

1 **Sewage Sludge Ash: A Comparative Evaluation with Fly Ash for Potential Use as Lime-**
2 **pozzolan Binders**

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12 **Abstract**

13 The disposal of sewage sludge ash (SSA) has become an environmental issue due to the
14 limited available landfilling space. This study aims at applying the finely-ground sewage
15 sludge ash (FSSA) with quicklime and hydrated lime respectively to develop a new type of
16 lime-pozzolan system and study the effects of different types of lime on the mechanical
17 properties of the systems. A traditional pozzolanic material (i.e. coal fly ash (FA)) was also
18 used to compare with the FSSA. Multiple techniques including X-ray diffraction (XRD),
19 scanning electron microscopy (SEM) and thermogravimetric analysis (TGA) were used to
20 assess the hydration kinetics and microscopic characteristics of the lime-pozzolan systems.

21 The results showed that the quicklime system exhibited lower strength than the hydrated lime
22 system, which was due to its lower reaction degree with the FSSA and the higher porosity
23 caused by the expansion during the vigorous hydration reaction of quicklime.

24 Compared to FA, the FSSA attained higher strength for the whole curing period (up to 90 d)
25 in the lime system due to the highly porous nature of FSSA leading to a lower effective water
26 to binder ratio. However, in the hydrated lime system, due to the higher overall pozzolanic
27 activity of FA, its long-term strength values gradually improved.

28 Besides, calcium phosphate hydrate crystals were detected by XRD in the FSSA; while some
29 clinotobermorite was found in the FA both from XRD and SEM, which might govern the
30 strength gain in the lime-FA system. Overall, the application of FSSA as a pozzolan in the
31 lime-pozzolan system could be a promising option to both relieve the waste disposal pressure
32 and provide a potential sustainable construction material.

33
34 **Keywords**

35 Finely-ground sewage sludge ash (FSSA); fly ash (FA); lime-pozzolan; hydration;
36 construction material; waste management.

37

38 **1. Introduction**

39 Lime-pozzolan binders have been widely studied and applied in construction since ancient
40 times. Specifically, the lime-volcanic ash mortars were first applied as a wall covering in
41 Santorini Island in 1500 BC [1]. The Romans utilized lime together with volcanic ash and
42 crushed sintered clay products in concrete for the construction of buildings and coastal works,
43 which had resisted erosion from seawater for over 20 centuries [2].

44 However, the invention of Portland cement (PC) in the 19th century caused the recession of
45 the use of the lime-pozzolan binders. But they have again become popular as alternatives to
46 PC recently due to the high cost, carbon footprint and energy intensity of PC. In addition,
47 concrete based on PC would normally suffer from long-term degradation problems, especially
48 under harsh conditions [2]. More importantly, the application of PC-based materials on
49 repairing ancient buildings is not allowed according to the requirements regulated by
50 European supervisory authorities which specify that the repairing materials should be
51 compatible with the original materials [3].

52 Pozzolans are commonly used as partial replacements for PC, as they are not only cost-
53 effective, but also improve some properties of the produced mortars and concrete. The role of
54 pozzolans in the lime-pozzolan binders is their participation in pozzolanic reactions with
55 calcium hydroxide to form hydration products, which are similar to those produced from PC
56 hydration including calcium silicate hydrates (CSH) and calcium aluminate silicate hydrates
57 (CASH) [4].

58 Extensive studies have investigated the characteristics, hydration, carbonation and
59 applications of lime-pozzolan pastes/mortars. They mostly focused on natural pozzolans such
60 as volcanic ash, artificial pozzolans such as brick dust and some common industrial waste
61 pozzolans such as fly ash (FA) and ground granulated blast furnace slag (GGBS) or a
62 combination of these [5]. In addition, various combinations of limestone, metakaolin, natural
63 zeolites, silica sand, crushed bricks/rocks, and siliceous wastes had been studied for the
64 production of lime-pozzolan binders [6-8]. It was found that different pozzolans have varying
65 pozzolanic activities [9].

66 Incinerated sewage sludge ash (ISSA), a by-product generated from the incineration process
67 of sewage sludge of wastewater treatment plants, is increasingly being generated and the
68 common management method is landfilling. Considering the limited availability of
69 landfilling space, it is no longer a sustainable waste management solution for ISSA [10, 11].
70 In order to resolve this issue, different recycling methods have been developed. The
71 characteristics of ISSA may vary significantly from place to place, but typical elemental
72 compositions of ISSA include P, Ca, Si, Al and Fe [12]. ISSA normally only possesses a
73 relatively low pozzolanic reactivity due to the presence of a large amount of crystallised SiO₂
74 and the particles are with irregular shapes and large porosity [13, 14]. Even so, as a valuable

75 resource, the ISSA has a great potential to be applied for the production of cement, aggregates,
76 mortars, concrete and controlled low-strength materials [15]. In addition, many researchers
77 have studied the use of ISSA as supplementary cementitious materials [11, 16-18] and for soil
78 stabilisation [19-21].

79 For its application in the cement-based materials as a pozzolan, it was found that 20%
80 replacement of cement with ISSA would not adversely affect the mortar strength [11]. The
81 pozzolanic activity of ISSA was found to be lower than FA, but the ISSA could still improve
82 the strength of mortars containing ISSA at the later curing stage [22]. At 5% and 10%
83 replacement levels, the 28-day mortar strengths were comparable to the control [23]. Increase
84 the fineness of ISSA by grinding could improve the mechanical properties through
85 enhancement of its pozzolanic activity [24]. Generally, the ISSA could be used as a low-grade
86 pozzolan which can save natural resources for cement production and the associated carbon
87 emissions [10, 17].

88 The use of wastes as secondary building materials has attracted increasing attention due to
89 environmental protection issues. Using the ISSA as a binder for soil stabilisation has been
90 demonstrated to help to facilitate the use of soft soils [25]. Some studies used up to 16% ISSA
91 to treat cohesive soils or clay and obtained improved geotechnical properties such as
92 increased unconfined compressive strength and decreased swelling pressure [19, 26].
93 However, in order to have higher strength, a small portion of cement or hydrated lime were
94 added to promote the pozzolanic reaction of ISSA [27-29]. A previous study reported that a
95 maximum of 26% strength increase was attained in the soil-cement mixture with 20% ISSA
96 [20]. By using ISSA and hydrated lime, soft subgrade soil could be stabilised to achieve 3 to 7
97 times higher unconfined compressive strength and improved shear strength. It was also
98 concluded that an 8% ISSA-hydrated lime mixture was the optimal content added in the soils
99 for effective stabilisation treatment [28].

100 Different types of lime also affected the reactions with the pozzolans and hence the strength
101 development. The heat liberated from quicklime (CaO) hydration could accelerate the early
102 pozzolanic reactions, because the freshly-produced Ca(OH)_2 from lime hydration had a higher
103 solubility than the commercially-available hydrated lime (Ca(OH)_2) [30]. It was found that the
104 hydrated lime-pozzolan pastes had a higher porosity than the lime-pozzolan pastes, which
105 indicated that higher compressive strength was obtained by the lime-pozzolan pastes [30].

106 However, very limited information is available on the lime- sewage sludge ash (SSA) system.
107 In order to better understand the reactions between quicklime/hydrated lime and SSA, finely-
108 ground SSA (FSSA) that possessed a relatively higher pozzolanic activity than the as-received
109 SSA was used in this study. FA, as a representative of common supplementary cementitious
110 materials, was also used for comparison. The hydration heat of the fresh pastes and the
111 mechanical properties of the hardened pastes were evaluated. The reaction products and

112 microstructural properties were assessed by using X-ray diffraction (XRD), scanning-electron
113 microscopy (SEM) and thermogravimetric analysis (TGA) to elucidate the mechanisms
114 governing its performance.

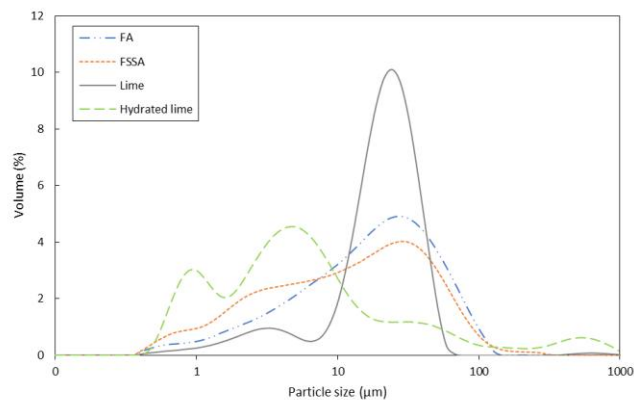
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116 2. Materials and methods

117 2.1 Materials

118 The FSSA used in this study was collected from the T-Park (a sewage sludge incineration
119 plant) in Hong Kong. The ash was dried in a 105°C oven overnight and finely ground in a ball
120 mill for two hours. The quicklime and hydrated lime were commercially available from local
121 suppliers, and FA was provided by a local coal power plant.

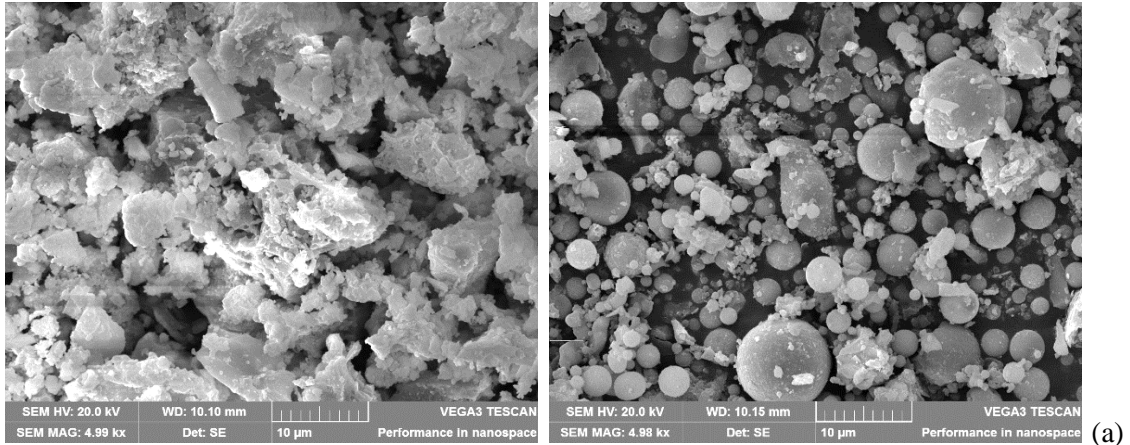
122 The particle size distributions of quicklime (lime, CaO), hydrated lime (CH), FSSA and FA
123 measured by using a Malvern Mastersizer 3000 are presented in Fig. 1. The mean particle
124 sizes were 27.48, 40.11, 23.77 and 26.6 μm for lime, hydrated lime, FSSA and FA,
125 respectively. The FSSA particles were slightly finer than the FA particles.



126

127 **Fig. 1** Particle size distributions of raw materials

128 The morphologies of gold-coated FSSA and FA samples were obtained by using a scanning
129 electron microscope (SEM, TESCAN VEGA3), as presented in Fig. 2. It is clear that the
130 FSSA particles were porous and a relatively rough surface. The FA particles were largely
131 spherical in shape. Fig. 3 presents the XRD spectra of FSSA and FA. The main crystalline
132 phases of FSSA are quartz (SiO₂), hematite (Fe₂O₃), magnetite (Fe₃O₄), leucite (KAlSi₂O₆)
133 and anorthoclase ((Na,K)AlSi₃O₈). In the FA, there are mainly quartz, mullite
134 (3Al₂O₃·2SiO₂), magnetite, and hematite.

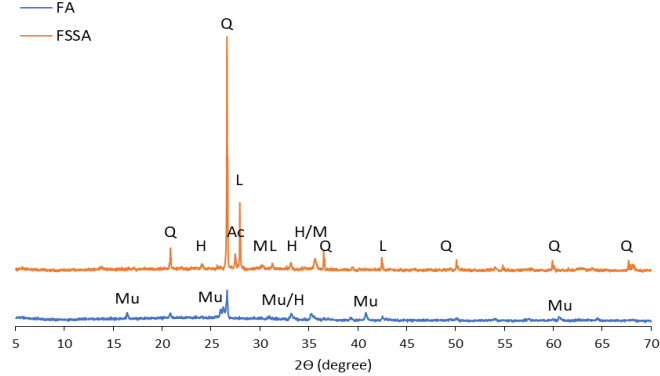


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FSSA

(b) FA

Fig. 2 SEM images of (a) FSSA and (b) FA



138

139 **Fig. 3** XRD patterns of FSSA and FA (Q: quartz; Mu: mullite; L: leucite; H: hematite; M:
140 magnetite; Ac: anorthoclase)

141 The chemical compositions of quicklime, hydrated lime, FSSA and FA determined by using a
142 Rigaku Supermini200 X-ray fluorescence (XRF), are shown in **Table 1**. The major
143 constituents of FSSA were SiO_2 , Al_2O_3 and Fe_2O_3 and the FA was mainly consisted of SiO_2
144 and Al_2O_3 . The Brunauer-Emmett-Teller (BET, Micromeritics ASAP 2020 PLUS) surface
145 areas and loss on ignition (LOI) of raw materials under 950°C are also listed in **Table 1**.

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155 **Table 1.** Oxide compositions (%) and physical characteristics of materials

Oxides	Hydrated lime	Quicklime	FSSA	FA
SiO ₂	1.22	1.47	37.04	44.42
Al ₂ O ₃	0.15	0.68	15.24	32.56
Fe ₂ O ₃	0.13	0.16	14.03	6.49
CaO	97.69	96.56	6.91	6.67
MgO	0.59	0.72	2.80	1.86
K ₂ O	0.06	0.12	2.77	1.81
Na ₂ O	/	/	7.11	1.79
TiO ₂	/	/	0.38	1.24
SO ₃	/	0.13	3.66	2.27
P ₂ O ₅	0.12	0.12	9.12	0.44
BET (m ² /kg)	10889	3479	2866	1742
Specific gravity (kg/m ³)	2210	3340	2740	2330
LOI (%)	/	/	1.41	3.76

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157 **2.2 Paste samples preparation**

158 The pastes preparation followed the mix design in **Table 2** and the water to binder (w/b) ratio
 159 was fixed at 0.38 for all the mixes, as several w/b ratios were tried and 0.38 was able to
 160 produce a cohesive mixture without bleeding.

161 **Table 2.** Mix design of different mixtures

Sample ID	Quicklime	Hydrated lime	FSSA/ FA	w/b	Curing age
Control			1	0.38	
10%lime-FSSA/FA	0.1		0.9	0.38	
20%lime-FSSA/FA	0.2		0.8	0.38	
30%lime-FSSA/FA	0.3		0.7	0.38	7, 28 & 90d
10%CH-FSSA/FA		0.1	0.9	0.38	
20%CH-FSSA/FA		0.2	0.8	0.38	
30%CH-FSSA/FA		0.3	0.7	0.38	

162

163 The powder was dry mixed for one minute at a low speed by a standard mechanical drum
 164 mixer before adding water, and then another three minutes of mixing (1 min at low speed, 2
 165 mins at high speed) was carried out to obtain the homogenous pastes. After mixing, the fresh
 166 mixture was cast into cubic steel moulds with a size of 20*20*20 mm and then subjected to
 167 vibration for about one minute to remove entrapped air in the samples. Lastly, the moulds
 168 were covered by a plastic film and placed at room temperature (about 25 °C) for three days.
 169 Afterwards, the pastes were demoulded and subsequently placed in an environmental chamber
 170 controlled at 95% RH and 23±2 °C until testing.

171

172 **2.3 Testing programme**

173 **2.3.1 Thermal liberation of fresh pastes**

174 A Calmetrix I-Cal 4000 calorimeter was used to perform the isothermal calorimetry test
 175 where 10~30 wt.% lime and hydrated lime was used to replace FSSA and FA. The fresh
 176 pastes first were mixed outside the calorimeter at 500 rpm for three minutes and then placed

177 in the four-channel calorimeter which recorded the heat evolution profiles at 20°C for 72
178 hours.

179 2.3.2 Compressive strength of harden pastes

180 For the compressive strength test, the 20*20*20 mm cubic paste samples were tested by a
181 TESTOMETRIC CXM loading machine. The loading rate was 0.3mm/min and triplicates
182 were tested to obtain the average strength values.

183 2.3.3 Microscopic and chemical analysis

184 Small pieces of the broken paste samples were placed in absolute ethanol (99.8%+,
185 International Laboratory USA) for one week to arrest the hydration. After solvent exchange,
186 the samples were kept in a vacuum desiccator for three days prior to the tests. For TGA and
187 XRD tests, further grinding manually in an agate mortar and sieving (< 75 μm) was necessary.
188 The crystalline-phase mineralogy of the samples was evaluated by using a high-resolution
189 powdered X-ray diffractometer (Rigaku SmartLab, Japan) under the condition of 20 mA and
190 40 kV with CuKα radiation. The 2θ range of 5 to 70° with a step interval of 5°/min was used
191 in the tests. Then, a Highscore plus software with an ICDD PDF 2010 database (International
192 Centre for Diffraction Data, Powder Diffraction File) was used to identify the peaks.

193 For the SEM test, the representative fresh fractured surfaces of the pastes were selected, and
194 gold coated first. Their morphologies and element distributions were then tested by using a
195 SEM-energy dispersive X-ray (EDX) analyser operating at a working tension of 20 kV.

196 A Rigaku Thermo Plus EVO2 was used for TGA. A total of 10 mg sample was heated from
197 50 to 1000°C with a rate of 10°C/min, under 20 ml/min Argon flux. The weight loss from
198 TGA at different temperature ranges was useful for quantifying the content of Ca(OH)₂ and
199 CaCO₃.

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201 3. Results and discussion

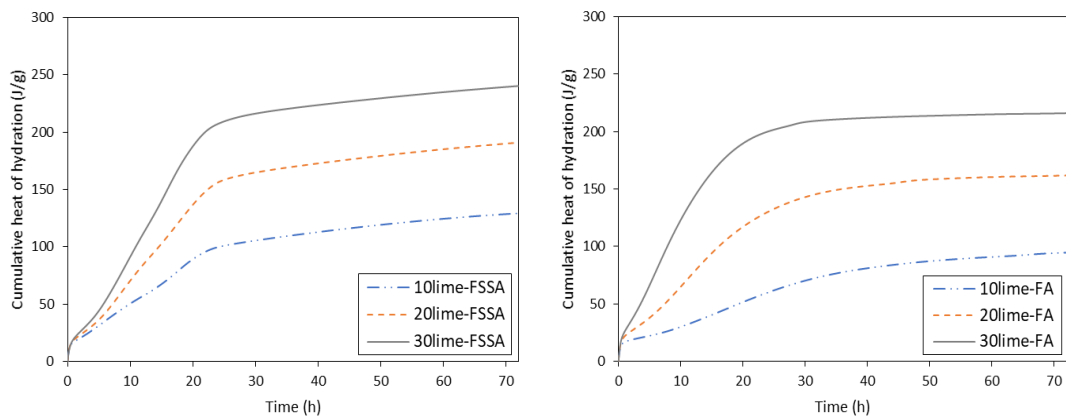
202 3.1 Hydration evolution

203 Fig. 4 shows the cumulative hydration heat of hydration of both the quicklime (lime) and
204 hydrated-lime (CH) pastes preapred with FSSA and FA incorporation. From Fig. 4 (a), it is
205 clear that the total heat evoloved increased with a greater amount of lime with both the FSSA
206 and FA pastes, and the total heat of the lime-FSSA pastes was larger than that of the lime-FA
207 pastes at all replacement levels within the first 72 hours. This may indicate that higher
208 reaction rate and degree were achieved by FSSA due to its larger surface area and lower
209 effective w/b ratio as the porous FSSA particles took up a certain quantity of the mixing water.
210 The evoloved heat of the quicklime pastes mainly came from the lime dissolution and the
211 generated heat probably promoted the reactions between lime and FSSA/FA. This is
212 consistent with the gradually increased gradients of the heat flow curves when higher amounts

213 of lime were employed. Therefore, the use of lime indeed accelerated the lime-FSSA/FA
214 reactions at the early stage, especially within the first 24 hours.

215 However, as shown in Fig. 4 (b), the cumulative heat evolution of the CH pastes was very low
216 compared with the quicklime pastes and could be nearly regarded as neglectable, even though
217 a higher amount of CH led to greater amounts of heat. This may be because the CH was
218 relatively insoluble in water and could not react with the FSSA/FA immediately at the early
219 age. As mentioned by Shi [30], several days were needed to reach the dissolution equilibrium
220 of $\text{Ca}(\text{OH})_2$. The extremely low amount of heat evolved rendered the calorimeter very
221 susceptible and sensitive to the changes in the room temperature at which the calorimeter
222 was placed. That might explain why there were fluctuations of the measured values. The total
223 heat generation of the CH-FSSA pastes was also greater than that of the CH-FA pastes at all
224 replacement levels, which again verified the higher reaction rate and degree of FSSA
225 compared with FA within 72 hours.

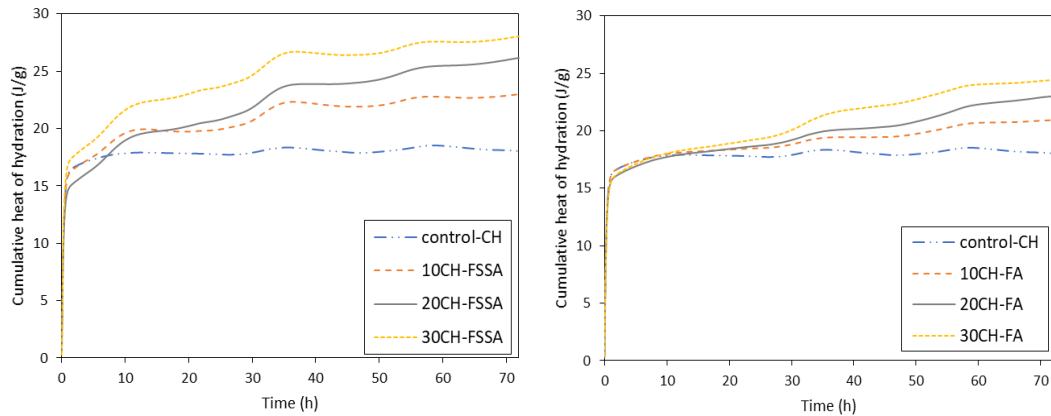
226 For better interpretation of the differences between the lime and CH system, the heat
227 evolution rate of the lime-FSSA paste was plotted to compare with that of the CH-FSSA
228 paste, as presented in Fig. 4 (b). There was a sharp peak at around 10 hours after mixing
229 which corresponded to the lime-FSSA reactions and the higher peak rate was attained by a
230 higher amount of lime. The sharpness could be due to the very fine particles of FSSA and its
231 large surface area. However, in the CH system, the scale of heat evolution rate (Y-axis) was
232 much smaller than the lime system and the seemingly regular fluctuations can be attributed to
233 the sensitivity of the calorimeter to daily temperature changes of the laboratory environment.



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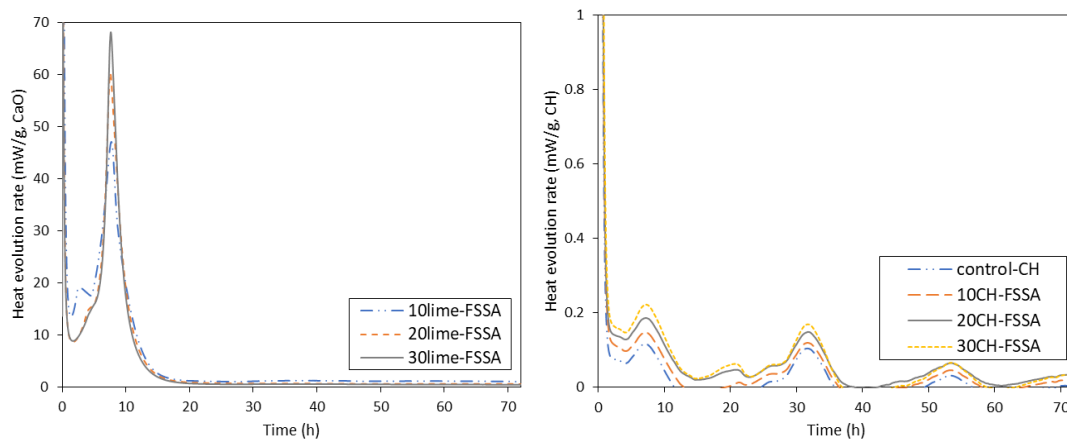
(a)



236

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(b)



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(c)

Fig. 4. Heat flows of (a) lime-FSSA/FA, (b) CH-FSSA/FA pastes, and (c) Heat evolution rates of lime-FSSA & CH-FSSA

240

241

242

243 3.2 Compressive strength

244 The compressive strength of the lime/CH pastes containing FSSA and FA at the curing age of

245 7, 28 and 90 days are presented in Fig. 5. As shown in Fig. 5 (a), a higher compressive

246 strength was obtained by the lime-FSSA pastes than the lime-FA pastes at all curing ages.

247 This was also consistent with the higher cumulative hydration heat of the lime-FSSA pastes in

248 the previous section. Donatello and Cheeseman [12] concluded that SSA had a strongly

249 positive pozzolanic activity in the saturated lime test. The strength of the lime-FSSA pastes

250 increased with the increasing amount of lime used (from 10% to 30%) at all the studied curing

251 time. However, the lime-FA pastes showed much lower strength than the lime-FSSA pastes at

252 7 and 28 days, while the strength suddenly picked up at 90 days as the FA was mostly

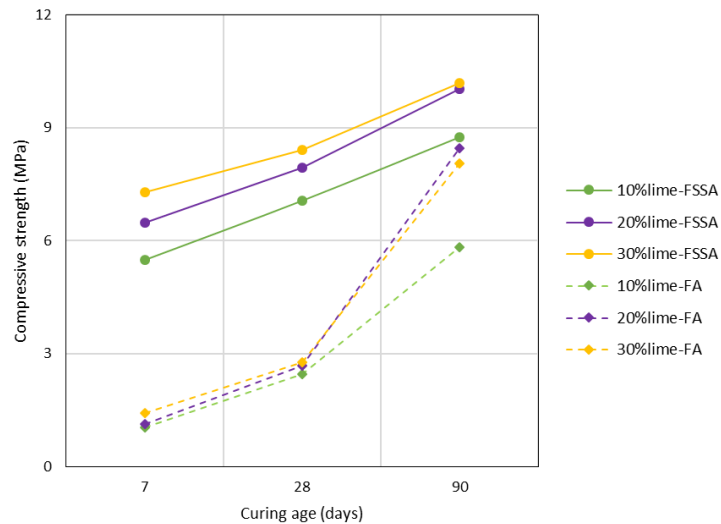
253 activated and reacted at the later age [31]. The higher strength of the lime-FSSA paste at the

254 early age was partly attributed to the lower effective w/b ratio due to the high water

255 adsorptivity of porous FSSA particles [22]. Besides, it had been suggested that the reaction

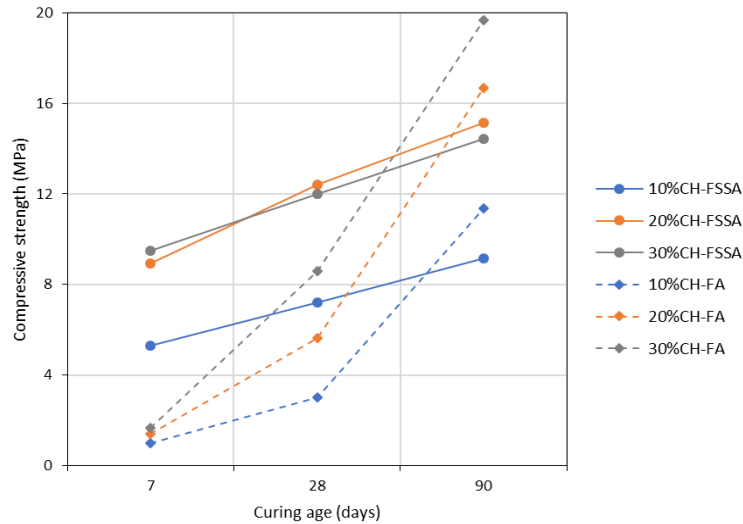
256 between $\text{Ca}(\text{OH})_2$ and SSA had mechanical outcomes as they led to moderate but critical

257 strengthening of the pastes at 21 and 150 days [18]. Therefore, it can be envisaged that some
 258 new phases formed in the lime-FSSA pastes contributed to the strength development.
 259 It can be seen that from Fig. 5 (b) the 20% and 30% CH-FSSA pastes had similar strength
 260 values and were much higher strength than the 10% CH-FSSA pastes. One possible reason
 261 was that 10% of CH was too little to well react with the FSSA. The strength of the CH-FSSA
 262 pastes almost linearly increased with the curing time. For the CH-FA pastes, all the strength
 263 values were comparable at 7 days, and were lower than that of the CH-FSSA pastes, which
 264 might be due to the slow pozzolanic reaction of FA at the early stage. Also, there might be
 265 insufficient soluble $\text{Ca}(\text{OH})_2$ for the reaction with FA. Helmuth [32] found that the minimum
 266 amount of $\text{Ca}(\text{OH})_2$ for complete reaction with class F FA was 45% by weight. However,
 267 after 28 days of curing, the CH-FA pastes experienced a faster rate of strength gain and the
 268 strength increased with the higher amount of CH. When the curing age was extended to 90
 269 days, the strength values attained were even higher than the strengths of the CH-FSSA pastes
 270 at each level of CH addition. That was because the FA had a higher overall pozzolanic
 271 reactivity than the FSSA, which was confirmed by using the R3 method [Ali submitted
 272 manuscript to CBM]. This was related to more amorphous (reactive) phases were present in
 273 FA than FSSA providing more precursors for the formation of hydration products [22, 32].



274
 275

(a)



(b)

Fig. 5. Compressive strength of (a) lime pastes and (b) CH pastes with FSSA and FA

Apart from that, the slight expansions of the lime system due to exothermic reactions of lime hydration were observed which might cause some micro cracks in the paste matrix reducing the strength of the lime system. The strength difference within each system was also dependent on the consumption/reaction degree of $\text{Ca}(\text{OH})_2$ which will be discussed in section 2.4.

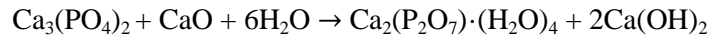


Fig. 6. Photo of lime-FSSA pastes showing slight expansion (10%~30% quicklime from left to right)

3.3 Microstructural analyses

3.3.1 Crystalline phases by XRD analyses

The XRD spectra of lime and CH pastes prepared with FSSA and FA at 28 and 90 days are presented in Fig. 7. As depicted in Fig. 7 (a), anorthoclase (Ac), hematite (H) and leucite (L) were the original phases of FSSA. Calcium phosphate hydrates (CPH) were newly formed in the lime-FSSA pastes. According to Donatello and Cheeseman [12], phosphorous (P) existed as $\text{Ca}_3(\text{PO}_4)_2$ in the SSA. Hence, the formation of CPH could be deduced as the following reaction:



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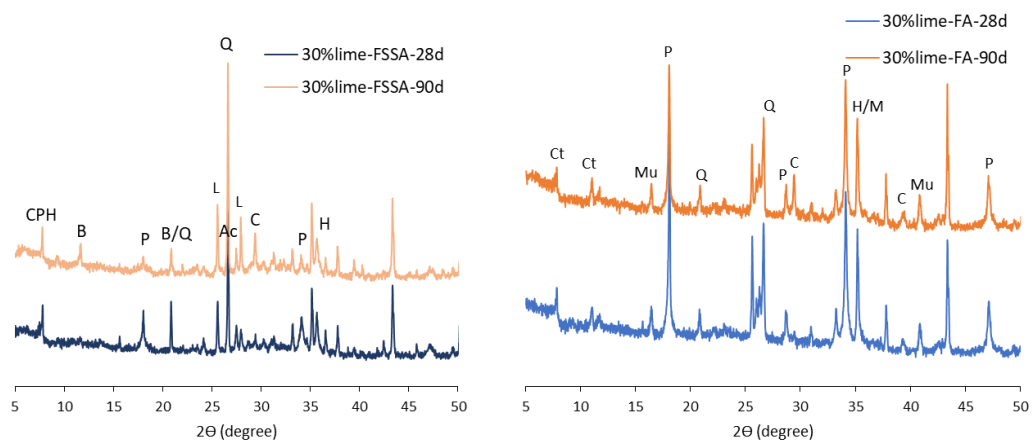
297 A new phase (brushite) was also weakly detected in the lime-FSSA pastes, especially at 90
 298 days. Besides, the calcite peaks were barely present at 28 days but became distinguishable at
 299 90 days due to the continuing carbonation of lime.

300 For the lime-FA pastes (Fig. 7 (a)), except the phases that were originally present in the FA
 301 (mullite, quartz, hematite/magnetite), calcite and portlandite peaks were detected at both 28
 302 and 90 days, especially for the intensive peaks of portlandite. This might indicate the
 303 moderate reactions between FA and lime, resulting in the lower strength of the lime-FA
 304 pastes, compared with the lime-FSSA pastes. Besides, at both curing ages, the formation of
 305 calcium silicates – (clinotobermorite) was also observed due to the pozzolanic reactions
 306 between $\text{Ca}(\text{OH})_2$ and FA. According to He et al. [33], clinotobermorite was produced in
 307 cement pastes prepared with FA [34].

308 As shown in Fig. 7 (b), portlandite was weakly detected in the CH-FSSA paste, and the 90-d
 309 peak intensity slightly decreased when compared to 28-d, due to the pozzolanic reactions
 310 between $\text{Ca}(\text{OH})_2$ and FSSA. The produced CPH was only present at 28 days, and it is noted
 311 that calcite due to its limited amount was not identified by XRD.

312 The CH-FA paste at 28 days showed strong intensities of clinotobermorite, suggesting its
 313 relative higher amount in the paste. However, the relative amount of clinotobermorite
 314 significantly decreased after reaching 90 days. Compared with the lime-FA pastes, the calcite
 315 peak in the CH-FA paste was much weaker, as the former porous lime pastes were easier to
 316 be carbonated.

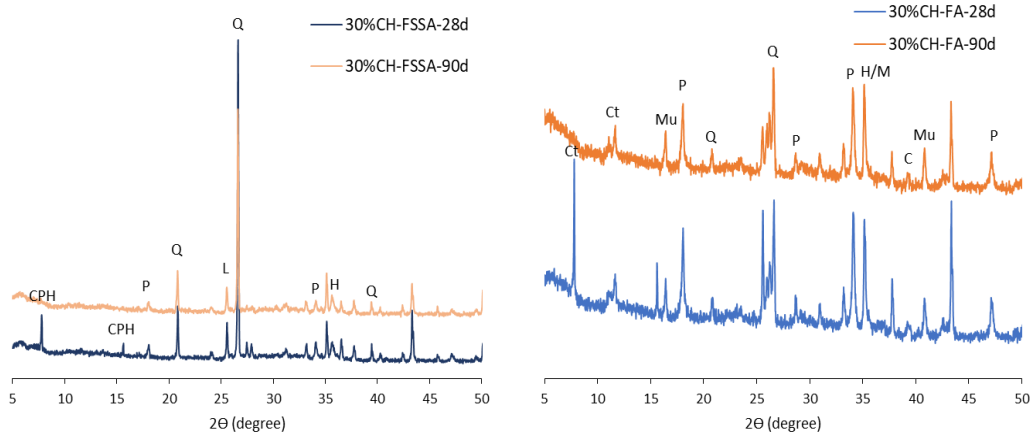
317 Based on the above, the CPH and clinotobermorite might separately contribute to the strength
 318 gain of the lime-FSSA and the lime-FA pastes, while these phases could only be beneficial for
 319 the strength gain of the CH-FSSA/FA pastes at a rather late stage. However, some amorphous
 320 phases that could not be identified by XRD will be investigated in the following section.



321

322

(a)



(b)

Fig. 7. XRD spectra of 30% (a) lime pastes and (b) CH pastes, at 28 & 90d

(CPH: Calcium phosphate hydrate $\text{Ca}_2(\text{P}_2\text{O}_7)(\text{H}_2\text{O})_4$; B: Brushite ($\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$); C: Calcite (CaCO_3); P: Portlandite; Ct: Clinotobermorite; H: Hematite; M: Magnetite; L: Leucite; Ac: Anorthoclase; Q: Quartz; Mu: Mullite)

3.3.2 SEM

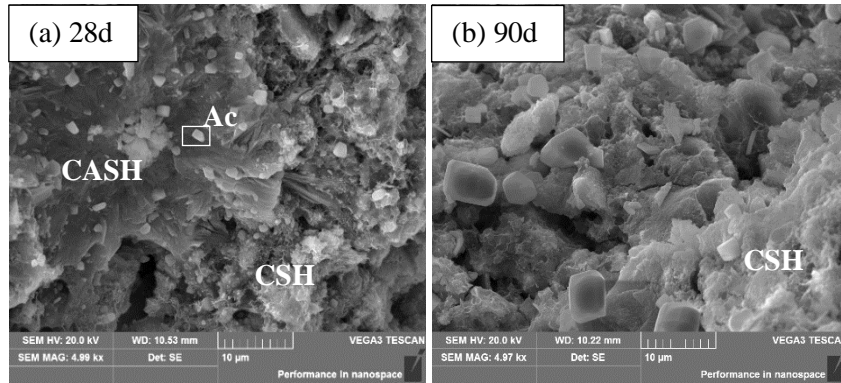
Fig. 8 shows the selected SEM images together with EDX results obtained from the 30% lime and CH pastes prepared with FSSA and FA respectively after 28- and 90-day curing. In **Fig. 8(a)**, it can be seen that the calcium alumina silicate hydrate (CASH) and CSH gels were formed and the structure of the 28-d lime-FSSA paste was less dense than that of the 90-d paste (**Fig. 8 (b)**). In the lime-FSSA paste at 90 days, there were some unreacted FSSA particles intersecting with CSH gels.

For the lime-FA pastes, as shown in **Figs. 8 (c) and (d)**, significant amounts of hydration products were formed and covered the spherical FA particles, some of which were still visible, suggesting the relatively low reactivity between FA and lime. The pores in the pastes were gradually filled with those hydration products that made the microstructure of the hydrated matrix denser, leading to the strength improvement at 90 days [35]. This agreed well with the strength results of the lime-FA pastes.

In **Fig. 8 (e)**, there were some calcite crystals located inside and on the surface of the FSSA particles. But after 90 days (**Fig. 8 (f)**), the FSSA particles were almost covered by hydration products and the paste structure became much denser, compared with the 28-d paste. Similar findings could be seen in the CH-FA pastes too, shown in **Fig. 8 (g) and (h)**. Calcite crystals were also present at both curing ages and some unreacted FA particles were observed in **Fig. 8 (g)**. However, the spherical FA particles and platy portlandite crystals were hardly seen after 90 days of curing, indicating high consumption of $\text{Ca}(\text{OH})_2$. This explained that the strength of the CH-FA pastes caught up with that of the CH-FSSA paste at this age. Besides, less pores

351 can be found in the 90-d paste, which was consistent with a noticeable increase in the strength
 352 development. Also, except for the crystallised clinotobermorite, some amorphous CSH could
 353 be observed in the CH-FA paste at 28 and 90 days. Based on EDX spectrum in Fig. 8 (g1), the
 354 Ca/Si ratio was about 0.82, similar to 0.83 which was the Ca/Si ratio of 11.3 Å tobermorite
 355 (clinotobermorite) [36, 37].

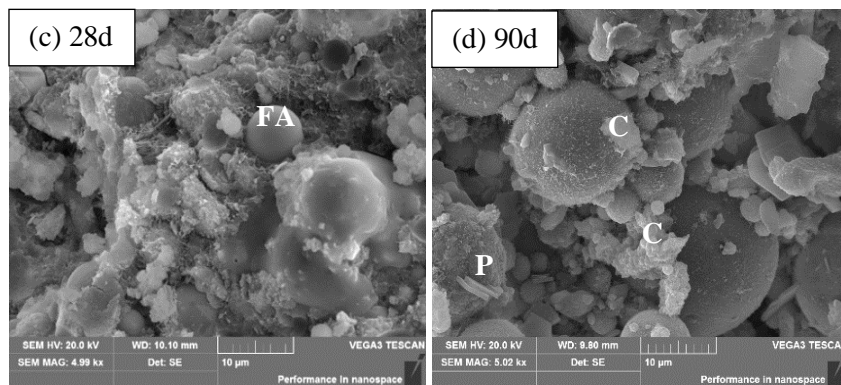
356



357

358

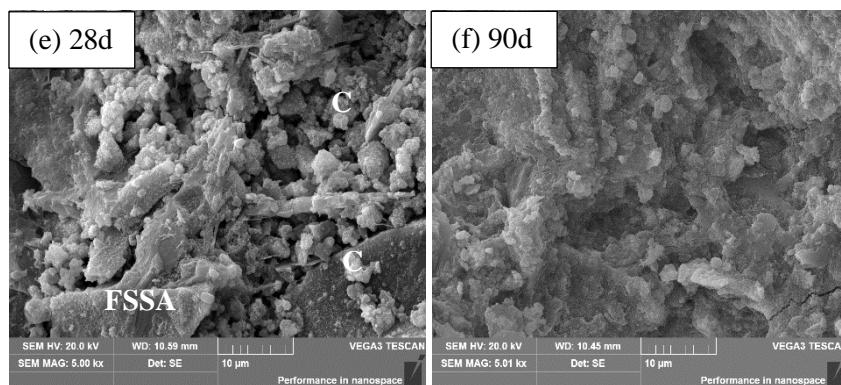
lime-FSSA



359

360

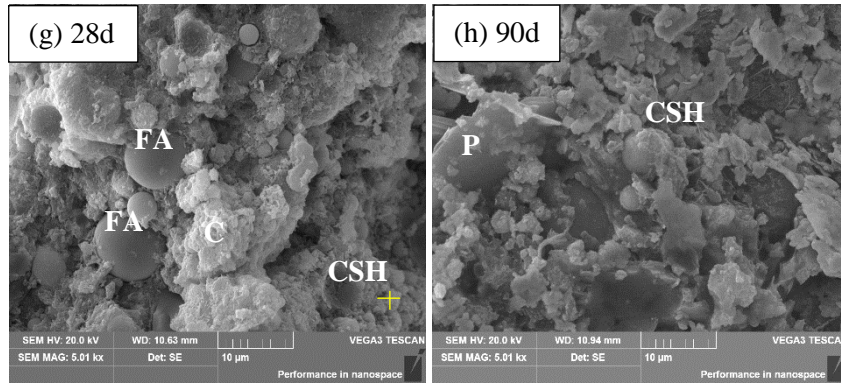
lime-FA



361

362

CH-FSSA



CH-FA

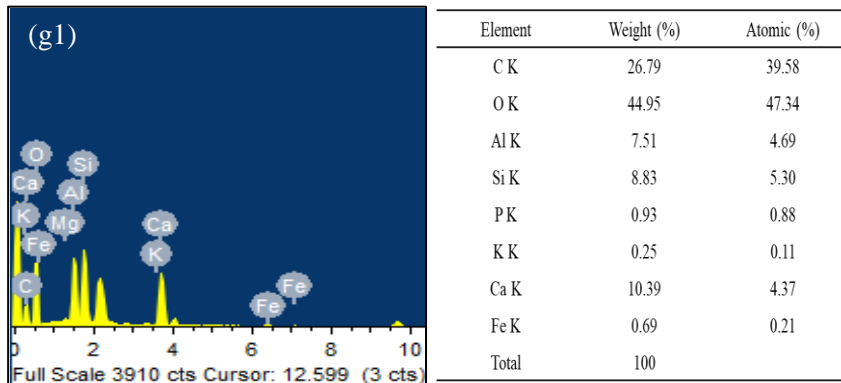
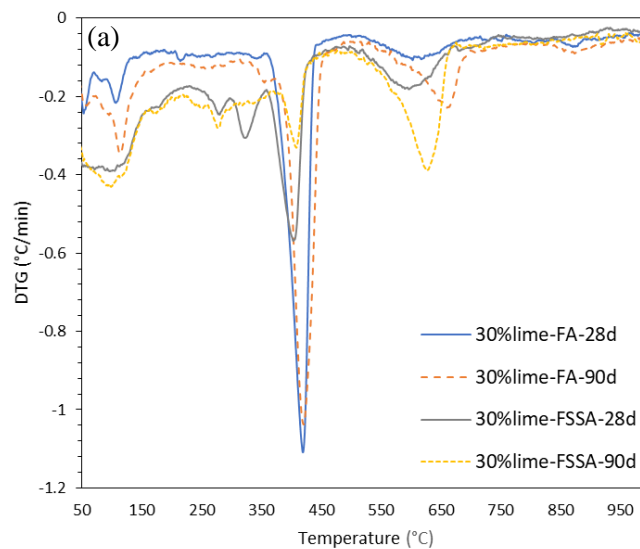
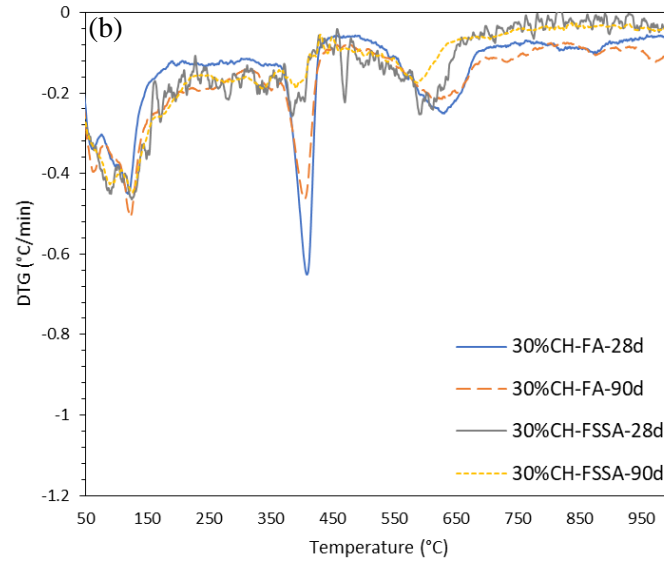


Fig. 8. SEM micrographs of the lime-FSSA pastes (a) 28d, (b) 90d; lime-FA pastes (c) 28d, (d) 90d; CH-FSSA pastes (e) 28d, (f) 90d; CH-FA pastes (g) 28d, (h) 90d; and (g1) EDX result of (g)

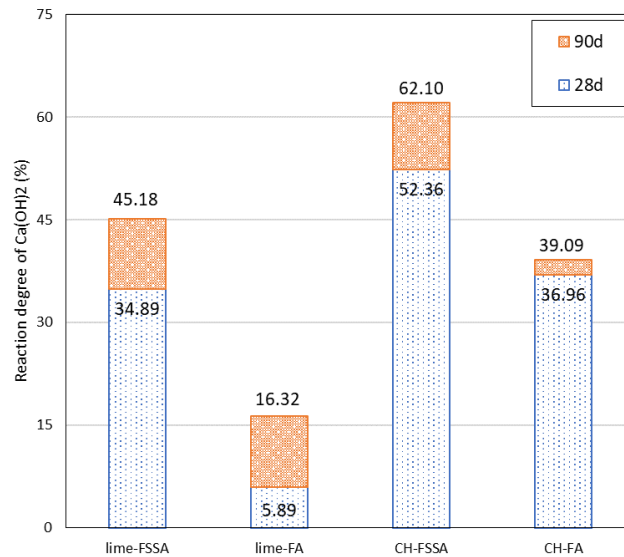
3.4 TGA results





372

373 **Fig. 9.** DTG profiles of (a) lime pastes and (b) CH pastes at the curing age of 28 & 90 days



374

375 **Fig. 10.** Reaction degree of calcium hydroxide of lime/CH pastes containing FSSA, FA at 28
376 and 90d

377 The DTG data of 30% lime/CH pastes containing FSSA and FA at the curing ages of 28 and
378 90 days are plotted in Fig. 9. The main endothermic peak around 450°C was attributed to
379 dehydroxylation of Ca(OH)_2 and that at the temperature range of 600-800°C was
380 corresponded to the decarbonation of calcium carbonates (CC). For the lime system (Fig. 9
381 (a)), the lime-FA pastes had a much stronger dehydroxylation peak of Ca(OH)_2 than the lime-
382 FSSA pastes. Fig. 10 illustrates the reaction degree of CH in both the lime and CH pastes. The
383 reaction degree of Ca(OH)_2 was computed by the following equation:

$$\begin{aligned} \text{Ca(OH)}_2 \text{ reaction degree (\%)} &= 100 - \text{Ca(OH)}_2 \text{ content (\%)} \\ &= 100 - (4.11\text{MassCa(OH)}_2 + 1.68\text{MassCC}) \end{aligned}$$

384 where MassCa(OH)_2 is the mass loss (%) of Ca(OH)_2 decomposition between 350-500°C,
385 MassCC is the mass loss (%) of CC decomposition between 550-750°C [38].

386 It can be seen that a significant amount of Ca(OH)_2 was left in the lime-FA pastes and the
387 amount remained relatively stable with the curing time. This may explain the relatively lower
388 reactivity and strength of the lime-FA pastes, implying that the FSSA was more reactive in
389 the lime system than the FA. Also, a high content of unreacted Ca(OH)_2 was left in the lime-
390 FA pastes lowered the strength of the lime-FA paste [30].

391 For the lime-FSSA pastes, the reduction of Ca(OH)_2 content suggested a much higher reaction
392 degree of FSSA in the lime system, as shown in Fig. 10. It is noteworthy that another small
393 peak was observed between 260-320°C in the lime-FSSA paste, which might be attributed to
394 decomposition of CPH, as a previous study reported that the hydrated calcium phosphate
395 dehydrated at around 300°C [39]. CPH was also identified in the XRD results. The
396 decarbonation peak was mainly due to the carbonation of lime and became more obvious
397 from 28 to 90 days.

398 As shown in Fig. 9 (b), at the same curing age, the Ca(OH)_2 decomposition peak intensity of
399 the CH-FSSA pastes was much weaker than that of the CH-FA pastes. Also, the CH-FSSA
400 showed a higher reaction degree of Ca(OH)_2 than the CH-FA pastes at 28 and 90 days (Fig.
401 10), which meant that the FSSA consumed more Ca(OH)_2 than the FA. This finding was
402 consistent with the higher strength value of the CH-FSSA pastes at 28 days. However, even
403 though the CH-FA paste showed a lower Ca(OH)_2 reaction degree at 90 days, the strength of
404 the CH-FA paste was higher than that of the CH-FSSA paste. This may be attributed to a
405 higher content of CC in the CH-FA paste at the later age leading to a lower reaction degree
406 and a higher pozzolanic activity of FA.

407

408 **4. Conclusion**

409 The use of lime and hydrated lime had different influences on the pozzolanic reactions and
410 strength development of the FSSA and FA pastes. The findings from the study may provide a
411 potential new type of lime-pozzolan binder option by recycling sewage sludge ash. The
412 following conclusions can be drawn from this study:

413 (1) The FSSA generated a higher amount of hydration heat and reacted faster than the FA
414 in the lime pastes, while the CH pastes generated very little amount of heat with both
415 FSSA and FA. The heat produced from lime hydration accelerated the reactions
416 between FSSA/FA but induced expansion to the paste samples.

417 (2) In both quicklime and hydrated lime systems, the strength of FSSA pastes were
418 higher than that of FA pastes at the early curing ages due to the porous nature of

419 FSSA which enabled a lower effective w/b ratio. The CH-FA pastes showed a
420 comparable strength in the long term due to the higher pozzolanic activity of FA.

421 (3) The use of FSSA was beneficial for the management of wastes with very high water
422 contents such as contaminated marine sediments.

423

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429

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