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1 Long-term performance of novel high-calcium one-part alkali-activated  
2 cement developed from thermally activated lime kiln dust

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9  
10 **Highlights**

- 11 • One-part earth alkaline activated dry cementitious mixture was produced  
12 using thermally activated lime kiln dust (LKD) waste material as a source of  
13 CaO activator and metakaolin and natural pozzolan as a source of alumina-  
14 silicate
- 15 • The proportion of LKD and metakaolin in the ternary blend and thermal  
16 activation of LKD not only increased the reactivity but also induced higher  
17 degree of alkali-activation.
- 18 • Significant increase in the rate of strength development after 28 days of curing  
19 and continued until 180 days that reached 51 MPa for increased proportion of  
20 950°C calcined LKD and metakaolin in the blend was revealed.
- 21 • Considerable changes in mineralogy and amorphicity were evidently  
22 accredited to thermal treatment.
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30 Abstract

31 The traditional activation approach for alkali-activated cement AAC has several problems  
32 resulting mainly from the hazardous and corrosiveness of the alkaline chemicals, such as (NaOH,  
33 Na<sub>2</sub>SiO<sub>3</sub>), which in turn impede the utilisation of AAC in the construction fields. In this study, A  
34 second generation of alkali activated binder was developed using Metakaolin (MK) and natural  
35 pozzolan material (NP) (as a source of alumina-silicate), these materials were activated using high-  
36 calcium lime kiln dust as solid activator to transform the alumina-silicate crystalline phases to  
37 cementitious hydrated products. This was achieved with the aid of heat treatment of materials at  
38 different temperatures. Raw materials and final AAC samples were characterised using analytical  
39 methods, such **X-Ray powder diffraction (XRD)**, Thermogravimetric Analysis (**TG-DTA**), Fourier  
40 Transform Infrared Spectroscopy (**FTIR**) and Scanning Electron Microscope (**SEM**). Additionally,  
41 long-term compressive strength, chemical and microstructural performance were investigated. The  
42 transformation of raw materials from crystalline to amorphous phases happened due to the effect  
43 of the heat treatment and the formation of stratlingite products in the final AAC paste, which were  
44 evidenced using the mentioned characterisation methods. The findings of the present study proved  
45 that the compressive strength of the new binder reached 27 MPa and 51 MPa after 28 and 180 days  
46 of curing, respectively, **ensuring a progressive as well as a higher degree of alkali-activation and**  
47 **disappearance of unreacted alkaline substances in the final AAC products.**

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52 Keywords

53 Alkali activated cement, heat treatment, Lime waste, XRD, TG-DTA, FTIR.

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## 63 1. Introduction

64 The world production of the Ordinary Portland Cement OPC is significantly growing, where the  
65 estimated global production in 2018 was 4.1 billion metric tonnes [1]. The use of OPC in concrete  
66 construction is under serious evaluation because of the substantial challenges that are facing the  
67 method of production of conventional cement. Firstly, the high energy consumption through the  
68 whole process [2]. Secondly, a high quantity of carbon dioxide gas (CO<sub>2</sub>) is released to the  
69 atmosphere during the production of cement [3]. To overcome these challenges, several  
70 investigations were commenced to develop other types of cements that are entirely free of OPC  
71 and based principally on supplementary materials. Mineral Products Association (MPA), in the  
72 UK, listed a number of novel low carbon (non-Portland) cements with low energy of production  
73 in their fact sheet [4]. One of these novel cements is Alkali Activated Cement AAC, which is a  
74 cementitious material formed as a result of an alkaline activation of amorphous or vitreous  
75 alumina-silicates. When mixed with alkaline activators (solid or liquids), in a chemical reaction  
76 called alkali-activation or alkanination, these materials set and harden, yielding a material with  
77 good binding properties [5,6]. There is a conventional two-part alkali activation, wherein a solid  
78 raw material is activated with alkaline chemical solution. This solution is categorised as extremely  
79 corrosive materials. From an operational viewpoint, they are difficult and expensive to handle,  
80 with significant occupational health and safety concerns. On the other hand, the activator solution  
81 provide the highest single contribution to the embodied carbon dioxide of AAC [7–9]. Until now,  
82 it cannot be said that AAC is based on user friendly and low carbon process, as the production of  
83 the chemical solutions releases large amounts of CO<sub>2</sub> to the environment [10].

84 Several efforts have been made to develop a second generation of geopolymers with one-part AAC  
85 that can be found under different terminologies in the literature, such as one-part alkali activated  
86 cement, self-activating cement or one-part alkali alumina-silicate hydraulic cement [11–13]. In  
87 one-part AAC, only a dry mixture is needed in addition to water. The dry mixture is prepared by  
88 mixing a solid alkali-activator with a solid alumina-silicate precursor, with or without an assisted  
89 activation approach [8,12].

90 Provis [14] defined the activators as any substance that represents an alkaline source that raises  
91 the pH of the reaction mixture and simplifies dissolution. This includes alkali cations (Na<sup>+</sup>, K<sup>+</sup>) or  
92 earth alkaline cations (Ca<sup>+2</sup>, Mg<sup>+2</sup>). A significant body of literature explored the synthesis of one-  
93 part AAC using alkaline commercial solid chemical activators, including sodium silicates  
94 (Na<sub>2</sub>SiO<sub>3</sub>) and sodium carbonates (Na<sub>2</sub>CO<sub>3</sub>). Realistic reports revealed that these chemicals are  
95 expensive and primarily contribute to the total cost of production of AAC [9,15–17]. Therefore,  
96 many synthetic chemicals do not represent a realistic commercial or environmental optimised  
97 solution for use as an activator [8].

98 Previous studies focused on the use of (CaO) and Ca(OH)<sub>2</sub> as potential alternative activators, as  
99 these activators are cheaper than (Na<sub>2</sub>SiO<sub>3</sub>) and (Na<sub>2</sub>CO<sub>3</sub>) [17,18]. These activators provide  
100 alkaline earth cations in place of alkali cations, which ease the creation of different binding phases,

101 when compared to blends of low calcium content [8,19]. **Cabrera and Rojas [20]** investigated the  
102 reaction kinetics of metakaolin-lime in water at 60°C, using thermal analysis. The study confirmed  
103 the appearance of reaction products, mainly strätlingite ( $C_2ASH_8$ ) and ( $C_4AH_{13}$ ), as the reason for  
104 binder strength development. Kim et al. [17] used synthetic commercial CaO solid powder and  
105 ground granulated blast furnace slag (GGBS) (as the Si/Al source material) to produce non-cement  
106 binder . When compared to Ca (OH)<sub>2</sub>, CaO activator was found to yield a higher mechanical  
107 strength, 53 MPa after 56 days, due to the production of more calcium silicate hydrate (C-S-H)  
108 than Ca (OH)<sub>2</sub>. Nevertheless, when investigating the energy and ecological analysis of  
109 synthesising high purity chemical CaO powder, it was discovered that the synthesis process is  
110 complex, consuming high energy and it requires a high level of accuracy [21-22]. Different  
111 activation assisted methods were used in the production of a one-part AAC. For example, thermal  
112 activation (calcination) has been recently used as an assisted approach to improve the reactivity  
113 and the amorphicity of supplementary cementitious materials, such as natural pozzolans [11,23].  
114 During the thermal treatment of undisturbed material, crystalline bonds are broken down and  
115 transformed to a glassy or amorphous phase, which results in a more reactive binder. When  
116 reviewing recent attempts directed to develop the one-part AAC, limited research projects have  
117 been devoted to investigate the long-term performance and properties of these materials, especially  
118 in the presence of unreactive alkaline substances. In fact, the latter must be precisely investigated  
119 as the excessive unreacted alkaline solution can be diffused to a concrete surface, and reacts with  
120 CO<sub>2</sub> in the air, forming efflorescence on the surface and degrading the appearance and strength of  
121 concrete [24].

122 Recently, Abdel-Gawwad et al.[25] carried out a study about one-part AAC comprised of GGBS  
123 and concrete waste treated by dry NaOH, with the addition of lead bearing sludge. The study  
124 assessed the compressive strength of hardened one-part AACs after 120 days, which was 44 MPa  
125 for a blend containing 65.9 wt. % GGBS, 15 wt. % lead bearing sludge, 34.10 wt. % concrete  
126 waste, 3.10 wt. % NaOH, 2 wt. % Na<sub>2</sub>O, and water/powder ratio of 0.29. On the other hand, there  
127 is no study until now for evaluating the long-term strength of one-part AAC, activated by earth  
128 alkaline cations, such as CaO or MgO, embodied in the material.

129 The principal aim of this work is to develop of novel approach to produce a second-generation  
130 alkali-activated cement, that is completely free of commercial chemical activators. This was  
131 achieved by providing the earth alkaline source from lime kiln dust (LKD), which is primarily  
132 composed of reactive CaO. LKD is a by-product material of quick lime production process, which  
133 is usually disposed of in landfills worldwide. For instance, in the USA, it is estimated that about  
134 2.5 million metric tonnes of high-calcium LKD is produced annually [26]. Thus, LKD must be  
135 reused to minimise its environmental impacts, and to meet the sustainability requirements. LKD  
136 is mainly composed of a CaO compound and high alkaline materials (high pH). Metakaolin, which  
137 is the crystallo-graphically disordered layered product of dehydroxylation of kaolinite (an alumina-  
138 silicate clay) [27], was combined with volcanic tuff or natural pozzolan(as an alumina silicate  
139 (Al/Si) source) to formulate the one-part dry AAC powder that just needs water to induce the  
140 alkali-activation process. Volcanic tuffs provide an extensive variety of reactivity, depending on

141 their degree of crystallinity and mineralogy. The proposed alumina-silicate precursors, undertake  
142 dissolution and precipitation processes when mixed with lime or CaO, yielding calcium alumina-  
143 silicate hydrate (C-A-S-H) [28]. Furthermore, the current research examines the long-term (after  
144 180 days) mechanical and physico-chemical properties of the novel AAC cement, which helps to  
145 assess the sustained progression of alkali-activation in the absence of unreacted alkaline.

146

## 147 **2. Materials and methods**

### 148 2.1 Materials

149 The alumina-silicate source was produced by combining metakaolin (MK) and natural pozzolan  
150 (NP) from volcanic tuff material. LKD has been used as a source of alkaline material in the  
151 proposed blend. The elemental composition of raw materials was determined as they were received  
152 condition by using a Shimadzu EDX 720 and energy dispersive X-ray fluorescence (EDXRF)  
153 spectrometer. The key oxide elements and the physical properties of raw materials are listed in  
154 Table 1. Both MK and NP are composed of similar compositions of minerals, which could be  
155 considered as an ideal source of siliceous and aluminate materials, with minor quantities of CaO,  
156 Na<sub>2</sub>O, K<sub>2</sub>O and MgO in NP. LKD was mostly rich in CaO (about 80.1wt.%) with slight amounts  
157 of SiO<sub>2</sub> and Na<sub>2</sub>O, and therefore it was considered as calcareous source. The specific surface area  
158 was determined by air permeability test (Blaine method) that is described in BS EN 196-6 [29]. It  
159 was found that MK and NP have surface area of 19.6 and 17.2 m<sup>2</sup>/g, respectively, while LKD has  
160 10.1 m<sup>2</sup>/g, LKD has high alkalinity with pH of 12.3, which confirms its suitability to be alkaline  
161 activator. MK and NP have nearly the same density; 2.69 and 2.57 g/cm<sup>3</sup>, respectively, while LKD  
162 has a density of 2.7 g/cm<sup>3</sup> as measured by Quantachrome gas expansion multi-pycnometer purged  
163 with helium gas.

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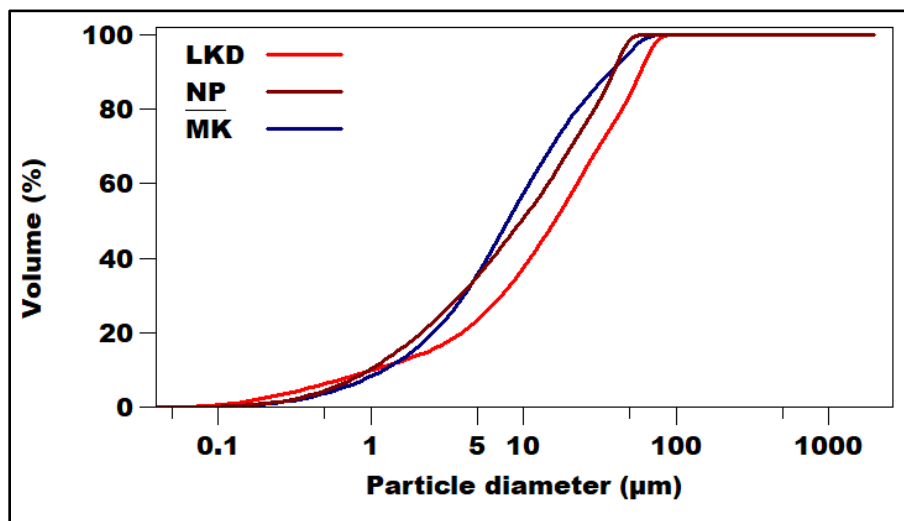
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Table 1. Chemical and physical properties of as received materials.

Chemical composition (Wt.%)	Raw materials		
	MK	NP	LKD
SiO <sub>2</sub>	55	46.6	14.6
Al <sub>2</sub> O <sub>3</sub>	40	30.4	0.2
Fe <sub>2</sub> O <sub>3</sub>	1.4	3.8	0.1
CaO	0.15	4.5	80.1
Na <sub>2</sub> O	0.4	3.9	1.8
K <sub>2</sub> O	0.4	6	0.5
MgO	0.95	4.2	0.6
TiO <sub>2</sub>	1.7	0.6	0.1
<b>Physical Properties</b>			
Specific surface area (Blaine) (m <sup>2</sup> /g)	19.6	17.2	10.1
Density (g/cm <sup>3</sup> )	2.69	2.57	2.7
pH	6	6	12.3

176

177 Particle size distribution (PSD), was determined using Beckman Coulter laser diffraction particle  
 178 size analyser, as displayed in Fig. 1. It has been found that MK and NP have similar size  
 179 distributions with D<sub>50</sub> of 9.22 and 7.87μm, respectively. Conversely, LKD was found to be coarser,  
 180 with a D<sub>50</sub> of 15.94μm. This particle size range of raw materials has a high positive effect on alkali  
 181 activation. Rashad et al. [30], reported that MK with grain size less than 32 μm yields a workable  
 182 mix with higher compressive strength.



183

184

Figure 1. Particle size distribution (PSD) of starting materials.

## 185 2.2 Testing methods

186 The set of experimental tests that have been conducted in this study comprises of fresh properties  
187 indexed by the setting time, mechanical properties indexed by the compressive strength, thermal  
188 analysis presented by thermogravimetric analysis (TGA) and microstructural analysis introduced  
189 by x-ray diffraction (XRD), Scanning Electron Microscope (SEM) and Fourier transform infrared  
190 spectroscopy (FT-IR).

### 191 2.2.1 Setting time

192 The standard consistency and setting time of the developed one-part AAC binder were investigated  
193 according to British standard 196-3 [31].

### 194 2.2.2 Compressive strength

195 The compressive strength of mortar prisms, was used as a function for evaluating the mechanical  
196 properties of specimens and was measured according to the British standard 196-1 [32]. The test  
197 was carried out using a Control Automax 5 compression tester, with a load rate of 0.4 MPa/sec.

### 198 2.2.3 Thermal analysis (TGA)

199 Thermal analysis used in this investigation, consisted of Thermogravimetric analysis (TG), which  
200 is usually used to determine weight loss of the samples that subjected to thermal events. However,  
201 this technique does not detect phase change, such as “melting”; thus, differential thermal analysis  
202 (DTA) is employed, which is the first derivative of the weight loss curve. DTA indicates phase  
203 changes as endothermic and exothermic peaks [33]. Therefore, it is donated as TG-DTA technique.  
204 The tests were completed using the Perkin Elmer TGA Q50 V20.13 Build 39. The thermal  
205 performance of materials was investigated by TG-DTA, with a heating range 20-900 °C and a  
206 heating rate of 10 °C/min. In this study, TG-DTA investigations were performed on raw materials  
207 to assess their thermal behaviours during individual calcination.

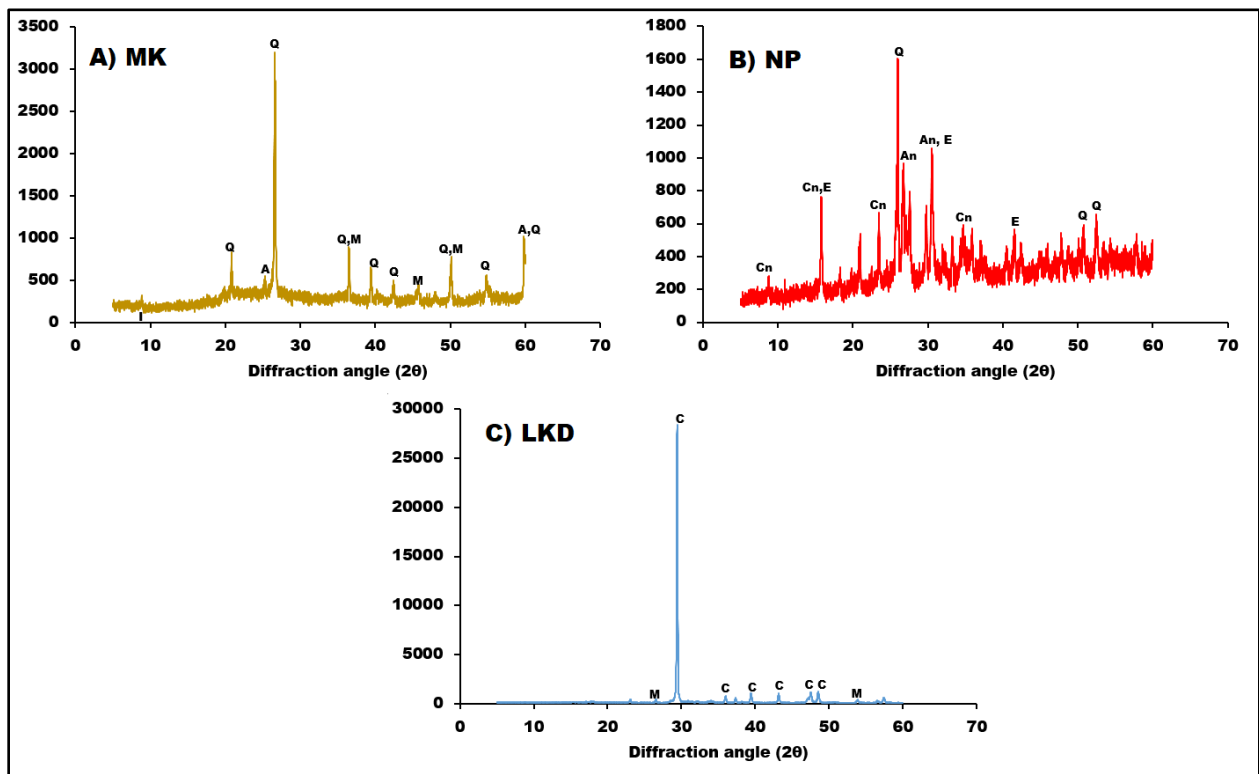
### 208 2.2.4 Microstructural analysis

209 The microstructural and morphological analyses were carried out on the raw materials before and  
210 after thermal treatment. Additionally, the selected pastes of AAC, with the highest strength after  
211 they have been hydrated and hardened, were finely ground, dried, and have gone through XRD,  
212 SEM and FT-IR analysis after 28, 90 and 180 days of curing. The XRD test was carried out using  
213 a Rigaku mini-flex diffractometer (mini-flex goniometer), with CuK X-ray radiation (30 kV  
214 voltage and 15mA current at scanning speed of 2.0 deg./min in continuous scan mode) and  
215 scanning range ( $2\theta$ ) of 5–60°. Scanning Electron Microscope (SEM) morphological analysis and  
216 observations were conducted using an Oxford Inca x-act detector (45nA prob current and 100 sec  
217 counting time) and a FEI Company SEM model Inspect S (20kV accelerating voltage). Fourier  
218 transform infrared spectroscopy (FT-IR) analysis was carried out using a Perkin-Elmer Spectrum  
219 BX series Fourier transform infrared spectrometer (FT-IR), equipped with a Miracle ATR  
220 accessory (Specac, UK). The spectrum of the sample was recorded using accumulating 16 scans  
221 at 4 cm<sup>-1</sup> resolution and wavelength between (550 cm<sup>-1</sup> and 2000 cm<sup>-1</sup>).



222 2.3 Characterisation of raw materials

223 The powder (XRD), (SEM) and (FT-IR) analyses of undisturbed materials are illustrated in Figs.  
224 2 to 4. From the X-Ray diffractions of MK, Fig. 2, it is observable that MK comprises many  
225 crystalline phases and is primarily composed of quartz ( $\text{SiO}_2$ ) and Mullite ( $\text{Al}_6\text{Si}_2\text{O}_{13}$ ), in major  
226 crystalline peaks. Illite ( $\text{K}, \text{H}_3\text{O}$ ) ( $\text{Al}, \text{Mg}, \text{Fe}$ ) $_2(\text{Si}, \text{Al})_4\text{O}_{10}[(\text{OH})_2, (\text{H}_2\text{O})]$  and anatas ( $\text{TiO}_2$ )  
227 compounds in minor peaks. The highest quartz peak occurred at ( $2\theta$ )  $26.73^\circ$ . Diffractogram  
228 patterns of NP, which have closely crystalline peaks of quartz peak and feldspars, such as anorthite  
229 ( $\text{CaAl}_2\text{Si}_2\text{O}_8$ ) and clinoptilolite ( $\text{KNa}_2\text{Ca}_2(\text{SiO}_{29}\text{Al}_7)\text{O}_{72} \cdot 24\text{H}_2\text{O}$ ). Additionally, edenite  
230 ( $\text{Ca}_2\text{NaMg}_5(\text{AlSi}_7)\text{O}_{22}(\text{OH})_2$ ) was present in NP diffractions with high quantities. Diffractions  
231 of LKD, showed one recognised crystalline peak of calcite ( $\text{CaCO}_3$ ) with a higher intensity.  
232 Smaller peaks exist in the forms of mullite. The presence of such compounds increases alkalinity  
233 activity in LKD; these compounds are similar to the content of commercial activator water glass  
234 ( $\text{Na}_2\text{SiO}_3$ ).



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Figure 2. Powder XRD-patterns of initial materials

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*Q: Quartz, M: Mullite, A: Anatas, I: Illite, Cn: Clinoptilolite, An: Anorthite, E: Edenite, C: Calcite.*

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SEM images illustrate the particles of original materials, as shown in Fig.3. MK seems to have fine and lamellar particles with random non-uniform shapes. Particles of LKD look coarser and have less surface area with flaky shapes. The raw NP particles, as shown in the SEM image, are irregular in shape and size.

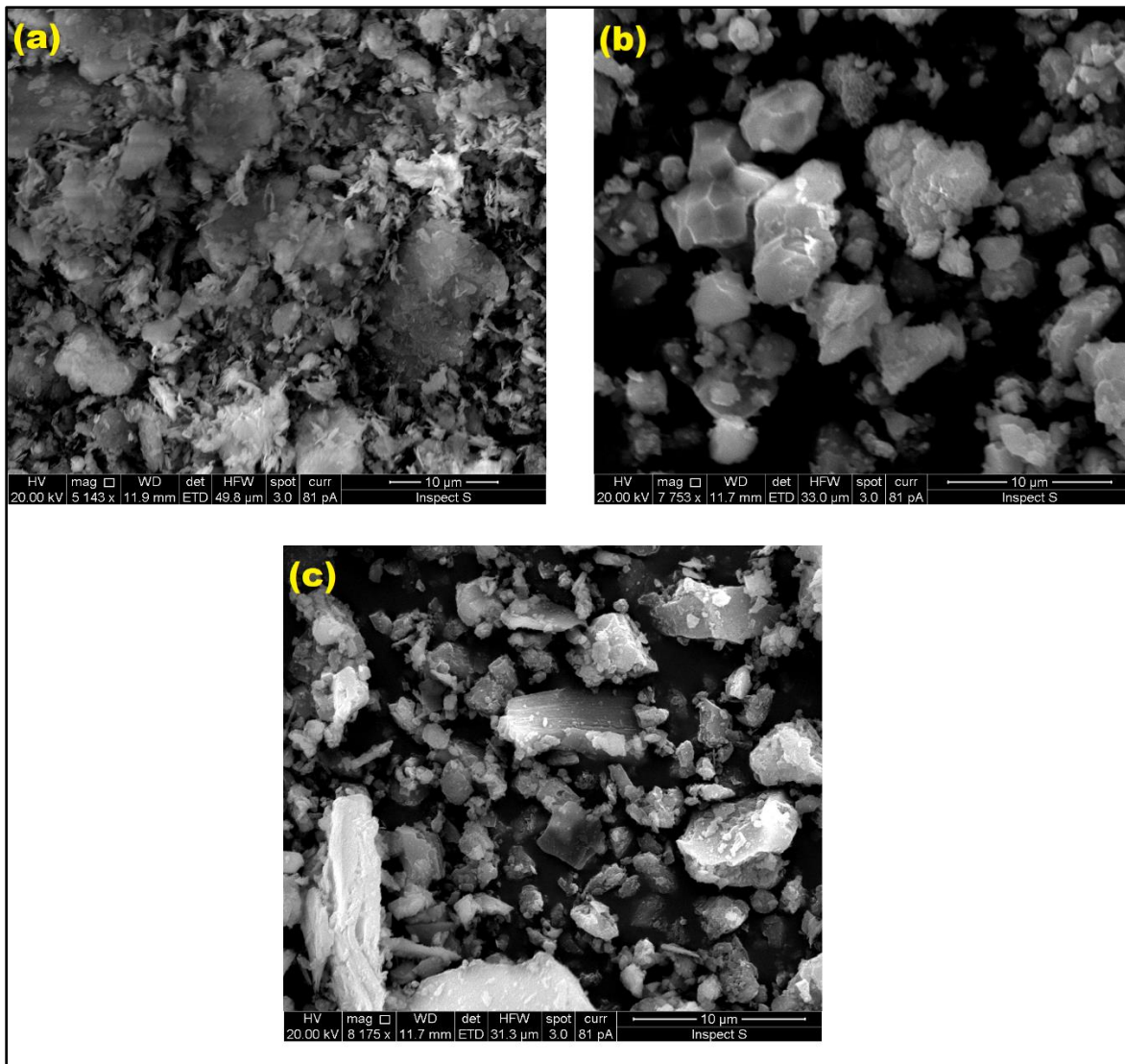


Figure 3. SEM micrographs of undisturbed materials a) MK, b) LKD and c) NP.

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246 Fig. 4 shows the results of the FT-IR measurements of raw MK and NP, which indicate that the  
 247 highest absorption band of these raw materials are  $1046$  and  $996\text{cm}^{-1}$ , respectively These values  
 248 are attributed to the strong bands of Si–O–Al and tetrahedral Si–O–Si of bridging oxygen (BO)  
 249 atoms of the original alumina-silicate framework [34]. The characteristic peaks appearing at  
 250  $794\text{cm}^{-1}$  and  $724\text{cm}^{-1}$ , are ascribed to the stretching vibration Si–O and stretching Al 6-coordinated  
 251 geometry (AL, Mg)–O–OH [35]. Both absorptions Si–O–Si and Si–O, are supporting the presence  
 252 of quartz, while Si–O–Al is supporting the presence of kaolinite [35,36]. These peaks are related  
 253 to the alumina-silicate prevalence phases in both MK and NP. The strong presence of calcite  
 254 ( $\text{CaCO}_3$ ), presented by the C–O bond, was evidently presented by the bands at  $1402$ ,  $872$  and  
 255  $712\text{cm}^{-1}$  [37]. The presence of these bands is accredited to the tendency of CaO to react naturally  
 256 with  $\text{CO}_2$  in the atmosphere.

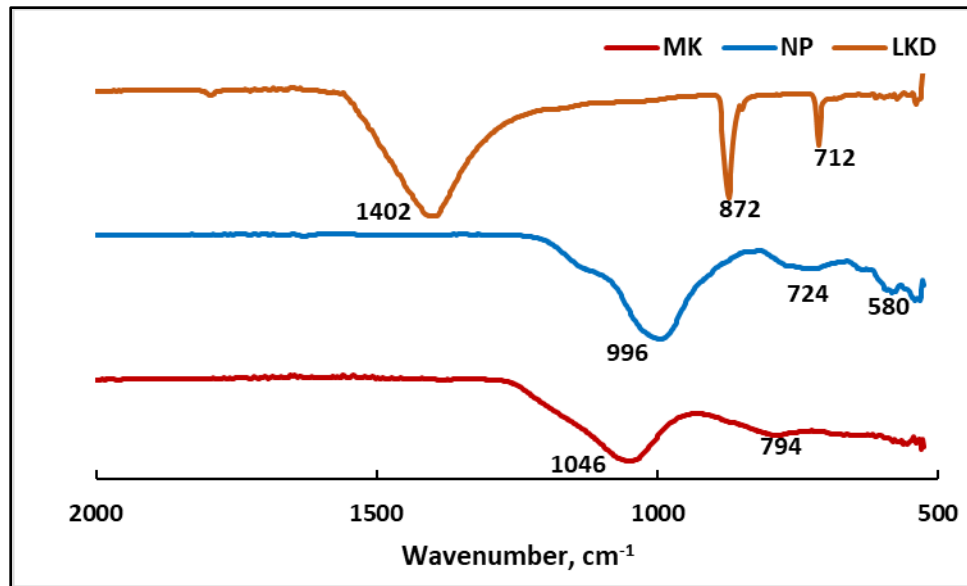


Figure 4. FTIR-spectra of *as received* materials

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## 259 2.4 One-part AAC cement preparation

### 260 2.4.1 Heat treatment mechanism

261 Thermal treatment or calcination, means heating the substance to high temperature (500-2000 °C)  
 262 within a controlled atmosphere, in order to increase their reactivity by changing their mineralogy  
 263 [23]. Thermal treatment is used as one of the assisting activation methods to produce one-part  
 264 AAC cement. Most of the materials, which are clayey in origin, require calcination in order to be  
 265 reactive. For instance, metakaolin ( $\text{Al}_2\text{Si}_2\text{O}_7$ ) is originally synthesised from the calcination of  
 266 kaolin clay ( $\text{Al}_2\text{SiO}_5(\text{OH})_4$ ) [38] at a temperature of 750 °C that used in current study. During the  
 267 calcination process, dehydroxylation is carried out, leading to the loss of the long-range order of  
 268 alumina and silica layers and conversion of the powder to an amorphous form so that when they  
 269 react with calcium hydroxide and water, they undergo a pozzolanic reaction to form hydration  
 270 products responsible of improving the strength and durability [39]. Another change is that the  
 271 alumina transforms from the octahedral coordination to tetrahedral coordination, due to calcination  
 272 [40]. On the other hand, recent studies demonstrated that the transformation of metakaolin into  
 273 spinal structure of Si-containing  $\gamma\text{-Al}_2\text{O}_3$  and amorphous silica does not occur in temperatures less  
 274 than 920 °C [41,42]. Therefore, in order to enhance the reactivity of calcined kaolins so that it can  
 275 be used as a pozzolanic cement products, a proper thermal treatment for metakaolin (for structure  
 276 disorder) is essential [41,43].

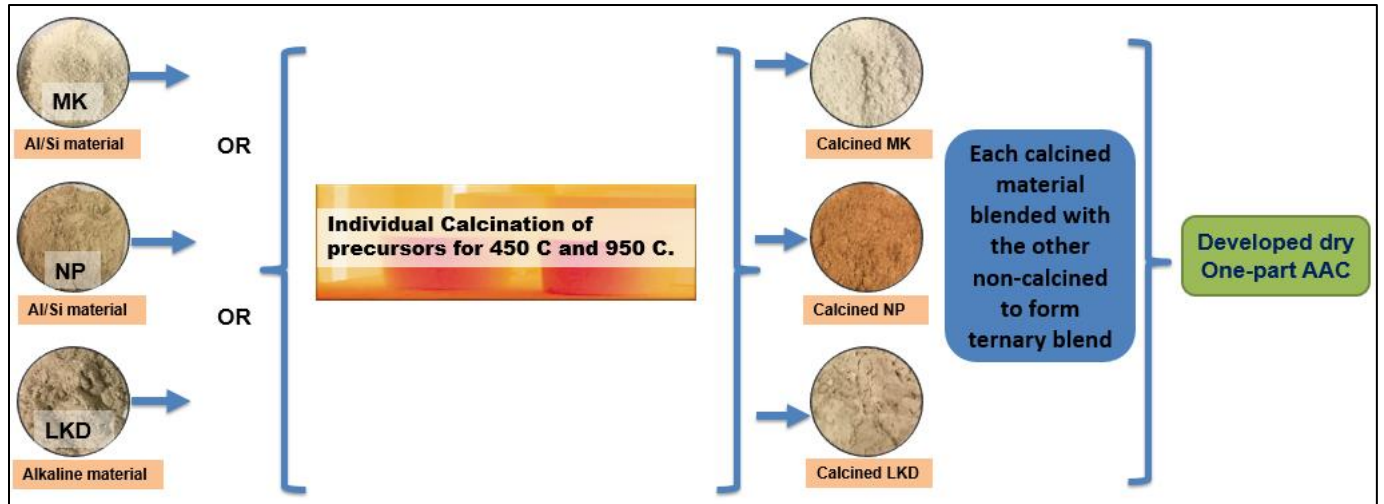
277 On another aspect, significant amounts of lime kiln dust (LKD) are produced, during the  
 278 calcination process of limestone, and collected from dust control systems. LKD usually contains  
 279 variable amounts of free/available CaO/and un-calcined calcium carbonates, and less amounts of  
 280 impurities, such as MgO, derived from the limestone [44] and dumped in waste landfills due to its  
 281 high amounts of impurities which is environmentally not favourable alternative. Earlier

282 investigation by Strydom et al.[45] to determine the reactivity and phase changes of precipitator  
283 dust from lime kilns upon heating to 500 °C and 1000 °C, has observed slight reduction in the  
284 intensity of the recognised calcite at temperature 500 °C and complete disappearance of calcite  
285 and transformation to CaO at 1000 °C, which makes LKD as hydraulic materials to participate in  
286 hydration reaction. Furthermore, NP, which is a natural pozzolan material, is considered as a good  
287 source of silicate and aluminate, but in a poor reactivity state it must be treated by various methods,  
288 such as chemically or thermally, to enhance its reactivity which was repeatedly confirmed  
289 previously by many literature [46–48].

290 Thermal activation was performed, in this study, for the candidate materials individually to  
291 increase their reactivity. Subsequently, mixing each calcined material with the other two non-  
292 calcined precursors, according to the blending concept (Si/Al+alkaline), was carried out to  
293 synthesise a dry hydraulic cement, as illustrated in Fig. 5. Thermal treatment was conducted in a  
294 muffle furnace with a ramping temperature of 20 °C/min for 2 hours. Two foundry cylindrical  
295 silicon carbide graphite crucibles (500ml) and two alumina cylindrical crucibles (175ml), were  
296 used during calcination process. The relevant literature demonstrated that crystallinity and  
297 reactivity of most materials rich in alumina-silicate content can increase with temperatures up to  
298 950-1000°C, but after this temperature range, their reactivity can be decreased [49,50]. In this  
299 study, activation at temperatures of 450°C and 950°C have been investigated to evaluate and  
300 compare the phase transitions. Activation at 450°C temperature at mid-range between 0 and 950°C  
301 to evaluate and compare the phase's transitions aimed for saving energy. It is noteworthy to  
302 mention that the choice of activation temperatures was based on TGA and XRD analysis  
303 (discussed later), which revealed that most phase transitions occur at these levels.

304 Additionally, it is noteworthy that during thermal activation process; some of the material colours  
305 have been changed in both stages of temperatures, as displayed in Fig 5. The colour of MK was  
306 becoming whiter, with the increase of temperature, while, the colour of NP has shown considerable  
307 change from greyish to brown, as the temperature rose. The colour transformation of NP attributed  
308 to the increment of anorthite mineral that has been observed clearly in XRD patterns, which turns  
309 to be brown in nature [51]. LKD originally white but turned into light grey at 950°C.

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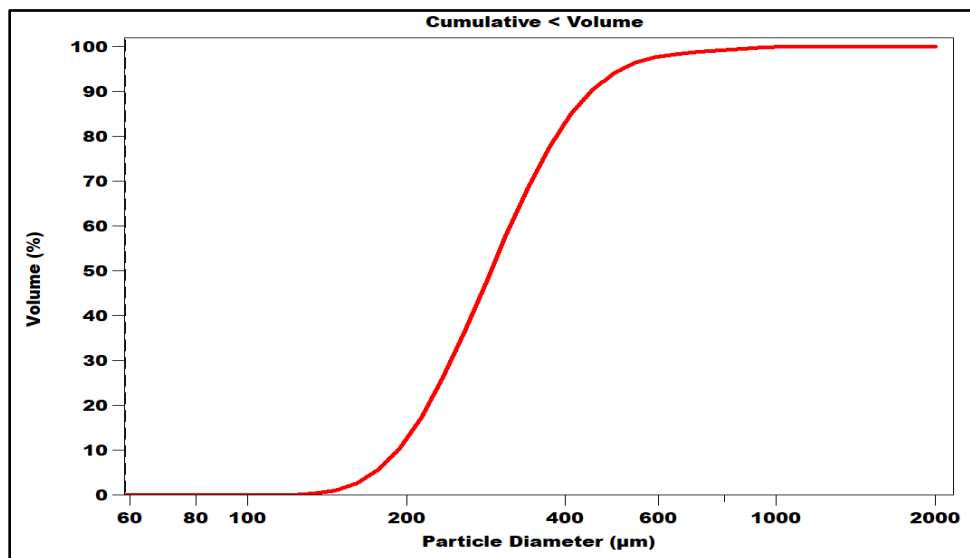
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Figure 5. Preparation steps of one-part alkali activated cement.

313 2.4.2 Preparation of one-part AAC mortars

314 Mortars were mixed with kiln dried sand, with a specific gravity of 2.62, with sand to binder ratio  
 315 of 1:2. The particle size distribution of the used sand is shown in Fig. 6. The mixing process was  
 316 conducted according to the requirements of British standard 196-1 [32]. Initially, 0.55 water/binder  
 317 ratio was chosen, but it was found that the mortars were extremely flowable, therefore; it was  
 318 reduced to 0.45 for all the mortars, where appropriate consistency was achieved at this stage.



319

320

Figure 6 Particle Size Distribution of the sand used for mortar.

321 Mortars were mixed, using a Hobart 5L countertop Mixer N50, and poured in steel prism moulds  
 322 (40mm x 40mm x 160mm). Mortars were prepared using ternary blended binder with proportions,  
 323 as shown in Table 2. These mix proportions, in the ternary blends, were achieved after several  
 324 initial trials, as the ternary blended with only non-calcined constituent, showed no cementing  
 325 property after 28 days of curing.

Table 2 Mixing proportions of ternary blends used for preparing mortar specimen.

Mix ID	Binder contents	Calcination temperature (°C)	Calcined constituent
AAC1	40%MK 40%LKD 20%NP	950	NP
AAC2	40%MK 40%LKD 20%NP	950	MK
AAC3	40%MK 40%LKD 20%NP	950	LKD
AAC4	40%MK 40%LKD 20%NP	950	NP, MK, LKD
AAC5	35%MK 35%LKD 30%NP	950	NP
AAC6	35%MK 35%LKD 30%NP	950	MK
AAC7	35%MK 35%LKD 30%NP	950	LKD
AAC8	35%MK 35%LKD 30%NP	950	NP, MK, LKD
AAC9	35%MK 35%LKD 30%NP	450	NP
AAC10	35%MK 35%LKD 30%NP	450	MK
AAC11	35%MK 35%LKD 30%NP	450	LKD
AAC12	35%MK 35%LKD 30%NP	450	NP, MK, LKD

327

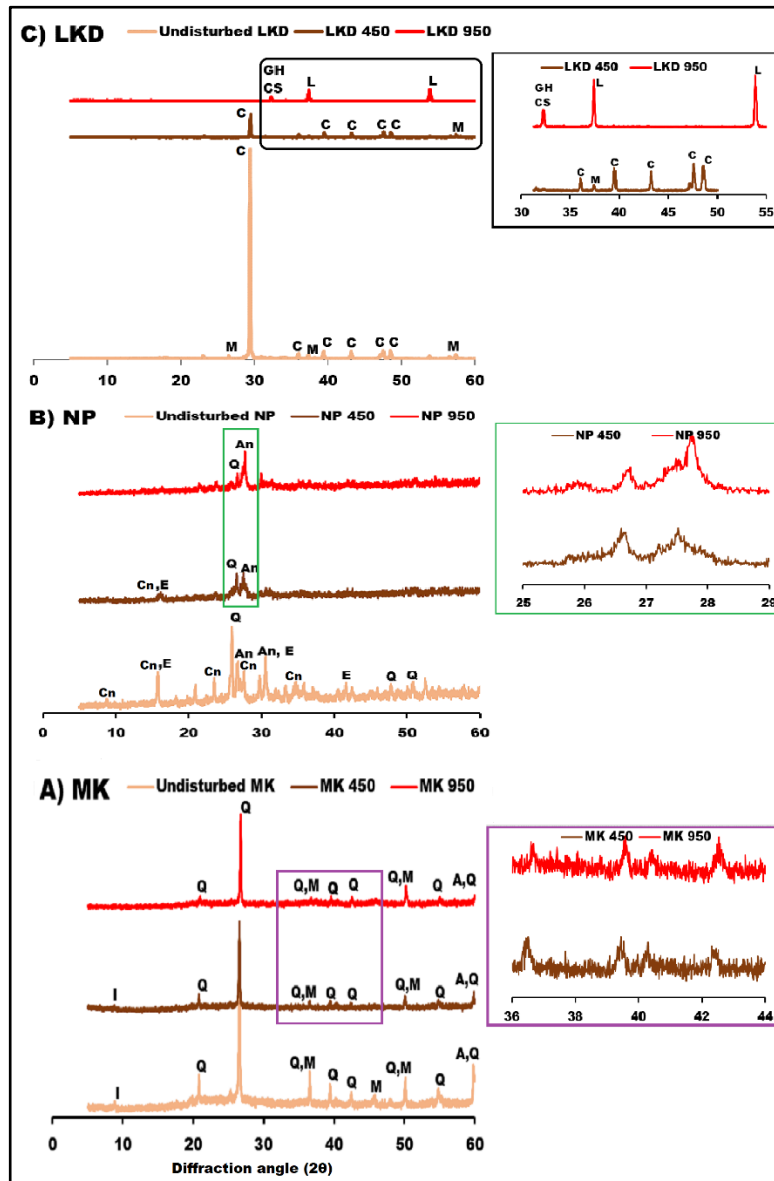
328 The mortars were cured in a hot water curing regime, to enhance the reactivity and the alkali-  
 329 activation rate of the mortars and to avoid potential problems, such as efflorescence and micro  
 330 cracking, which can lead to a reduction in the compressive strength [52]. Curing temperature was  
 331 fixed to 50 °C for the first 7 days, and then cured in normal water (20 °C) for the desired period,  
 332 as suggested by Perera et al. [53] and Singh et al. [54].

### 333 3. Results and discussion

#### 334 3.1 XRD analysis after heat treatment

335 The powder diffraction of calcined materials has been presented in Fig. 7. Diffractograms of MK  
 336 revealed a significant reduction and disappearance of peaks, such as mullite and illite, after both  
 337 levels of calcination indicating semi-transformation of material to an amorphous phase. While  
 338 peak of quartz has shown no change on both calcination level, XRD spectrums of calcined NP at  
 339 both levels, indicated a huge transformation of crystalline phases to amorphous phases through the  
 340 loss of quartz and clinoptilolite, in a range of  $2\theta$  from  $30^\circ$  to  $60^\circ$ . It can be noticed, that crystalline  
 341 quartz, which peaked at  $2\theta = 26^\circ$ , has disappeared entirely when NP calcined to  $450^\circ\text{C}$  and  $950^\circ\text{C}$ ,  
 342 introducing strong evidence of the increment of amorphicity. This was confirmed by the  
 343 formulation of a broad amorphous hump of anorthite existed at  $2\theta = 26.7^\circ$  and  $2\theta = 27.8^\circ$  at  $950^\circ\text{C}$   
 344 patterns. Remarkably, this intense calcite peak in LKD was reduced markedly when calcined to  
 345  $450^\circ\text{C}$  and **completely disappeared in calcination at  $950^\circ\text{C}$**  which could indicate the total  
 346 decomposition of calcite  $\text{CaCO}_3$  to lime  $\text{CaO}$  and introduced as new peaks at  $2\theta = 37^\circ$  and  $54^\circ$ .

347 The appearance of new diffraction patterns of calcined LKD at 950°C, indicated the formulation  
 348 of new compounds, but it was composed of the same elements as untreated LKD (CaO, SiO<sub>2</sub> and  
 349 Al<sub>2</sub>O<sub>3</sub>), in the form of periclase cementitious minerals, di-calcium silicate C<sub>2</sub>S (2CaO.SiO<sub>2</sub>) and  
 350 gehlenite (Ca<sub>2</sub>Al(Al Si)O<sub>7</sub>) [45]. This would suggest that mullite (3Al<sub>2</sub>O<sub>3</sub>.2SiO<sub>2</sub>) exist in raw LKD  
 351 has decomposed and reacted with CaO to formulate these new phases [45]. The complete  
 352 disappearance of the intense crystalline CaO after 950°C calcination, which indicated that  
 353 calcination had caused a large combination of lime CaO with the compounds of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>,  
 354 to form the above hydraulic minerals.

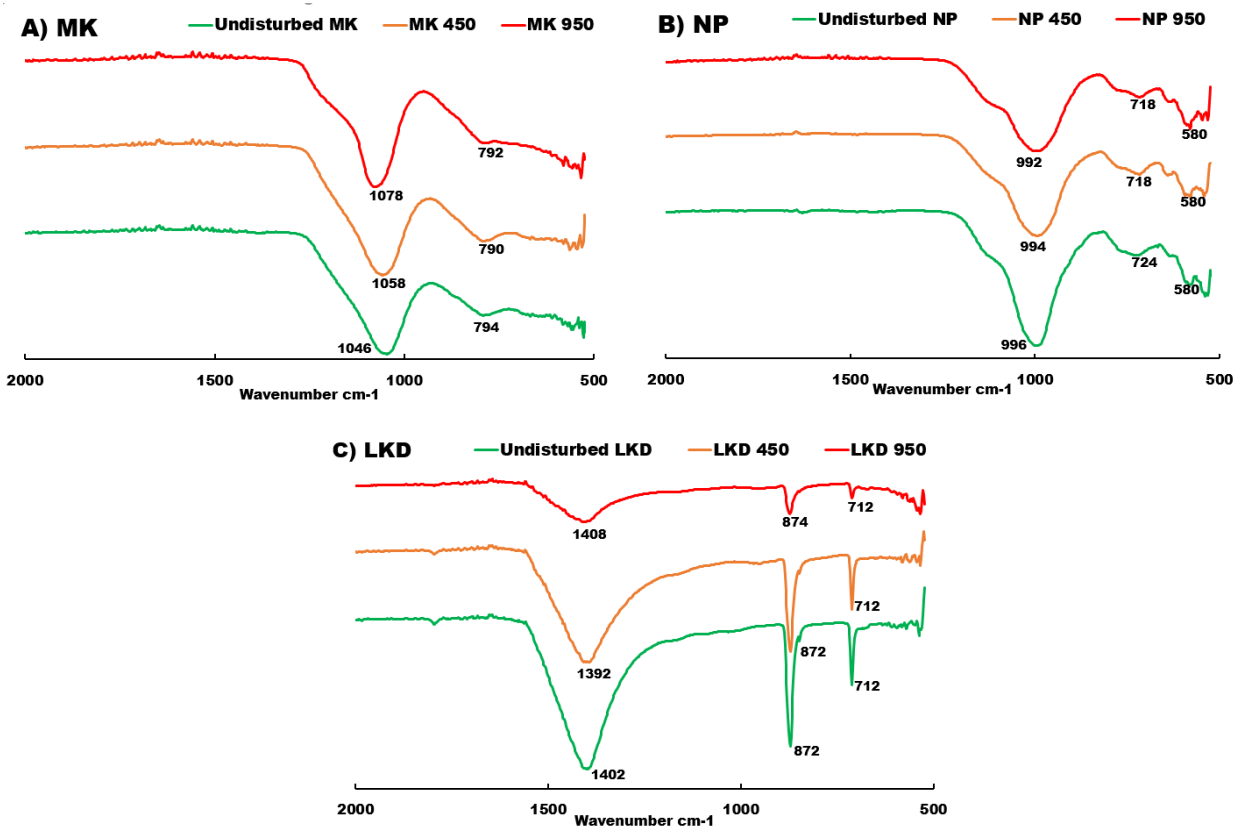


355  
 356 *Figure 7. Powder XRD patterns of materials after thermal activation, A) MK, B) NP, and C) LKD.*

357 *Q: Quartz, M: Mullite, A: Anatas, I: Illite, Cn: Clinoptilolite, An: Anorthite, E: Edenite, L: Lime, C:*  
 358 *Calcite, CS: dicalcium-silicate, GH: gehlenite*

359 3.2 FT-IR analysis after heat treatment

360 The FT-IR spectrums of thermally treated materials are illustrated in Fig. 8. The alumina-silicate  
361 bands Si-O-Si and Si-O-Al have become higher from 1046  $\text{cm}^{-1}$ , in untreated MK, to 1058  $\text{cm}^{-1}$  at  
362 450 °C and 1078  $\text{cm}^{-1}$  at 950 °C. This is while the opposite to this tendency took place with the  
363 spectra of NP. This kind of shifting and enhancement of broadness of these bands, leads to the  
364 transformation of crystalline phases to an amorphous structure and high deformation in the lengths  
365 and angles of Si-O-Al and Si-O-Si bonds [25]. This transformation is satisfied by the increment  
366 of non-bridging oxygen atoms due to thermal treatment [55]. A considerable reduction and width  
367 increment of the intensity of C-O bond during the treatment at 950°C, indicates the transformation  
368 of the crystalline phase into the glassy phase and interprets the increment of CaO amounts released  
369 from  $\text{CaCO}_3$  [56,57].



370

371

Figure 8. FTIR spectra of materials after thermal activation.

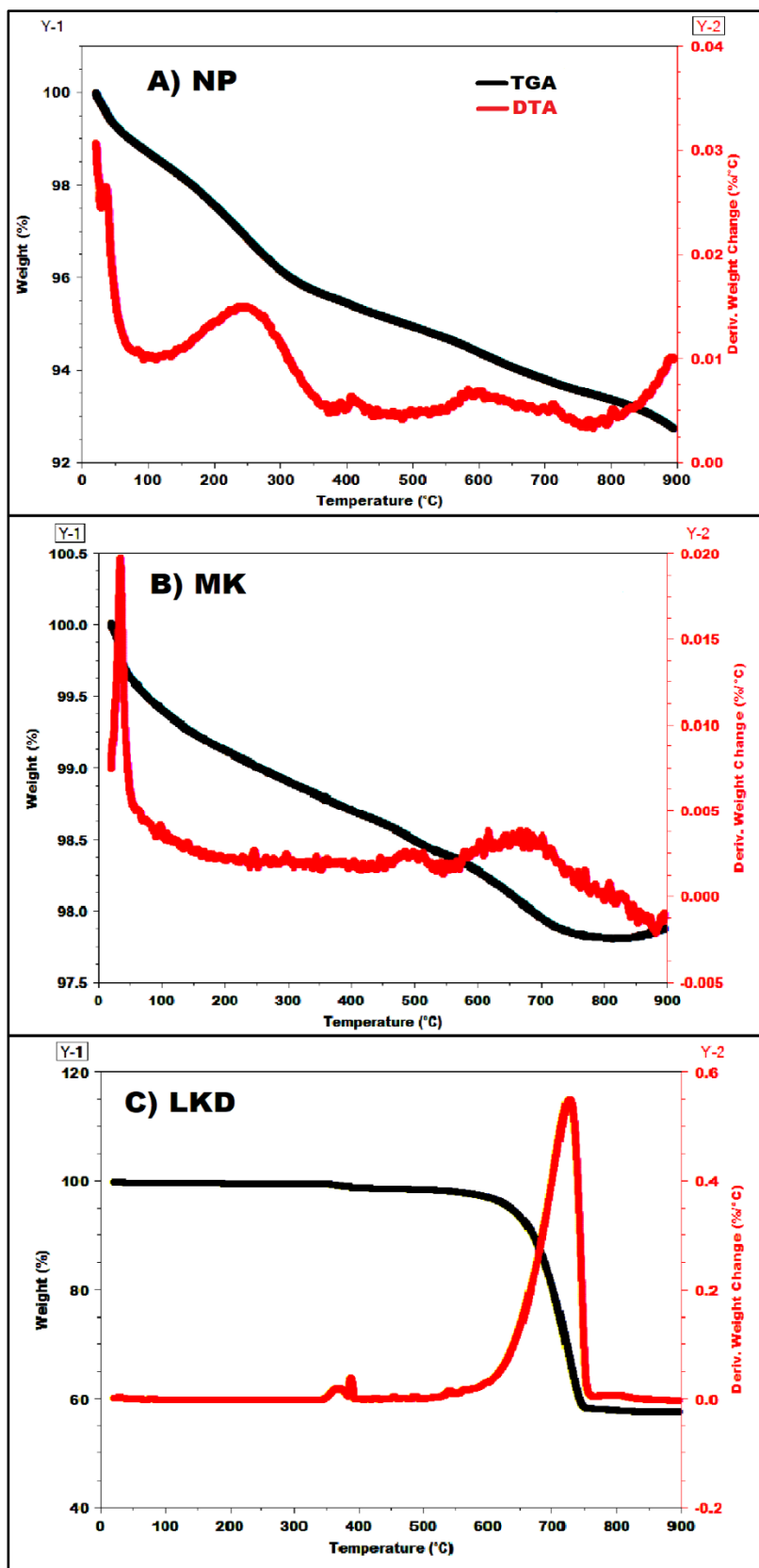
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373 3.3 Thermogravimetric Analysis (TG-DTA) after heat treatment

374 Fig. 9 (A) exhibits the thermal performance of NP investigated by TG-DTA, when calcined to  
375 900°C. As can be seen, some mass-loss phases are present in its patterns caused by endothermic  
376 and exothermic reactions. The Thermogravimetric (TG) patterns of NP mass-loss from room  
377 temperature to 900°C is approximately 7% of total weight, which might be attributed to the loss of



378 chemically and physically adsorbed water and breakdown of crystal phases, which was evidenced  
379 by XRD analysis. The loss of weight can be divided into three stages. The loss in temperature  
380 range of 20-320°C is due to the evaporation of adsorbed water. The loss in the range of 320 –  
381 620°C, is because of calcining amounts of impurities and contaminants. While loss in the range  
382 620 - 900°C is due to decomposition of unburnt carbons [58,59]. As for the DTA curve, there was  
383 endothermic convexity at 119°C, which relates to the evaporation of water followed by exothermic  
384 concavity at 250°C. The TG curve of MK has shown a very small weight change within 2% of the  
385 total weight, with a reasonably straight DTA curve, as shown in Fig. 9B. The TG patterns of LKD,  
386 revealed that it has not lost any weight until 350°C; when there was only a slight weight loss until  
387 620°C, when the weight changed dramatically with a 35 % loss of original weight and an  
388 exothermic sharp peak of the DTA curve, which is shown clearly in Fig. 9C. This loss is markedly  
389 attributed to the transformation and loss of the CaO crystalline intense peak, which was further  
390 evidenced by the XRD analysis of 950°C calcination, when this peak disappeared from the  
391 diffraction pattern.



392

393

Figure 9. TG-DTA analysis of initial materials, A) NP, B) MK, and C) LKD

### 394 3.4 Influence on compressive strength after heat treatment and blending.

395 One-part mortar samples were prepared and cured using the mix proportion, with two levels of  
396 thermally activated materials as stated in table 2, and their resulting compressive strengths are  
397 displayed in Figs. 10 and 11. The blend containing 950°C calcined LKD (AAC3), has shown a  
398 compressive strength of 24.54 MPa after 28 days. The regular growth in strength of up-to 28 days  
399 for this blend, indicates the progressive alkali-activation that attributes to the calcination of LKD  
400 with the presence of sufficient amounts of reactive silicate and calcium oxide from the NP. A  
401 considerable amount of strength was developed within the blends, containing calcined materials  
402 individually and collectively, which showed no cementitious properties prior to thermal treatment.  
403 Calcination at 950°C, clearly contributed to a huge variation in LKD mineralogy and substantial  
404 reduction in its crystallinity, which was observed in the XRD patterns. In order to optimise the  
405 strength, these mortar mixes were repeated with same procedures, but with increased proportion  
406 of NP (35% MK, 35% LKD, and 30% NP). The increment of the NP was due to the suitable  
407 composition of NP, which contains a various range of alkaline elements including CaO, Na<sub>2</sub>O and  
408 K<sub>2</sub>O, as shown in XRF results. Noticeably, the highest compressive strength was achieved at this  
409 stage with 27.3 MPa in 28 days for the blend that contains 950°C calcined LKD (AAC7), as shown  
410 in Fig. 10. The increment of NP content by 10% within the blends, resulted more growth of C-A-  
411 S-H products. The addition of more NP contributed to extra dissolution of quartz silicate, caused  
412 by CaO and Na<sub>2</sub>O that exists in the calcined LKD and NP, yielding high binding properties.  
413 Therefore, a higher transformation to vitreous mineralogy led to a higher strength of AAC7.  
414 Moreover, the reduced strength for AAC4 and AAC8, when all three components were calcined,  
415 indicated less dissolution of Si/Al compounds from MK and NP (at both composition), which  
416 means that less activation was caused by the LKD.

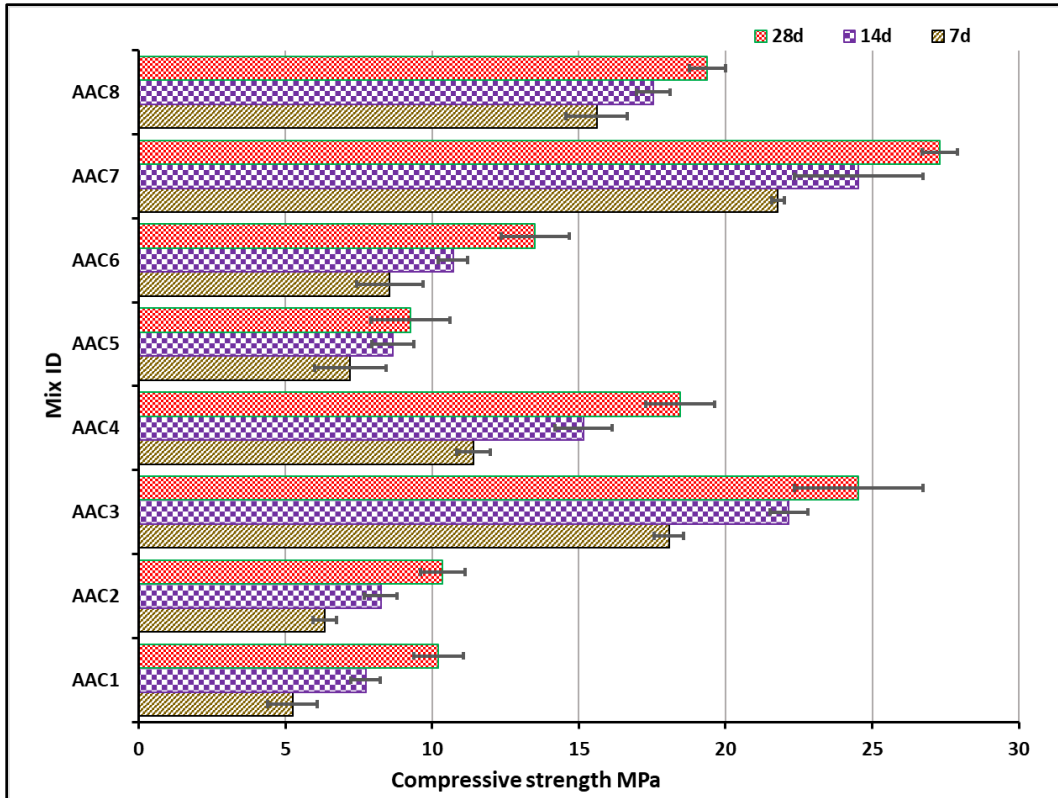


Figure 10. Compressive strength of blends with 950 °C calcined materials.

417

418

419 The TG-DTA curves have shown that the weight loss of the individual constituent of the ternary  
 420 blend, starts at the temperatures in the range 350-500°C. This was evidenced by XRD patterns,  
 421 where raw materials after 450 °C treatment have displayed a considerable change in their  
 422 diffraction. Therefore, the ternary blends of 35% MK, 35% LKD and 30% NP were formulated  
 423 after calcination of 450°C. The resulting compressive strength offered by the ternary blends after  
 424 450°C thermal activation, has been shown in Fig. 11. A similar trend of higher strength in the case  
 425 of calcining LKD individually at 450 °C, was evident in AAC11 with 23.4 MPa at 28 days.  
 426 Although both early and longer-term strengths were higher in the case of 950°C treatment, no  
 427 remarkable strength generation was observed compared to a lower temperature (450°C). This  
 428 similar development of strength is ascribed to mineralogical and chemical changes that were  
 429 noticed in both XRD and TG-DTA results, which indicated that most of diffraction patterns were  
 430 starting to transform around and after 450°C.

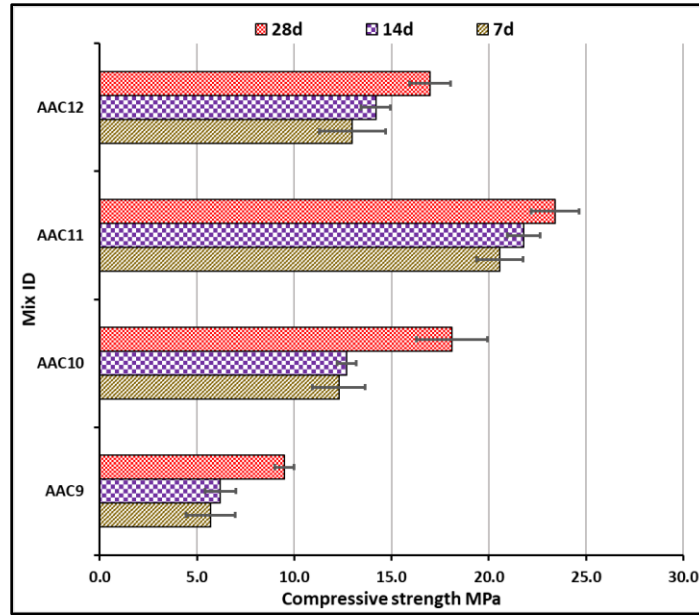


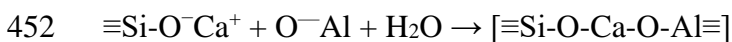
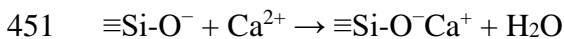
Figure 11. Compressive strength of blends with 450 °C calcined materials.

431

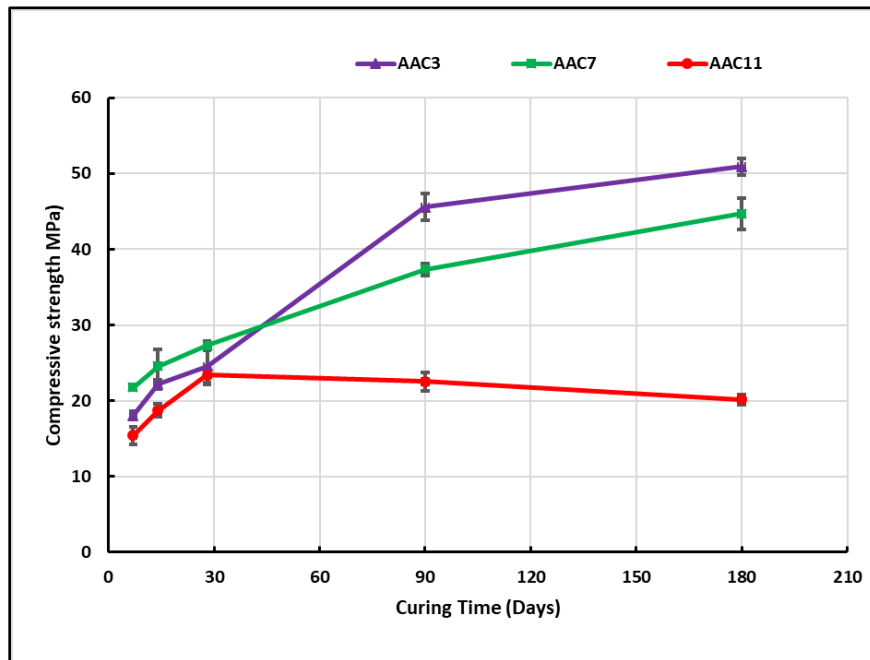
432

### 433 3.4.1 Long-term compressive strength for optimal blends

434 The compressive strength after 90 and 180 days of curing was investigated for the blends, which  
 435 showed a higher alkali-activation rate in the case of calcined LKD at both level of calcination and  
 436 mix proportion (AAC3, AAC7 and AAC11), as presented in Fig. 12. Although AAC3 has a lower  
 437 compressive strength than AAC7 until 28 days of curing, it showed significant increase in rate of  
 438 strength development just after 28 days and continued till 180 days. The higher rate of strength  
 439 growth has been witnessed at the later age of 180 days which has shown increment of strength to  
 440 reach 51 MPa for blend AAC3 and 45 MPa for AAC7 with increment of about 11% and 18% at  
 441 age of 90 days of both mixes respectively; indicating that there was a high degree of alkali-  
 442 activation within the system until 180 days, without showing any marks of efflorescence. The  
 443 higher degree of polymerisation within AAC3 compared to AAC7 was due to the formulation of  
 444 balanced alumina silicate and alkaline materials, which was achieved through reducing 10% of NP  
 445 and adjusting the others accordingly. This is was due to the higher CaO contained in AAC3 that  
 446 promoted formulation of further amounts of geopolymeric products and more stable  
 447 microstructure. This can be justified that calcium cations ( $\text{Ca}^{+2}$ ) might be bonded in the  
 448 geopolymeric gel that results in charge balance by replacing cations within the geopolymer [60].  
 449 The chemical mechanism of combining calcium cations in geopolymeric gel is illustrated by the  
 450 following reactions [61,62]:



454 Contrariwise, sample containing calcined LKD at 450°C (AAC11), has promoted a slight  
 455 deterioration in strength immediately after 28 days and continued until 180 days. This signifies the  
 456 inhibition of the alkali-activation reaction after 28 days in this blend. While the combined  
 457 procedures of impurities disappearance that were contained in the chemical composition of LKD  
 458 as a by-product material and the mineralogical changes after thermal treatment have strongly  
 459 introduced the fact that reactivity of LKD has increased just after thermal treatment. To confirm  
 460 this fact, this was investigated by preparing a mortar sample similar to AAC7 but without the  
 461 application of thermal treatment to LKD, as this sample has the optimal strength at the age of 28  
 462 days, 51% lower compressive strength (18 MPa) after 28 days was achieved from that blend, that  
 463 comprised un-calcined LKD, and decreased to 16 MPa at age of 180 days.



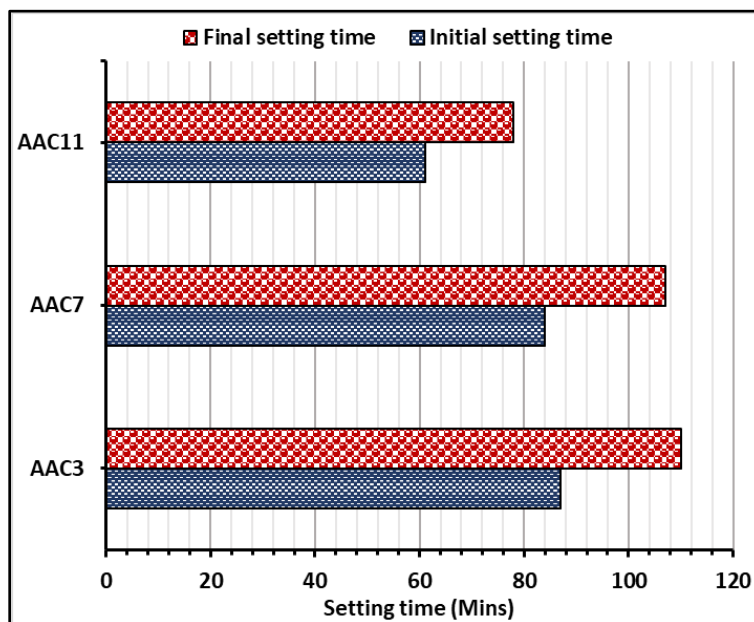
464  
 465 *Figure 12 : Long-term strengths of one-part blends containing activated LKD at 450 °C and 950 °C.*

466 **3.5 Setting time**

467 Fig. 13 shows the initial IS and final FS setting time of the optimum blends AAC3, AAC7 and  
 468 AAC11. It can be observed that the setting times for blends are fluctuating between 61-87 min for  
 469 the initial setting time and between 78-110 minutes for final setting time. It can also be noticed in  
 470 Fig.13 that nearly parallel IS and FS for blends AAC3 and AAC7, indicating that slower alkali  
 471 activation is due to the higher content of calcined LKD (40 Wt.% and 35 Wt.%), which acts here  
 472 as a key activation agent. Higher amounts of alkaline cations of Ca<sup>+2</sup> from LKD in the system is  
 473 leading to the formation of extra Ca(OH)<sub>2</sub> that causes more breaking and dissolving of Si and Al  
 474 chains that accelerates the alkali-activation reaction, and consequently increases the setting time  
 475 [63]. However, lower IS and FS were observed in AAC11, as shorter levels of reactions occurred  
 476 due to the low reactive LKD and, therefore, lower alkali activation. The effect of the alkaline  
 477 cations, from the activator (LKD), was clearly seen on the setting time. The result was evident, as

478 the alkaline part became more reactive, as there was more dissolution, leading to longer setting  
479 time.

480 In terms of application and use of AAC, a higher setting time is a significant parameter for  
481 transporting far distances, storage and a shorter setting time, which is important for quick repairs  
482 of damaged surfaces [63]. The current results of IS and FS are in a good agreement with the results  
483 of past studies. For example, Luukkonen et al. [8] revealed in their review that IS and FS of  
484 developed one-part AAC in past attempts were from 23-150 and 69-230 minutes, respectively.  
485 Additionally, the obtained results of initial setting time in this study are in agreement with initial  
486 setting times for cement types in British standards 197-1 [64]. Furthermore, setting times can be  
487 easily increased if needed in some applications with the aid of setting retardant agents such as  
488 lignosulfonate which was proven as a suitable retarder for one-part AAC in past studies [65].



489

490

*Figure 13 Initial and Final setting times of optimum one-part AAC blends.*

### 491 3.6 XRD analysis of hydration products

492 XRD analysis was performed on pastes of samples with the highest strength (AAC3, AAC7 and  
493 AAC11) at 28 days and for longer ages (at 90 and 180 days), as presented in Fig. 14. Major  
494 hydration products can be identified and are composed chiefly from CaO-Al<sub>2</sub>O<sub>3</sub>-MgO-SiO<sub>2</sub>  
495 compounds. The utilisation of a dry alkaline (CaO) activator, in the mix, transforms the  
496 crystallinity diffraction patterns into amorphicity status and leads to the presence of prevailing  
497 vitreous phases. The major products phases were specifically tetra calcium aluminate hydrate,  
498 CAH (C<sub>4</sub>AH<sub>13</sub>), calcium alumina silicate hydrates, St (stratlingite – C<sub>2</sub>ASH<sub>8</sub>), and Akermanite,  
499 (Ak) (Ca<sub>2</sub>MgSi<sub>2</sub>O<sub>7</sub>) [17][66][67][56]. Moreover, Gameiro [67] stated that calcium silicate hydrate  
500 (CSH) overlaps with the lime (calcite) peak in these blends, with higher MK (≥ 33% MK). XRD  
501 patterns of AAC3 as presented in Fig. 14a. This figure reveals that at the age of 28 days, a slight  
502 reduction of the dominant Quartz (SiO<sub>2</sub>) crystalline phase that existed in the raw MK at 2θ = 26°.

503 This slight reduction indicates the remaining large amounts of this crystalline quartz are in a non-  
504 reactant status. On the other hand, this reduction was sharp at age of 90 days and even more at age  
505 of 180 days, presenting more dissolution of crystalline SiO<sub>2</sub> and transforming into alkaline-  
506 silicates strains. This dissolution was induced by the formation of extra Ak products at the longer  
507 ages. The fact of reducing crystalline quartz phases and the formation of vitreous Ak, led to the  
508 development of significant strength after 90 and 180 days of (AAC3)[63]. Additionally, a glassy  
509 phase of alumina-silicate gel (C-A-S-H) can be indicated in the 2θ range of 25°-30° at age of 28  
510 days, however, due to the semi- amorphous nature, this product disappeared at age of 180 days  
511 [69,70]. At the same time, the strong presence of St, is noticed as a permanent phase in the matrix.  
512 The formulation of St as dominant substance in the three blends, indicating that this product is  
513 stable and is verified as the product responsible for the enhancement of mechanical strength  
514 [67,71]. Regardless of the slight increment of the Ak mineral peak at 90 and 180 days of AAC7  
515 (in Fig. 14b), crystalline quartz can be noticed in an increment trend from 28 to 180 days. The  
516 growth of St peaks can be noticed clearly, through the entire range of diffraction with increment  
517 of LKD, causing a higher strength for AAC7. This indicates that the lime (CaO) generated from  
518 LKD is causing a significant dissolution of aluminate and silicate species [72], and consumes all  
519 alkaline substances resulting no mark of any efflorescence. The high amounts of MgO from NP  
520 have strongly contributed to presence of amorphous phases, such as Ak together with the reactive  
521 CaO from LKD. This reaction introduced new bonds such as Ca-Mg-Si, which have a high  
522 contribution to the strength. C<sub>4</sub>AH<sub>13</sub> (CAH) was present in minor quantities due to the instability  
523 of this phase through the curing aging [67]. Additionally, portlandite (Ca(OH)<sub>2</sub>) peaks were not  
524 found extensively due to the modified LKD and MK in the blends, as this peak may appear in low  
525 MK blends [67]. A range of reaction products can be noticed similarly in AAC11 as in Fig. 14c,  
526 with LKD calcined at 450°C. The amounts of Ak can be observed with less quantities in the all  
527 ages of AAC11 with higher amounts calcite (C) than AAC3 and AAC7 that formulated through  
528 the ages. This was observed in the strength development for both levels of thermal treatment.  
529 However, the diffraction patterns of this blend are presenting growth of crystalline quartz in two  
530 major peaks at 2θ =26° and 47°. This caused a dip in the equivalent of 90- and 180-days strength,  
531 for AAC11.



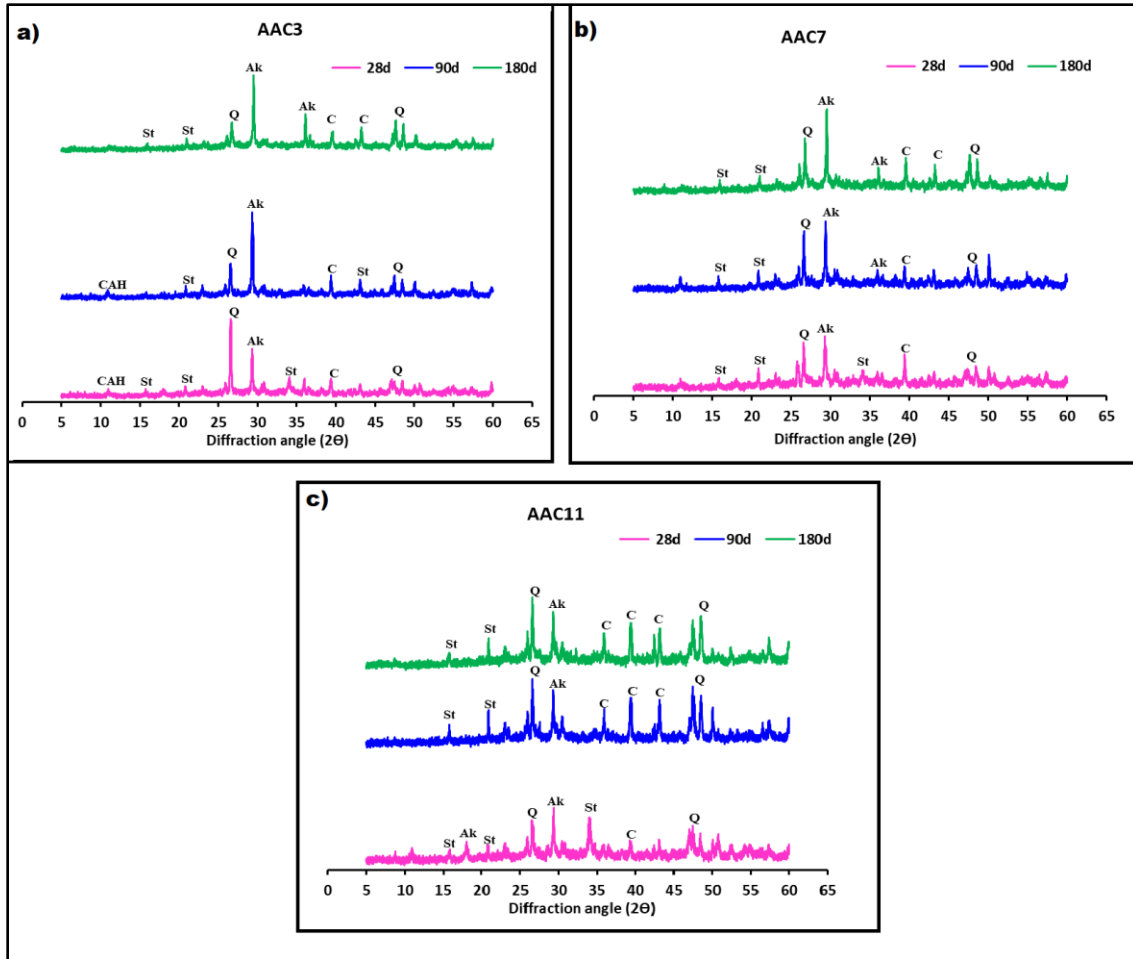


Figure 14. XRD patterns of a) AAC3, b) AAC7, c) AAC11 at 28, 90 and 180 days.

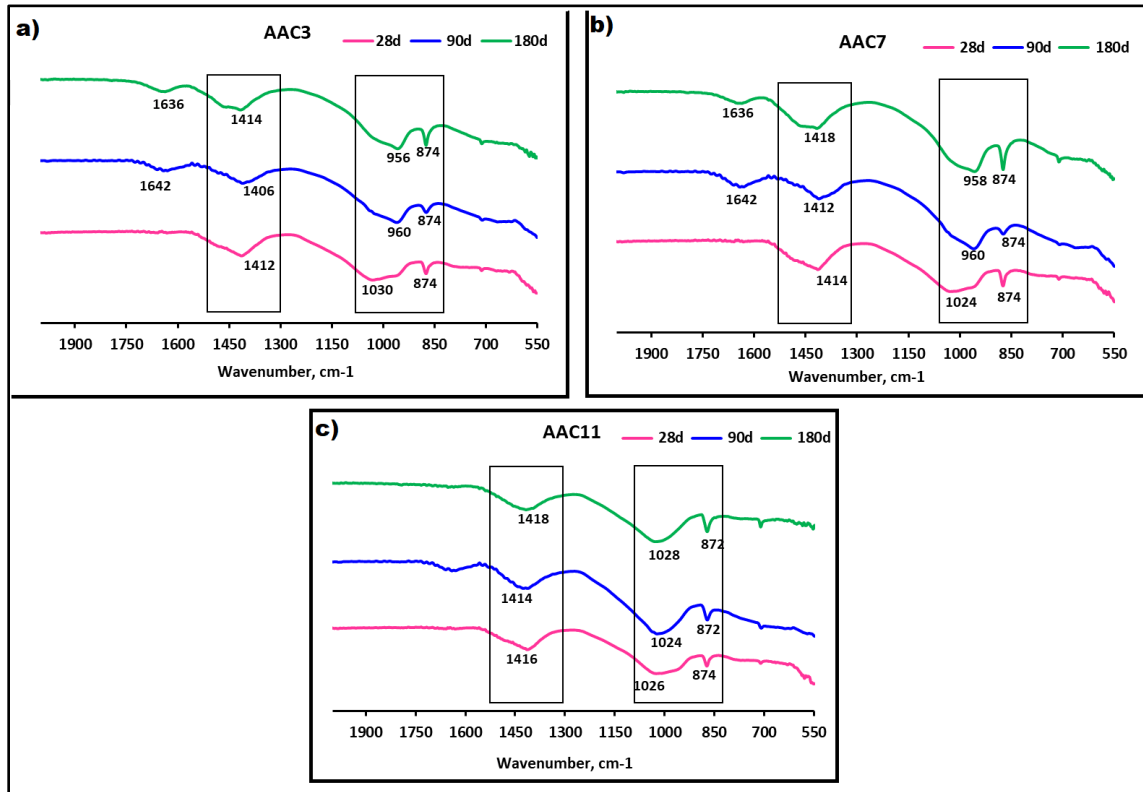
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533

### 534 3.7 FTIR analysis of hydration products

535 The FTIR (Fourier-transform infrared) spectra was reproduced for pastes of AAC3, AAC7 and  
 536 AAC11 blends, and were investigated after 28,90 and 180 days of curing, as shown in Fig. 15a  
 537 new absorption bands were recorded at 1642 and 1636  $\text{cm}^{-1}$ , for the vibration bending H–O–H in  
 538 all blends at 90 and 180 days. The appearance of water absorption bands are a clue for the  
 539 crystalline  $\text{H}_2\text{O}$  of the hydrated products such as C-S-H or C-A-H [73]. In all spectra, the typical  
 540 carbonate phases C-O band, appeared in the following bands 1406-1418 and  $874\text{cm}^{-1}$  was  
 541 observed. This is similar to infrared in the raw LKD in Fig. 4. However, the intensity of this band  
 542 was reduced with the right shifting of its location and has become wider at 90 and 180 days for the  
 543 blends, except for the AAC11 blend, as shown in Fig. 15a-c. This attributed to the calcined LKD  
 544 at  $950^\circ\text{C}$ , has purer and more reactive CaO than  $450^\circ\text{C}$  calcination in the case of AAC11, which  
 545 has more groups of  $\text{CaCO}_3$ . The asymmetric stretching band starting from  $1200\text{-}900\text{ cm}^{-1}$  region  
 546 of Si-O and Al-O were noticed in all spectra of the blends. These bands reveal the strong evidence  
 547 of Aft/AFm phases such as stratlingite ( $\text{C}_2\text{ASH}_8$ ) and mono-carboaluminate  $\text{Ca}_4\text{Al}_2(\text{CO}_3)(\text{OH})_{12}/$   
 548  $5\text{H}_2\text{O}$  [74]. It can be noticed that this band was very wide at 28 days and transformed to be sharp

549 and narrow at 90 and 180 days with shifting to lower wavenumber in both blends AAC3 and  
 550 AAC7. It was revealed that such this phenomenon is correlated to the breakdown of covalent bonds  
 551 of silicate networks, which become less strong and lead to the formation of more non-bridging  
 552 oxygen (Si–O bonds) that was simply undergone to more condensation and polymerisation [75].  
 553 Contrarily, these bands remained unchanged in blend AAC11 for ages of 28, 90 and 180 days.  
 554 This has led to the development of phases with a non-ordered structure, with a variation of bond  
 555 length and angles [76], which was in accordance with the reduced strength of AAC11.

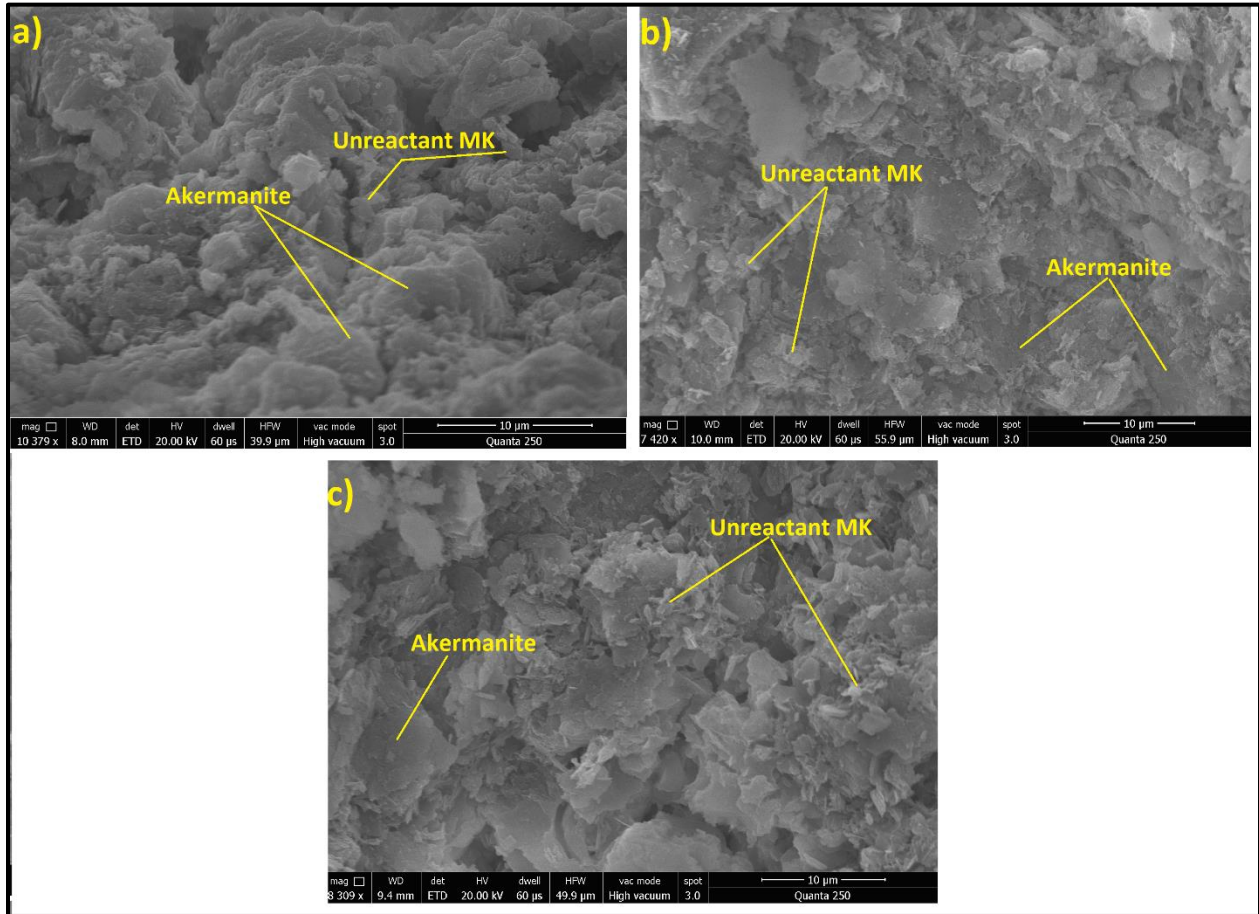


556  
 557 *Figure 15. FTIR-spectra of hardened pastes of a) AAC3, b) AAC7, c) AAC11 after 28, 90 and 180days*  
 558 *curing.*

### 559 3.8 SEM analysis of hydrates

560 The SEM micrographs for AAC3,7 and 11 at 28 days of curing are shown in Fig. 16. The  
 561 microstructure of hardened pastes shows the significant existence of the un-reactant MK in all  
 562 blends at 28 days, which can be identified by fine and lamellar particles with random non uniform  
 563 shapes and similar to particles of raw MK appeared previously in Fig. 3c. However, the un-reactant  
 564 MK has very limited quantities in blend AAC3, as shown in Fig. 16a, and highest quantities in  
 565 AAC11, as seen in Fig. 16c. This illustrates the pozzolanic reaction of Si and Al with Ca<sup>+2</sup> cations,  
 566 which encourages further dissolution and breaking the Si–O and Al–O bonds in both MK and NP  
 567 [77]. Ak products can be distinguished clearly as a predominant phase of flat continuous gels in  
 568 all blends, but its lowest appearance can be seen in AAC11, which agrees with the XRD patterns.  
 569 The trend of reaction mechanism during the hydration process is with the increment of reactive

570 calcined LKD at 950 °C, where there is an increment in the operation of extra dissolution of Al/Si  
571 constituents.

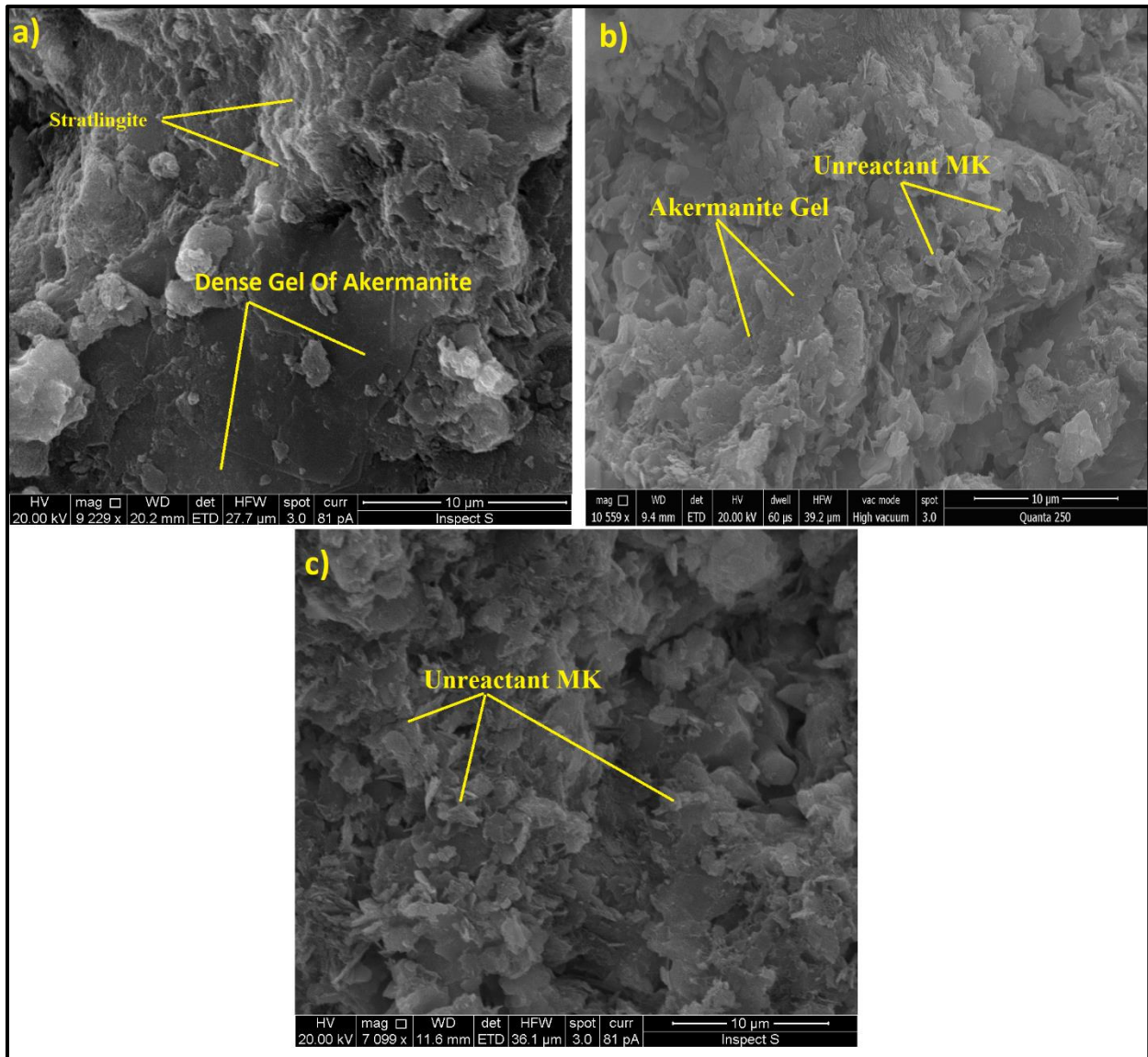


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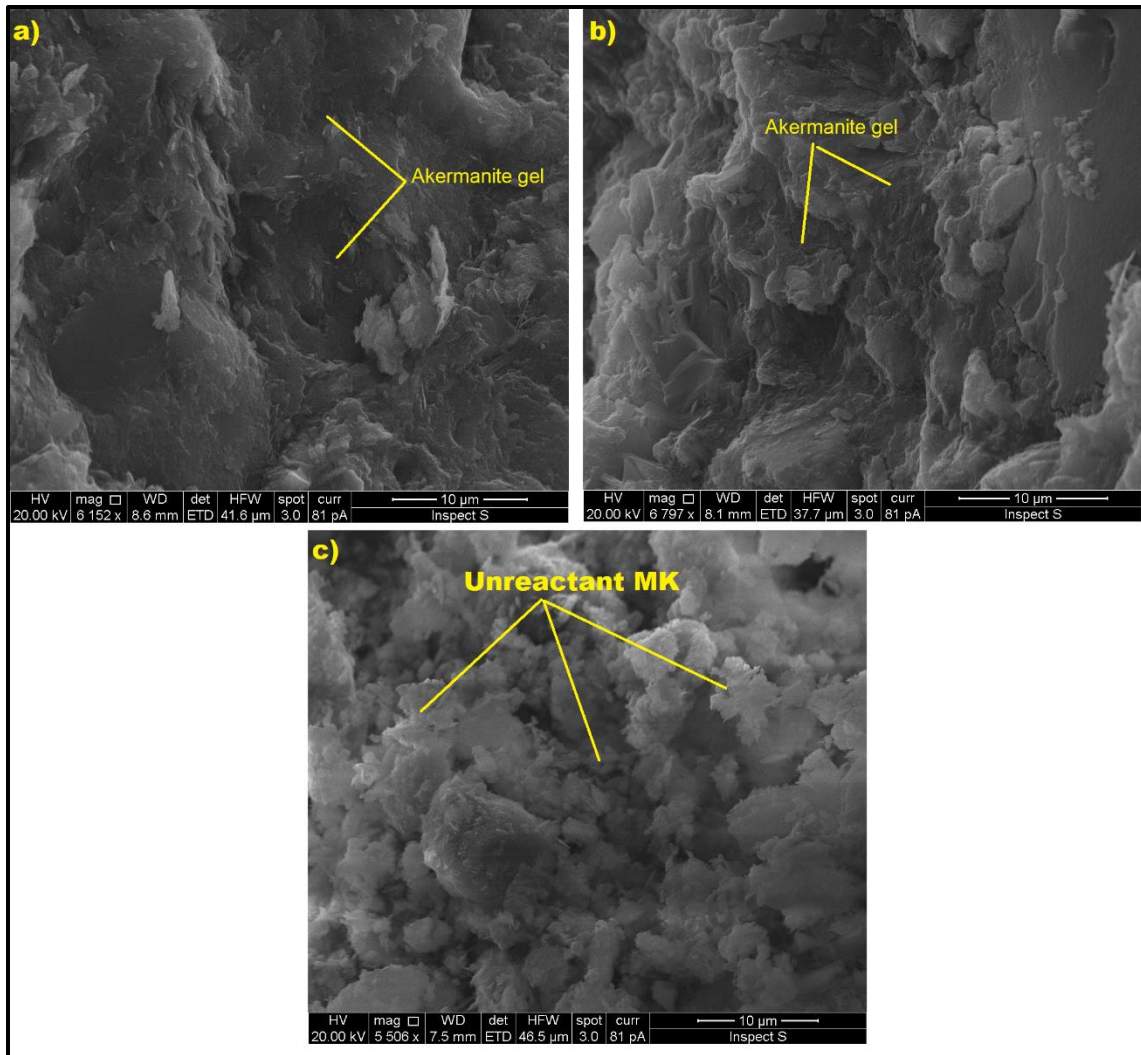
Figure 16. SEM micrographs for a) AAC3, b) AAC7 and c) AAC11 at 28 days

574 Fig. 17 presents the SEM micrographs of hardened pastes of optimum blends AAC3, 7, 11 at  
575 curing age of 90 days. While the matrix of blend AAC7 and AAC11, presented in Fig. 17b and c,  
576 is exhibiting un-reactant particles of MK. The amounts of these particles, decreased in blend AAC7  
577 at the age of 90 days compared to 28 days, with the development of more hydrates, such as Ak and  
578 St of darker grey tone. On the other hand, it can be significantly recognised from the microstructure  
579 of AAC3, as shown in Fig. 17a, that this blend is mostly free any un-reactant particles and showed  
580 higher microstructure density. This indicates a large transformation of MK and NP particles into  
581 dense gels of hydration products (Ak and St) through long term curing, which in turn resulted in a  
582 significant development of strength for the AAC3 blend.



583  
 584 *Figure 17. SEM micrographs for a) AAC3, b) AAC7 and c) AAC11 at 90 days*  
 585

586 While it could be clearly noticed that the internal matrix of AAC3 as in Fig. 18a at the age of 180  
 587 day, has significant increment of the formulation of a dense phase characteristic of cementitious  
 588 Ak compared to 90 day which strongly reveals the progression of alkali-activation till the age of  
 589 180 day and the total disappearance of any un-reactant grains. However as shown in Fig. 18b, this  
 590 dense gel was less presence in AAC7. On the other hand, the microstructure of AAC11 has shown  
 591 prevalence of un-reactant particles such as MK and very low gels of Ak as shown in Fig. 18c.  
 592



593  
 594 *Figure 18 SEM micrographs for a) AAC3, b) AAC7 and c) AAC11 at 180 days*  
 595

596 **4. Conclusions**

597 Based on the experimental outcomes of using thermally activated Lime Kiln Dust (LKD) that was  
 598 proposed as solid waste material rich in calcium oxide in the formulation of a second generation  
 599 one-part AAC cement, the following points can be summarised:

- 600 • More than 51% increase in strength after 28 days of curing just for highly reactive calcined  
 601 LKD at temperature of 950°C was achieved in the blend containing metakaolin (35 wt.%) and  
 602 natural pozzolan (30 wt.%).  
 603 • Significant increase of rate of strength development after 28 days of curing and continued until  
 604 180 days that reached 51 MPa for increased proportion of 950°C calcined LKD and metakaolin  
 605 was revealed.

- 606 • A high degree of alkali-activation was developing within the synthetic system until 180 days,  
607 without showing any marks of efflorescence or presence of unreacted alkaline substances were  
608 reported.
- 609 • The setting times of the optimised blends were in the acceptable range of setting times in the  
610 past studies of one-part AAC and cement types in British standards.
- 611 • According to the results of XRD, TG-DTA and FTIR, the thermal treatment at 950°C were  
612 evidently participating in breaking the crystalline phases of the original materials and showed  
613 strong evidence of transforming to amorphous phases.
- 614 • The characterisation findings by XRD, FTIR and SEM, have confirmed that the development  
615 of hydration products is responsible for the strength at 28, 90 and 180 days. These products  
616 include stratlingite, St ( $C_2ASH_8$ ) and Akermanite, (Ak) ( $Ca_2MgSi_2O_7$ ).
- 617 • The developed one-part AAC has introduced a comparable performance, in regard of the  
618 strength, setting times and morphology compositions, to the traditional AAC synthesised from  
619 commercial hazardous chemicals. However, there are some limitations, i.e. the durability  
620 investigations such as chloride and alkaline attacks which are recommended as a future work.  
621

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