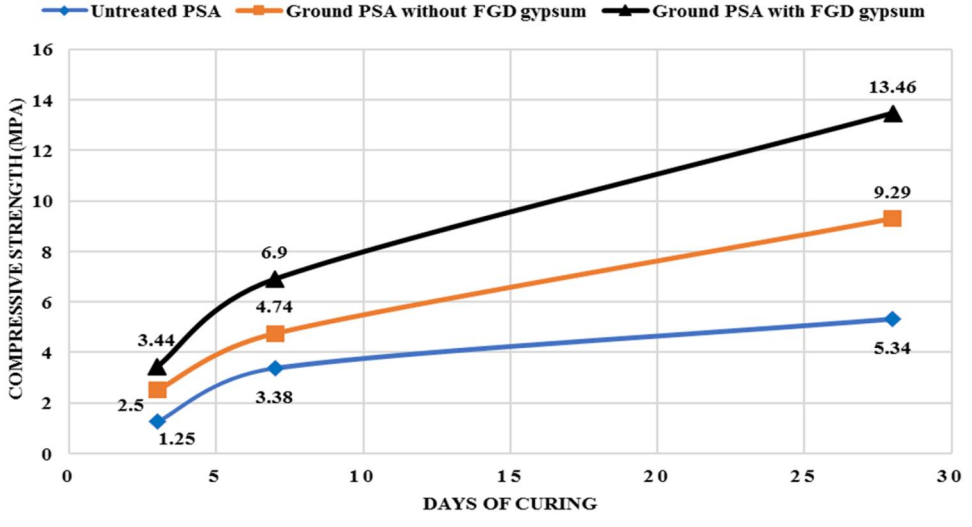

Figure 3. Influence of PSA grinding on development of the compressive strength.

curing ages of relative to untreated PSA samples with an enhancement in the compressive strength of about 174% at the age of 28 days. This can be since the hydration reaction continuous as high fineness materials gave more surface area to enhancement the hydration reaction that in turns aided in increasing the cementitious gel produced, therefore; the compressive strength improves as reported by Chindaprasirt et al. (2014) and Sobolev et al. (2016). Additionally, the highest compressive strength at all ages of curing was achieved for samples incorporated FGD gypsum with an improvement of about 252 and 145% relative to samples with untreated PSA and ground PSA without FGD gypsum, respectively. This could be recognized to the FGD gypsum function as a grinding aid where it could depolymerize the PSA glassy phase and certified more breakthrough of SO_4^{2-} and Ca^{2+}. This was also associated with an exchange of glass network modifiers that thus leaded to accelerated PH. This, therefore, improved the PSA glass phase dissociation and the creation of C-A-H and AFt products from the early curing ages (H. M. Jafer et al., 2018; Zhang et al., 2016).

3.2. Chemical activation of PSA by blending

After the improvement in the mortar performance due to the combined effect of grinding and the addition of FGD gypsum, further investigation was performed to investigate the influence of chemical activation of PSA by blending it with other materials as blending has several useful functions, including enhancing the strength relative to the single system of blending (H. Jafer et al., 2018). The chemical activation could be conducted by blending the base material (PSA) with high alkalinity and/or pozzolanic materials as reported by H. Jafer et al. (2018). In this section, the PSA was blended with poultry litter fly ash (PLFA) and cement kiln dust (CKD) as materials with high alkalinity and with silica fume (SF) and rice husk ash (RHA) as pozzolanic activators. Details of the mixing ratios for the explored binary systems are demonstrated in Table 4. Similar to the previous section, the S/B ratio and W/B ratio for all samples were fixed as 2.25 and 0.6, respectively. Table 5 presents the physical properties of the raw materials.

To investigate the performance of mortars made with four various binary blends, the compressive strength test was performed after 3, 7 and 28 days of curing. Figure 4 presents the results of the compressive strength development of the binary blends. It could be detected from Figure 4 that the compressive strengths of all the binary blends were greater than that of the unary blend (PSA ground with 5% FGD gypsum) at all ages of curing except the binary mixture (0.8 PSA + 0.2 CKD) that showed lower compressive strength at the 28 days curing age. This decrease in the compressive strength could be due to the larger particles of CKD relative to PSA ground with 5% FGD gypsum that could delay the mortars performance throughout hydration reactivity and that decrease the compressive strength (Shubbar et al., 2018). Figure 4 also shows that the incorporation of 20% PLFA resulted in enhanced compressive strength of the binary mix by about 20% relative to the unary blend (PSA ground with 5% FGD gypsum). This performance can be attributed to the alkaline activation of the PSA particles due to the high pH and alkali content of the PLFA that breaks the glass phase of the PSA and boost the dissolution of PSA by decreasing the amount of Al^{+3} and Ca^{+2} in the system to form solidify and ettringite the system (Al-Nageim et al., 2013; Konsta-Gdoutos & Shah, 2003; Sadique et al., 2013).

Table 4. Mix proportions of the binary mixtures.

Mixture	PSA ground with 5% FGD gypsum	PLFA	CKD	RHA	SF
0.8 PSA + 0.2 PLFA	80%	20%			
0.8 PSA + 0.2 CKD	80%		20%		
0.8 PSA + 0.2 RHA	80%			20%	
0.8 PSA + 0.2 SF	80%				20%

Table 5. The physical characteristics of the raw materials.

	Density (g/cm^3)	D_{50} (μm)	BET SSA (m^2/g)
PLFA	2.54	9.21	8.82
CKD	2.98	26.95	4.2
RHA	2.31	24.9	26.7
SF	2.69	26.84	26.21
Ground PSA with 5% FGD gypsum	2.88	4.23	4.68

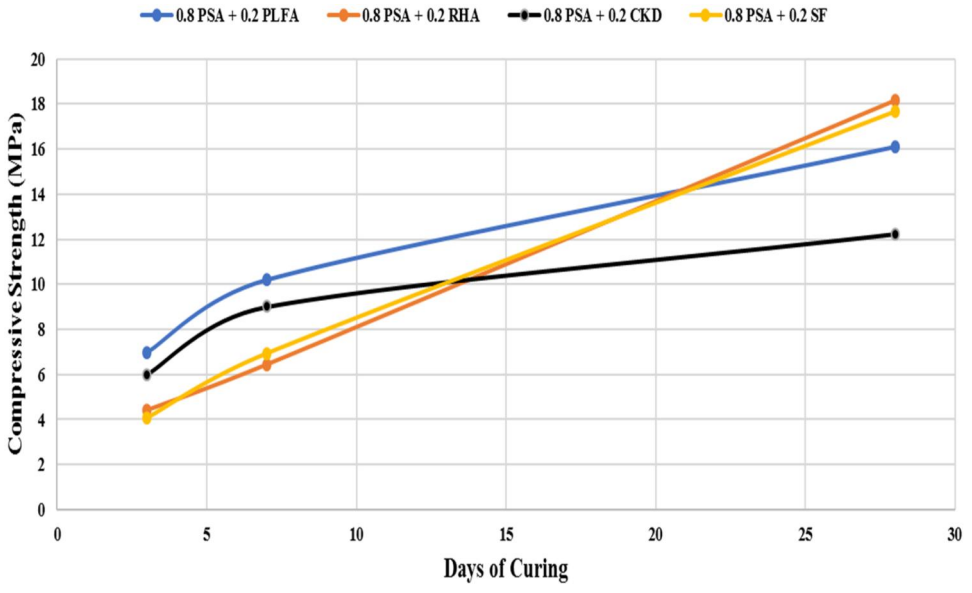

Figure 4. Improvement of compressive strength of the binary blends.

Table 6. Constitutive matrix of ternary blend and cement samples.

Blend	PSA ground with 5% FGD gypsum	Cement	PLFA	RHA	W/B	S/B
TB	60%	0%	20%	20%	0.6	2.25
Cement	0%	100%	0%	0%	0.35	2.25

Table 7. Chemical compositions of cement and TB.

	Cement	TB
Na_2O %	1.534	1.710
CaO %	62.389	43.169
SiO_2 %	26.638	33.296
Al_2O_3 %	2.436	3.943
MgO %	1.573	1.804
Fe_2O_3 %	1.744	0.516
SO_3 %	1.887	3.228
K_2O %	0.723	3.958
pH	12.9	11.78
Density (g/cm^3)	3.010	2.698
D_{50} (μm)	13.360	9.360
BET SSA (m^2/g)	6.741	9.912

Regarding the pozzolanic activation of the PSA, Figure 4 showed that blending RHA or SF with the PSA resulted in enhancing the compressive strength. This enhancement in the compressive strength can be due to the creation of extra Calcium Silicate Hydrate (C-S-H) gel as a results of the pozzolanic reaction between the silicate from the RHA or SF with hydrated lime produced from the PSA and also due to the successful conversion of soluble calcium hydroxide (C-H) that created during the hydraulic reaction, into dense C-S-H gel (Aïtcin, 2016; H. M. Jafer et al., 2018).

3.3. Development of ternary blend

According to the results from previous section, PLFA and RHA have provided the greatest compressive strength for the high alkalinity and pozzolanic materials, respectively. Therefore, this section was devoted to investigating the production of Ternary Blend (TB) using PSA + PLFA + RHA. As 20% of RHA and 20%

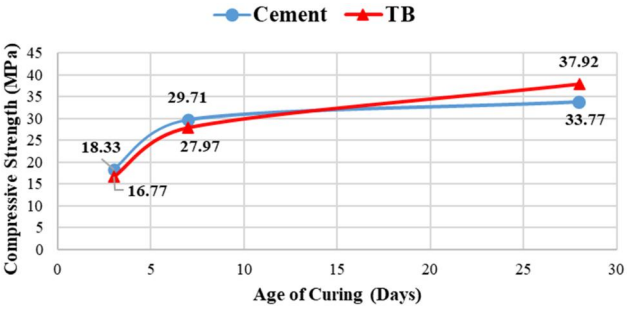

Figure 5. Compressive strength enhancement of the TB and cement.

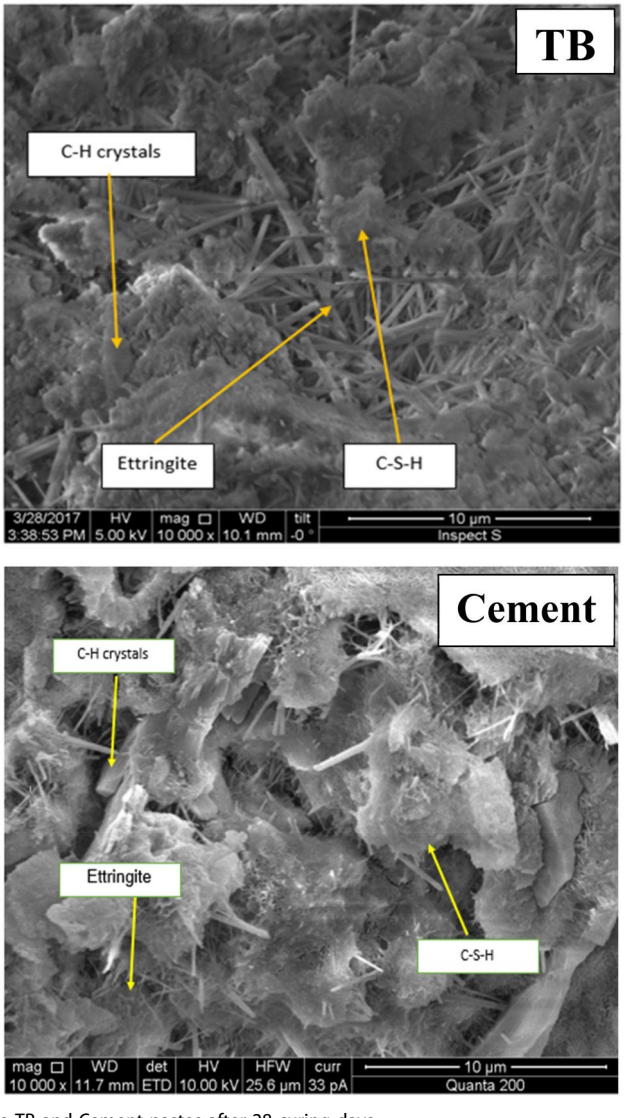

Figure 6. SEM images of the TB and Cement pastes after 28 curing days.

PLFA have showed an improvement in the compressive strength during the binary blends, therefore, these percentages were used in the ternary system (Table 6). In order to conduct a direct comparison with the TB, a cement mortar was prepared with 100% Cement, in which the S/B was similar to that of the ternary mixture (2.25), while the W/B ratio was 0.35 in order to obtain flow value for both mixtures in the range of (210 ± 5 mm). Similar approach was followed by Sadique et al. (2019). A comparison of the physical and chemical characteristics of the TB and the cement is presented in Table 7.

The findings of the compressive strength development of the TB and Cement mortars after 3, 7 and 28 curing days are demonstrated in Figure 5. As illustrated in Figure 5, the compressive strength of both mixes were improved with increasing the age of curing. At early curing ages (3 and 7 days), the compressive strength of the TB specimens was very similar to that of the cement mortar. The early strength of the TB mortars is believed to be due the increased concentration of SO_4^{-2} that react with the Al_2O_3 from the PSA and forms aluminosulphate, that in turs react with Ca^{2+} and create ettringite, which contributes strength at early ages (Qian et al., 2001; Sadique et al., 2019). The other reason is believed to be due to the presence of RHA that promotes pozzolanic reaction with calcium hydroxide from the PSA and creates C-S-H gel (H. Jafer et al., 2018; Sadique et al., 2019). At 28 curing days, the TB samples illustrated about 12% higher compressive strength relative to the cement mixture. This successful improvement in the compressive strength occurred since greater SSA and smaller particles of the TB relative to cement and the presence of a balanced oxide composition within the TB along with its high alkaline content that all leads to boost the chemical activation of PSA that in turns resulted in the creation of additional C-S-H gel (H. Jafer et al., 2018; Sadique et al., 2019). Usually gel is filling pores and grow into capillary spaces, which lead to increase dense and structural strength (Blanco et al., 2006; Shubbar et al., 2018).

3.4. Scanning electron microscopy (SEM) testing

The SEM analysis was employed in this research to evaluate the hydration degree of the developed TB binder and compare it with conventional cement. The SEM micrograph of the pastes of (TB and cement) after 28 days of water curing is presented in Figure 6. It could be detected from Figure 6 that both binders show the formation of the platy shape of Portlandite (CH) and the needle shape particles (Ettringite) along with the formation of C-S-H gel, which is the main strength-generating product (Sadique et al., 2012; Shubbar et al., 2018; 2020).

4. Conclusions

The main aim of this investigation was to utilise PSA as a base material to produce a ternary blend cement-free binder material for construction. Depending on the obtained findings, the following conclusions were drawn:

1. At the age of 28 days, grinding the PSA in the presence of FGD resulted in enhancing the compressive strength of mortars made totally from PSA by 252% (13.46 MPa) relative to mortars made with untreated PSA (5.34 MPa).
2. The chemical activation of the PSA with pozzolanic materials (SF and RHA) has improved the compressive strength, The improvement was by about 131% for SF and 134% for RHA relative to that of the unary PSA system at the age of 28 days.
3. The chemical activation of the PSA with high alkalinity material (PLFA) has improved the mechanical performance by 2.64 MPa relative to that of the unary PSA system, while the CKD resulted in reducing the compressive strength by 1.22 MPa at the age of 28 days.
4. Based on the useful roles of the ternary mixing, RHA and PLFA were added to the PSA that resulted in developing a ternary blend cement-free material with enhanced mechanical performance (37.92 MPa) relative to conventional cement (33.77 MPa) after 28 days of curing.
5. The results from the SEM analysis show the presence of morphological components (Ettringite, C-S-H gel and CH crystals) of the TB and it was comparable with that of the reference cement.
6. The production of cement-free binder material totally from waste and/or by-product materials with comparable properties to that of conventional cement would help in both reducing the negative

environmental impact of cement production and reduce the volume of waste materials being send to landfills.

Disclosure statement

The authors declare no conflict of interest.

Data availability statement

Data will be available on request.

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