

Suitability of Sarawak and Gladstone Fly Ash to Produce Geopolymers: A Physical, Chemical, Mechanical, Mineralogical and Microstructural Analysis

Author

Leong, Hsiao Yun, Ong, Dominic Ek Leong, Sanjayan, Jay G, Nazari, Ali

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1 **Suitability of Sarawak and Gladstone fly ash to produce geopolymers: A physical,**
2 **chemical, mechanical, mineralogical and microstructural analysis**

3
4 **Hsiao Yun Leong^a, Dominic Ek Leong Ong^a, Jay G. Sanjayan^b and Ali Nazari^{b*}**

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6 a) Research Centre for Sustainable Technologies, Faculty of Engineering, Science & Computing, Swinburne
7 University of Technology Sarawak Campus, 93350 Kuching, Sarawak, Malaysia.

8 b) Centre for Sustainable Infrastructure, Faculty of Science, Engineering and Technology, Swinburne
9 University of Technology, PO Box 218, Hawthorn Victoria 3122, Australia.

10 * Corresponding author, Email: alinazari@swin.edu.au; Tel: +61 3 9214 8370

11
12 **Abstract**

13 Two types of fly ash sourced from Sarawak, Malaysia and Gladstone, Australia reflect
14 differences in chemical compositions, mineral phase and particle size distributions. In this
15 paper, the Sarawak fly ash was used to produce geopolymer in comparison to the well-
16 developed Gladstone fly ash-based geopolymer. Characteristics of fly ash and mixtures
17 proportions affecting compressive strength of the geopolymers were investigated. It is found
18 that the variations of both fly ash types on particle size distributions, chemical compositions,
19 morphology properties and amorphous phase correspond to the compressive strength. The
20 results obtained show that after 7 days, geopolymer using Sarawak fly ash has lower
21 compressive strength of about 55 MPa than geopolymer using Gladstone fly ash with strength
22 of about 62 MPa. In comparison with Gladstone fly ash-based geopolymer, it showed that
23 Sarawak fly ash-based geopolymer can be a potential construction material. Moreover, the
24 production of Sarawak fly ash-based geopolymer aids to widen the application of Sarawak fly
25 ash from being treated as industrial waste consequently discharging into the ash pond.

27 **Keywords:** Sarawak fly ash; Gladstone fly ash; Geopolymer; Compressive strength;
28 Morphology

29

30 **1. Introduction**

31 Use of fly ash has been widely researched for making geopolymer concrete, a possible
32 alternative to ordinary Portland cement (OPC) concrete. The term ‘geopolymer’ was
33 introduced by Davidovits in 1979 [1]. Geopolymer has been known to exhibit ceramic-like-
34 properties as it comprises alumino-silicate materials such as fly ash in alkaline environment.

35 High emission levels of CO₂ during the manufacturing of OPC have become an issue since
36 this level of emissions is considered a threat to the existing global climate. The low carbon
37 footprint of geopolymer, which is approximately 80% lower than OPC [2], has shown that
38 geopolymer can be an attractive alternative construction material to OPC. The superior
39 properties of geopolymer such as better acid resistance and long durability have been
40 reported [3]. Geopolymer also shows better performance at elevated temperature as compared
41 to OPC concrete [4].

42 Properties of fly ash can be varied due to its origin, resources or coal type [5]. Fly ash
43 sourced from different places may have different chemical compositions, mineralogy,
44 morphology, particle size distributions or unburned carbon content [6]. Both chemical and
45 physical properties of fly ash can effectively influence the performance of geopolymer.

46 The particle size distribution of fly ash plays an important role on the strength
47 development of geopolymer [7]. Fly ash with higher amount of small particle size has been
48 found to exhibit excellent compressive strength [8]. In the alkali activation, small particles are
49 more active than larger particles [9]. The reactivity of fly ash is proportionally to the particle
50 size smaller than 10µm rather than the particle size greater than 45 µm [10]. The former
51 particle size increases the compressive strength while the latter particle size decreases the
52 compressive strength. It is known that the morphology is affected by fly ash particle size [11].

53 Small particle appears to have smoother surface than large particle [12]. The reduction of
54 particle size improves the workability of the mixture [8]. The formation of irregular grains in
55 fly ash could be due to the incomplete combustion process [11]. Fly ash consists of
56 crystalline phase and amorphous phase has been reported [13, 14]. The amorphous phase of
57 fly ash may be useful for the industry characterisation [14]. The most common phases in fly
58 ash such as quartz and mullite are important to the strength development. However, high
59 content of mullite and quartz decrease the reactivity of fly ash [1].

60 In Sarawak, few coal fired power stations are planned to be constructed within the
61 development masterplan of Sarawak Corridor of Renewable Energy (SCORE). The
62 production of Sarawak fly ash will increase in tandem with the development. Despite being
63 use on the construction of major dams in Sarawak such as Bengoh Dam, Murum Dam and
64 Bakun Dam, most of the Sarawak fly ash is still being treated as industrial waste and dumped
65 into the ash pond nearby. From the environmental point of view, the widening of Sarawak fly
66 ash application can effectively reduce the land for disposal, moreover, lowering the risk of
67 ground contamination due to improper management of discharged fly ash. From the
68 construction material point of view, the development of Sarawak fly ash-based geopolymer
69 provides an alternative to the ordinary Portland cement-based concrete. The world's first
70 building using geopolymer for structural purpose in Australia has proven the potential use of
71 geopolymer [15]. Therefore, it shows the significance of benchmarking of Sarawak fly ash
72 against the well-developed Gladstone fly ash-based for producing geopolymer.

73 In this paper, two different types of fly ash, namely Sarawak fly ash (SFA) from Malaysia
74 and Gladstone fly ash (GFA) from Australia have been used to fabricate geopolymer. Study
75 of geopolymer using GFA has been well reported by other researchers [16-20]. As the
76 research around SFA is very limited, the study of geopolymer made of SFA in comparison to
77 GFA can effectively identify the factors influencing the difference of their compressive

78 strength for future in-depth studies on geopolymer using SFA. It is postulated that the
79 geopolymers produced by GFA and SFA may behave very differently in their respective
80 strength capabilities.

81

82 **2. Background of the Fly Ashes**

83 *2.1. Sarawak fly ash (Malaysia)*

84 In Sarawak, Sejingkat Power Station and Mukah Power Station are the two main coal-
85 fired power stations that are used to generate electricity. Sejingkat Power Station is located in
86 the Sarawak State capital of Kuching in Malaysia. The coal is mainly supplied from the
87 hinterland. Approximately 1 million tons of coal is utilized for the combustion and the daily
88 production of coal ashes is estimated about 1,400 tons. The combustion of coal is carried out
89 in 2 boilers operating independently. The first or older boiler has a capacity to generate 2
90 units x 50 MW of electricity, while the second or newer boiler has a capacity to generate
91 slightly more electricity at 2 units x 55 MW. The combustion temperature of coal is set at 540
92 °C. The cooling system utilized water pumped from river and the flow rate of the cooling
93 system is about 10,275 m³/hr. The by-product of the power plant, fly ash, is efficiently
94 captured by the electrostatic precipitator (approximately 99 %) and only 1 % of the fly ash is
95 discharged to the environment through the 120 m chimney. The captured fly ash is disposed
96 into two ash ponds nearby. The coal used for the combustion is classified as sub-bituminous.
97 The geological age of the coal ranges from Miocene to Miocene-Pliocene. In this research,
98 the Sarawak fly ash is sourced from Sejingkat Power Station.

99 *2.2. Gladstone fly ash (Australia)*

100 Another fly ash used in this research is the Gladstone fly ash. GFA is sourced from
101 Gladstone Power Station. This power station is situated at Gladstone, Queensland, Australia.
102 It is the largest power station in Queensland, which generates electricity of 1,680 MW by 6

103 boilers. The combustion temperature of coal is 540 °C. The cooling water is pumped from
 104 Auckland inlet and the flow rate of the cooling system is about 51,840 m³/hr [21]. It is
 105 estimated that 4 million tons of coal is used for the combustion annually [22]. The coal is
 106 supplied from the mining field at Central Queensland. In this region, the coal is classified as
 107 bituminous coal with the coal age ranged from Permian, Triassic, Jurassic and Cretaceous
 108 [23]. This fly ash is chosen for comparison because there are already a relatively large
 109 number of publications associated to GFA and hence, it would be easier to develop
 110 benchmarks against GFA [24].

111

112 3. Experimental works

113 3.1. Materials

114 The chemical composition of both fly ash types was studied using WD-X-ray
 115 Fluorescence Spectrometer (WD-XRF) and the results are shown in **Table 1**. Both SFA and
 116 GFA are classified as Class F in accordance to the ASTM C618 standard [25].

117 **Table 1** Chemical Composition of Fly Ash

Elements (%)	Gladstone Fly Ash (GFA)	Sarawak Fly Ash (SFA)	Manganese Slag[26]	Ordinary Portland cement (OPC)[27]
SiO ₂	51.1	43.8	28.3	21.8
Al ₂ O ₃	25.7	18.1	10.5	5.8
Fe ₂ O ₃	12.5	7.7	0.3	3.3
CaO	4.3	3.9	11.0	63.0
MgO	1.5	0.5	14.9	2.0
MnO	0.2	22.8	26.0	-
K ₂ O	0.7	2.0	5.1	0.3
Na ₂ O	0.8	0.3	2.7	0.5
SO ₃	0.2	0.1	-	2.4
TiO ₂	1.3	0.6	-	-
P ₂ O ₅	0.9	0.1	-	-

LOI	0.6	0.5	-	1.0
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118

119 From the vision observation, SFA is darker in shade (*i.e.* grey color) when compared to
 120 GFA (*i.e.* brownish) as shown in **Fig. 1**. It has been observed that fly ash with lighter in shade
 121 may consists of finer particle size [7] and produced from anthracite or bituminous coal. It
 122 may be an indicator for better quality of the fly ash.



123

124 **Fig. 1.** Physical appearance of (a) GFA-brownish and (b) SFA-grey colour

125 From the geological point of view, the coal used to produce SFA (*i.e.* sub-bituminous) is
 126 geologically younger (Balingian Formation of late Miocene age – Begrih Formation of Early
 127 Pliocene age) and it is mined nearer to the ground surface compared to the coal used to
 128 produce GFA (*i.e.* bituminous) (Permian age – Cretaceous age). The quality of coal is in the
 129 increasing order from subbituminous to bituminous.

130 The basicity index and hydration modulus of both SFA and GFA are evaluated using the
 131 equations as given in Equation (1) and (2):

132
$$K_b = \frac{CaO+MgO}{SiO_2+Al_2O_3} \quad (1)$$

133
$$HM = \frac{CaO+MgO + Al_2O_3}{SiO_2} \quad (2)$$

134 The basicity index of GFA and SFA is 0.08 and 0.07, respectively. Both are considered
 135 acidic (≤ 1). The acidic character of the fly ashes shows some pozzolanic activity due to the

136 high presence of SiO_2 [28]. The hydraulicity of acidic fly ash in the presence of alkaline
137 solution is weaker in comparison to basic fly ash.

138 From the hydration moduli obtained, it implies the hydraulicity of both fly ashes is very
139 low. The self-cementing properties are very poor due to the lack of CaO content. Being
140 different from Portland cement and Class C type fly ash, it does not harden in the presence of
141 water. Consequently, alkaline activator is essentially needed to activate both fly ashes.

142 *3.2. Sample Preparation*

143 In this paper, the alkaline solution used for the experiment was 8M sodium hydroxide
144 (NaOH) solution, which was prepared from the NaOH pellet with 97% purity, and sodium
145 silicate (Na_2SiO_3) with $\text{Na}_2\text{O}=14.7\%$ and $\text{SiO}_2=29.4\%$. Washed sand was selected as
146 aggregate in the experiments. Two kinds of sand conditions were prepared: (a) without
147 saturated surface dry (SSD) condition and (b) with SSD condition in accordance to the
148 Australian standard AS1141.5 [29].

149 The original moisture of sand at room temperature was 9%. However, the water absorption
150 of the sand was only 1% and it is thought that this amount of water would be absorbed into
151 the sand particles, namely absorbed water. Therefore, sample with and without SSD
152 condition would be differed by moisture of 8%. This moisture level is believed to have
153 contributed to the free moisture on the surfaces of the sand particles. From vision observation,
154 the free moisture is therefore making the appearances of the sand particles shiny. As the
155 absorbed water is not considered when the liquid/ash ratio is evaluated, the sample with and
156 without SSD condition would be differed by the free moisture. The effect of SSD on
157 compressive strength is being investigated here-in-after.

158 The materials were mixed according to proportions shown in **Table 2**. The ratio of fly ash
159 to sand used for these series of experiments was 1: 2 by mass ratio, whereas the ratio of
160 $\text{Na}_2\text{SiO}_3/ \text{NaOH}$ was 2.5 by mass ratio. Different ratios of liquid to ash were used on GFA

161 and SFA in order to achieve similar workability. It is found that the mixture becomes very
162 viscous and difficult to flow when the ratio of liquid to ash on SFA is similar as for GFA.

163 Fly ash and sand was premixed using a mortar mixer. Then, Na_2SiO_3 and NaOH solutions
164 were added into the mixture and further mixing took place for another 5 minutes. The mixture
165 was casted into cube moulds measuring 50 mm \times 50 mm \times 50 mm. The moulds were
166 vibrated on the vibrating table to dissipate air bubbles from within samples. The moulds were
167 then sealed with plastic sheet and placed into the curing box to be heat-cured in the oven at
168 60°C for 24 hours. After 24 hours of curing, the samples were taken out from the oven and
169 demoulded. The samples were then tested for compressive strength (*i.e.* strength after 1 day).
170 The remaining samples were cured at room temperature until the date of strength test (*i.e.*
171 strength after 7 days).

172 3.3. Tests

173 X-Ray Diffraction (XRD) test was carried out using (Bruker AXS D8 Advance XRD) to
174 determine the phase component of the tested ashes. The test was operated with CuK_α
175 radiation at a scanning rate of 0.02 time per step over a 2θ range scanned from 10 to 90 steps.
176 The results obtained from this test were qualitative in nature.

177 The particle size distributions (PSD) of the fly ashes were obtained using Laser Particle
178 Size Analyser (CILAS 1190). The morphology of the fly ashes was studied using the
179 Scanning Electron Microscopy (SEM) (ZEISS SUPRA 40 VP SEM).

180 Loss on Ignition (LOI) test was conducted using the Muffle Furnace. The fly ash was
181 heated in the furnace at 1100 °C with temperature increase rate of 3.33 °C/min. Subsequently,
182 the temperature was decreased to 110 °C and samples were cooled in the furnace for 2 hours.
183 The weight of the sample was measured before placing into the furnace and after the test
184 finished.

185 **Table 2** Details of geopolymer samples

Group	Sample	Ash/ sand ratio	Na ₂ SiO ₃ /NaOH ratio	Liquid/ash ratio
Sand without	GFA	1:2	2.5	0.51
SSD	SFA	1:2	2.5	0.61
Sand with	GFA	1:2	2.5	0.35
SSD	SFA	1:2	2.5	0.45

186 Note: Sample of sand with and without SSD condition differs by free moisture of 8%.

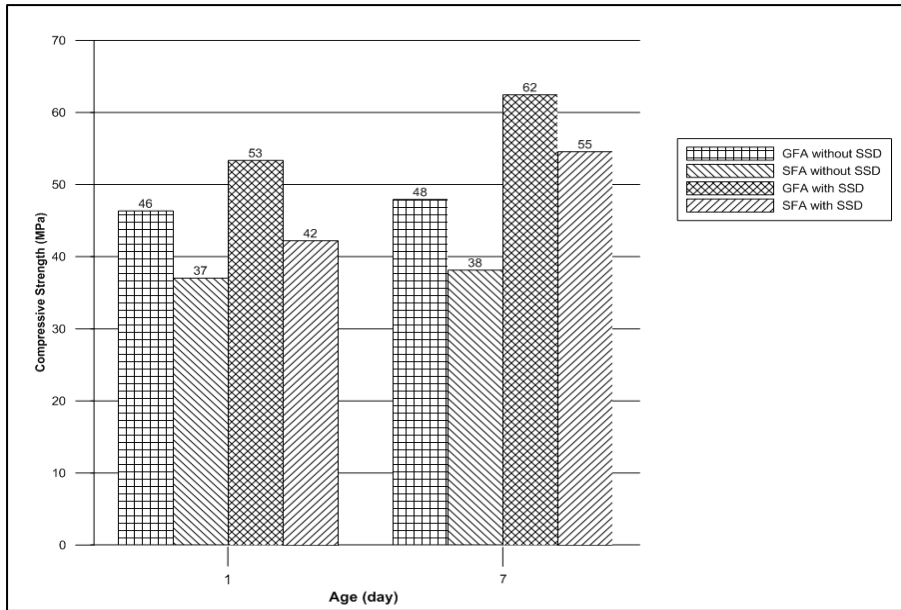
187 The workability of the sample was measured using the flow table in accordance to the
188 ASTM-C1537 standard [30]. The compressive strength test was carried out in accordance to
189 the ASTM-C109/C109M standard [31]. Three samples from each mix were tested for the
190 strength and the average value was reported as the compressive strength of the corresponding
191 mixture.

192

193 **4. Results and Discussions**

194 *4.1. Compressive Strength*

195 The compressive strengths of geopolymer using GFA and SFA were determined after 1
196 day and 7 days of curing as shown in **Fig. 2**. The test results show that the compressive
197 strength of GFA geopolymer was generally higher than SFA geopolymer. The highest
198 compressive strength was obtained on GFA samples after 7 days (*i.e.* 62 MPa) and SFA
199 samples after 7 days (*i.e.* 55 MPa). Factors affecting the strength of SFA and GFA
200 geopolymers are discussed in the following section.



201

202 **Fig. 2.** Compressive strength of geopolymer using GFA and SFA

203 *4.2. Chemical Composition*

204 As presented in **Table 1**, the chemical compositions of GFA and SFA are rather similar
 205 but different in proportions. The major components of both GFA and SFA are SiO₂, Al₂O₃
 206 and Fe₂O₃, followed by CaO, MgO and K₂O. Other components present in small quantities.
 207 Basically, all the elements on GFA are higher than SFA with the exception of K₂O and MnO.

208 It is important to note that SFA contains relatively large quantities of MnO. As compared
 209 to the other fly ashes or OPC, MnO content in SFA is similar to manganese slag as presented
 210 in **Table 1**. The influence of MnO content on the quality of fly ash is assessed with regard to
 211 the quality index as given in Equation (1):

212
$$I_a = \frac{CaO + 0.5MgO + Al_2O_3 + CaS}{SiO_2 + MnO} \quad (1)$$

213 The large quantities of MnO content in SFA significantly reduce the quality of fly ash by 2
 214 times as compared to GFA, *i.e.* 0.6 for GFA and 0.3 for SFA. High amount of MnO content
 215 reduces the geopolymer strength as it has poorer hydraulicity and high porous structure. It
 216 also inhibits the early strength development [32]. The reason of high MnO content can be
 217 most likely attributed to the existence of pyrite and illite minerals bound to the original coal

218 and should be removed during pre-combustion coal cleaning [33]. The Mn compound is
219 predominantly deposited in larger fly ash particle size and the amount increases with the
220 particle size. The particle size distribution of SFA as discussed in the following section is
221 shown to agree well to this statement.

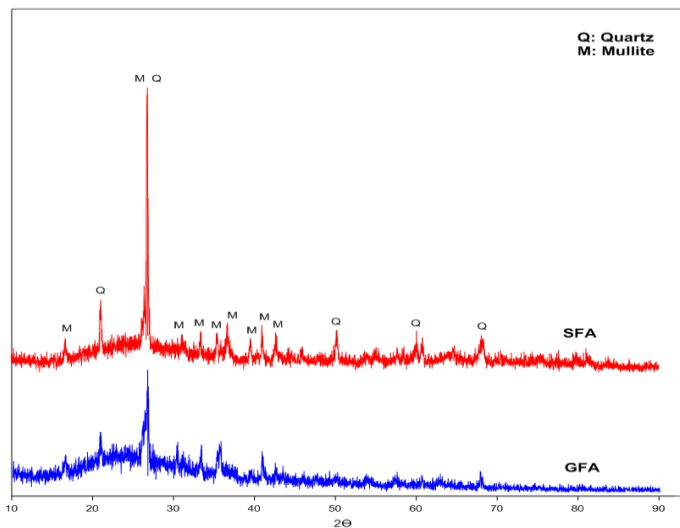
222 High amount of unburned carbon content hinder the geopolymerisation reaction. Besides,
223 it may affect the workability and strength capability [5]. However, the LOI values of GFA
224 and SFA in this research were found to be 0.57% and 0.49%, respectively. The results
225 indicate that both fly ash types have very low unburnt carbon content and hence, the effect of
226 LOI on geopolymer strength can be neglected.

227 *4.3. Mineralogy*

228 The mineralogy of GFA and SFA was studied based on the XRD test results. The peaks
229 represent the presence of the crystalline phases. The XRD result of GFA is similar to the
230 corresponding XRD result of SFA as shown in **Fig. 3**. The dominant minerals observed in
231 both GFA and SFA are mullite and quartz. The highest peak for both GFA and SFA was
232 observed at approximately 26.5° at 2θ . This broad hump indicated that the presence of
233 greater amount of amorphous material. However, the identification of crystalline peaks on
234 amorphous materials can be difficult due to overlapping some of the peaks [28]. The MnO
235 content in SFA is believed to be amorphous-like instead of crystalline-like thus making it
236 difficult to be identified from the XRD result.

237 The XRD pattern of GFA shows wider band from 20° to 40° . This shows that the
238 amorphous nature on GFA is more prominent than SFA. It can be attributed to the instant
239 cooling after the high temperature of coal combustion which causes the conversion of
240 material phase from crystalline to amorphous [34]. As provided in the background of both
241 coal fired power plant, the combustion temperature at both power plant is 540°C ; however,
242 the flow rate of the cooling system at Gladstone Power Plant is 5 times faster than at

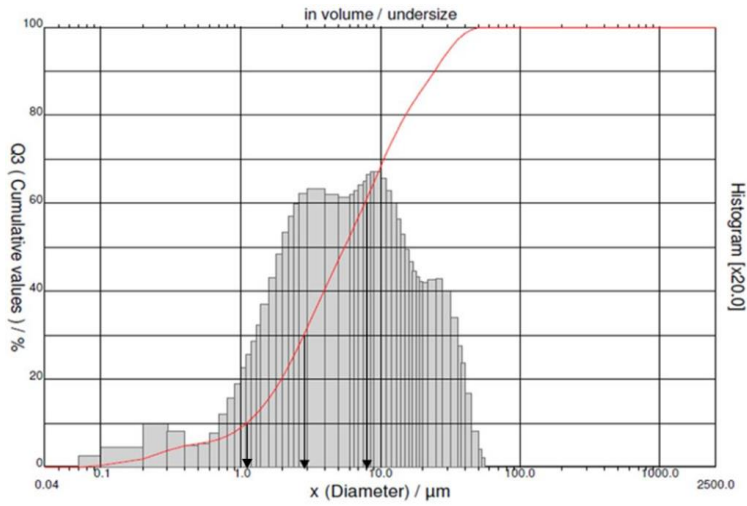
243 Sejngkat Power Plant. The mineral matter is quenched into the particle after the combustion
244 of coal and therefore, the particle tends to behave more spherical to rounded shape with
245 glassy exterior surface [35]. The greater flow rate ensures the combustion is cooled at a faster
246 rate thus the temperature gradient is greater. This quenching associates with the instant
247 cooling, controls the growth and the agglomeration of particles. More amorphous content can
248 be quenched into the fly ash particles and thus, as a result of better amorphous nature of fly
249 ash. Consequently, GFA shows richer amorphous phase than SFA and enhances the strength
250 development. More amorphous silica and alumina can be dissolved from GFA and contribute
251 to the geopolymerisation and therefore, form stronger geopolymer structure.



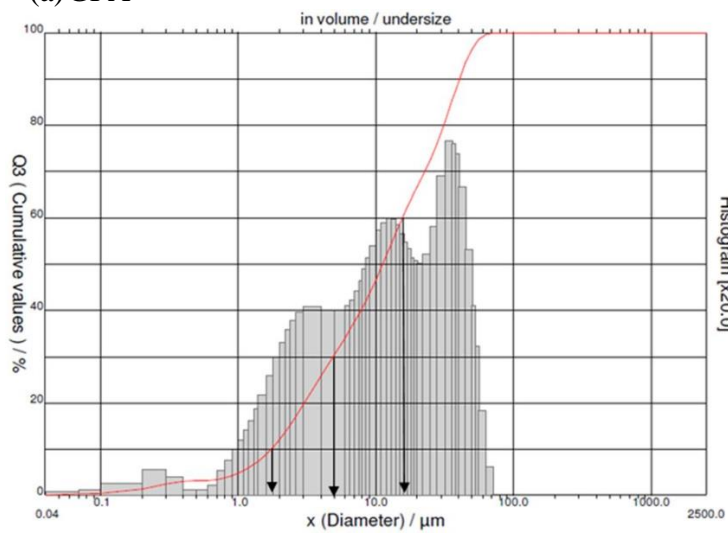
252
253 **Fig. 3.** Result of X-ray diffraction (XRD) on GFA and SFA

254 4.4. Particle Size Distribution

255 The results of the particle size distribution on GFA and SFA are presented in **Fig. 4. Table**
256 **3** shows the test results of particle diameter at 10% (d_{10}), 50% (d_{50}) and 90% (d_{90}), and the
257 mean diameter. Both GFA and SFA show broad distribution pattern in their respective
258 particle size distribution plots. However, GFA particles are observed to be approximately two
259 times smaller than SFA. For GFA, it is predominantly smaller than 24 μm whereas SFA is
260 predominantly smaller than 40 μm , both comprising 90% of the total fly ashes. The mean
261 particle diameter of GFA is 9.3 μm whereas SFA is 16 μm .



(a)GFA



(b)SFA

Fig. 4. Particle size distribution of (a) GFA and (b) SFA

262

263 **Table 3** Diameter of GFA and SFA particles at 10%, 50% and 90% of the total fly ash

264 content; mean diameter and specific surface

Sample	GFA	SFA
Diameter at 10% (d_{10})	1.1 μm	1.8 μm
Diameter at 50% (d_{50})	5.5 μm	11 μm
Diameter at 90% (d_{90})	24 μm	40 μm
Mean diameter	9.3 μm	16 μm
Specific surface (cm^2/g)	31382	24920

265 The particle size distribution of both GFA and SFA is categorized into 4 groups as
 266 presented in **Table 4** to ease the comparison, namely (a) ultrafine: < 0.6 μm (b) fine: 0.6 to 6
 267 μm (c) coarse: 6 to 20 μm and (d) coarser: > 20 μm .

268 **Table 4** Particle size distribution

Mode	Particle Size Group (μm)	GFA (%)	SFA (%)
Ultrafine	< 0.6	6	4
Fine	0.6 – 6	47	30
Coarse	6 – 20	32	31
Larger Coarse	> 20	15	35
	< 10	68	46
	> 45	2	7

269 Most of GFA has distributed in the range of fine portion but it gradually decreases from
 270 fine portion (47%), coarse portion (32%) to coarser portion (15%). For SFA, the particle size
 271 has evenly distributed among fine portion (30%), coarse portion (31%) and coarser portion
 272 (35%). The highest percentage of particle size distribution is found in the coarser portion. For
 273 both GFA and SFA, lesser amount of particle sizes is observed in the ultrafine portion at 6%
 274 and 4% respectively. This could be due to the difficulty of capturing such fine particle sizes
 275 and mostly escape to the environment through the chimney. Nevertheless, the amount of this
 276 particle size in the overall fly ash particle is insignificant.

277 As the fine portion is found to be highest on GFA, this fine portion of particles plays an
 278 important role on the strength development and the rate of geopolymerisation. Smaller
 279 particles are more active in the alkali activation process thus leading to higher compressive
 280 strength [9]. As most of GFA particles are distributed in fine portion, higher specific surface
 281 enhances the reactivity of the particle and hence, provides larger active surface area for the
 282 reaction. The higher fine portion on GFA also explains why lesser liquid content is required
 283 for GFA to achieve similar workability as SFA because smaller particle size improves the
 284 flow of the mixture.

285 The Coefficient of Uniformity (C_u) and Coefficient of Curvature (C_c) were also used to
286 compare the uniformity of particle sizes and the gradation of the particle size distribution
287 curve for GFA and SFA as given in Equations (4) and (5).

$$288 \quad C_u = \frac{d_{60}}{d_{10}} \quad (4)$$

$$289 \quad C_c = \frac{d_{30}^2}{d_{60} \times d_{10}} \quad (5)$$

290 It was found that the C_u for GFA is 7.3, which is lower than SFA with a value of 9.4.
291 Higher C_u value represents the particle size distribution being more non-uniform and contains
292 wider ranges of particle sizes. However, the higher amount of fine particles in GFA enhances
293 the filler effect. This in turn suggests that GFA has capability of more closely packed
294 particles than SFA and therefore, lesser voids and higher strength potential. The structure
295 formed tends to be more compact and rigid.

296 C_c value indicates the degree of curvature of the particle size distribution plot. Both GFA
297 and SFA have a C_c value of approximately 1, which is within the range of 1 to 3. Therefore,
298 both fly ash particle size distributions are classified as well-graded.

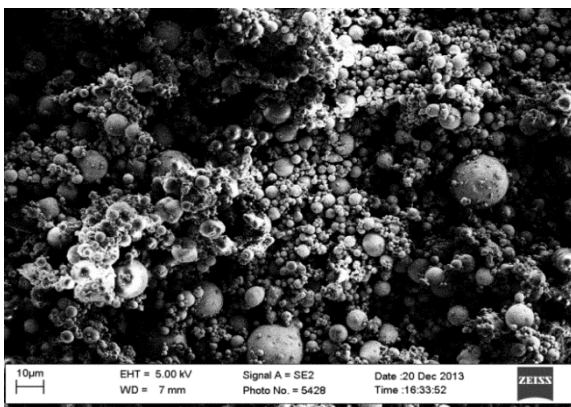
299 *4.5. Scanning Electron Microscopy*

300 As shown in the SEM micrographs, the fly ash particles of GFA are observed to be
301 predominantly smaller than SFA (see **Fig. 5a and b**). The distribution of both types of fly ash
302 are generally heterogenous and non-uniform as observed under the microscope. Distinct
303 distribution of particle size between large particles and small particles are obvious on GFA.
304 For SFA, the particle sizes are evenly distributed. This observation is consistent to the results
305 of the PSD.

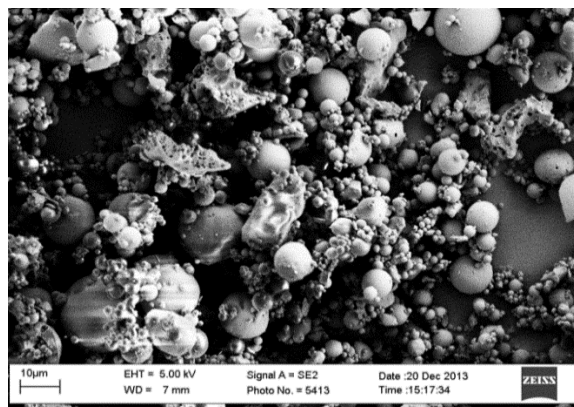
306 The morphology of ultrafine and fine particles are basically presented in smooth spheres
307 and they are rounded in nature [12]. GFA has higher percentage of ultrafine and fine portion,
308 which explains its morphology consisting of smooth spherical particles with less irregular
309 grains (see **Fig. 5c**). Fly ash with smooth spherical particle shape enhances the ball bearing

310 and lubricant effects. Having this capability, it improves the workability and flow ability of
311 the mixture. Therefore, less liquid content is required for GFA to achieve similar workability
312 as SFA. Inversely, higher percentages of coarse particles exhibit particle with shape
313 irregularities. Therefore, this explains the reason why SFA is observed to be less rounded in
314 shape and appear with some angularities (see **Fig. 5d**). Moreover, SFA consists of irregular
315 grains in which some are greater in size than those spherical particles (see **Figs. 5e and f**).
316 These irregular grains could be formed by the incomplete combustion as most of the organic
317 matters should be volatilized during the coal combustion process.[11] Additionally,
318 agglomeration of fly ash particles is observed on SFA particles from the microstructure
319 image.

320 From the workability point of view, the coarse particles of SFA with less rounded particle
321 shape and angularity increases the internal friction. The irregular grains occupy high surface
322 area which increases the sorption ability and the liquid demand. Therefore, higher amount of
323 liquid solution is required to obtain the similar workability as GFA. The angularity of particle
324 shape also affects the orientation of particles in the geopolymer matrix consequently
325 increases the void between the particles. Therefore, it could in turn reduce the strength
326 development on SFA. The reduction of coarse particle size on SFA is suggested and may
327 possibly increase the geopolymer strength.

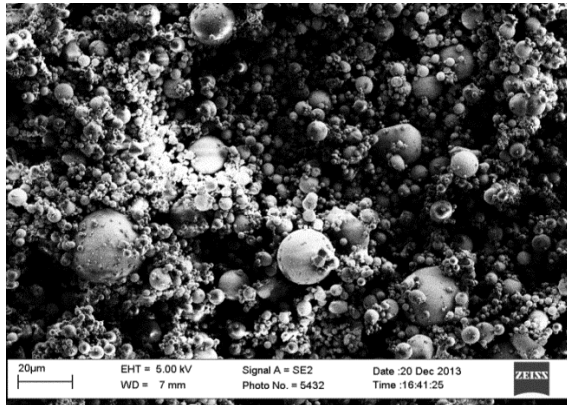


(a) GFA- distribution from fine to coarser portions is in decreasing order

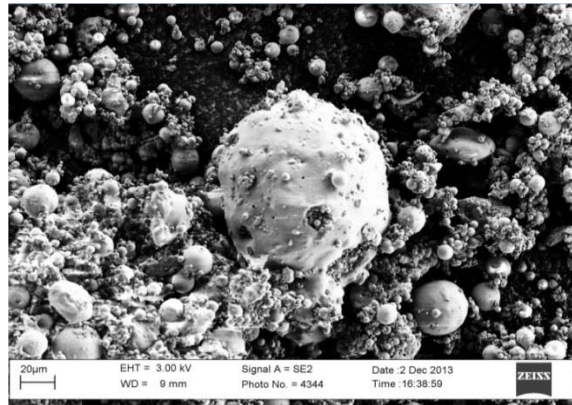


(b) SFA- distribution from fine to coarser portions is evenly (magnification to 10µm)

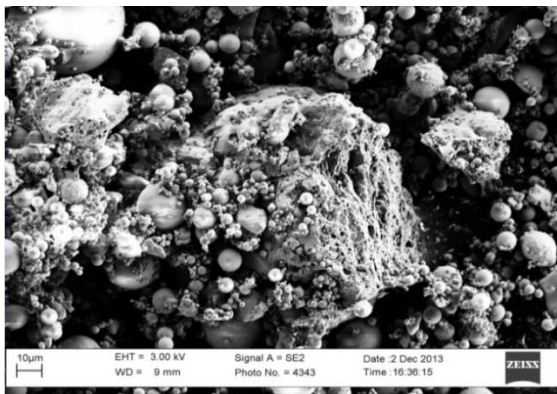
(magnification to 10 μ m)



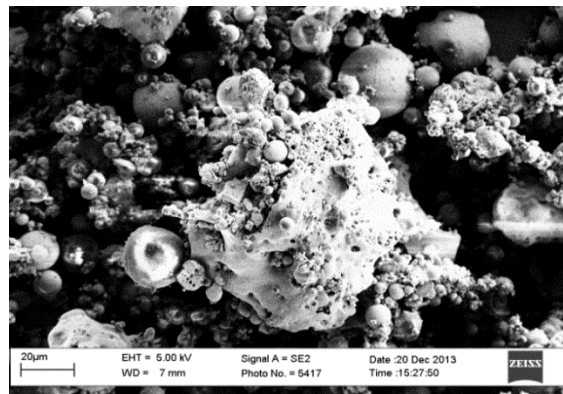
(c) GFA- smooth sphere and rounded in shape (magnification to 20 μ m)



(d) SFA- less rounded in shape and appear with some angularities (magnification to 20 μ m)



(e) SFA- irregular grain (magnification to 10 μ m)



(f) SFA- irregular grains in which some are greater in size than those spherical particles (magnification to 20 μ m)

328 **Fig. 5.** SEM on GFA and SFA

329 *4.6. Liquid to ash ratio*

330 It is observed that SFA requires higher ratio of liquid to ash but the compressive strength
331 obtained is lower than GFA. Although higher amount of alkaline solution should be able to
332 leach more silica and alumina from the fly ash and consequently enhances the
333 geopolymerisation process, it is not the case as observed on SFA performance. It could be
334 due to higher usage of alkaline solution that may obstruct the water evaporation and the
335 structure formation [36]. Other than that, fly ash with mostly amorphous phase enhances the
336 leaching capability of SiO₂ and Al₂O₃ [37]. It is believed that the better amorphous phase of

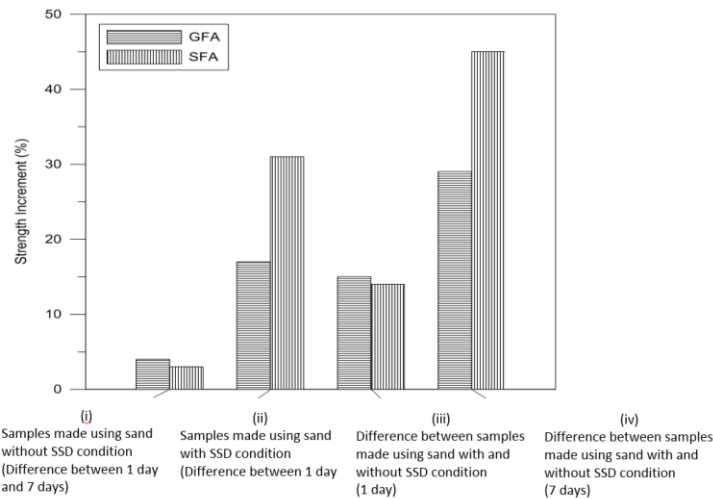
337 GFA than SFA demonstrates greater leaching capability and enhances the geopolymerisation
338 between the alkaline solution and fly ash, thus increasing the compressive strength.

339 *4.7. Sand with SSD condition*

340 It is worth mentioning that the effect of sand condition on geopolymer strength is
341 significant as shown in **Fig. 6**. For samples using sand without SSD condition, the strength
342 increments from 1 day to 7 days were 4% and 3% for GFA and SFA respectively. As free
343 moisture exists in sand without SSD condition, it weakens the concentration of alkaline
344 solution. Additionally, this free moisture is not involved in the chemical reaction of
345 geopolymer because the geopolymerisation is activated by the alkaline solution, unlike
346 cement the strength development is built up through water hydration. Therefore, it is believed
347 that the free moisture in the samples have evaporated when subjected to heat drying in the
348 oven. This will subsequently weaken the matrix formed and cause a decrease in strength.

349 The strength increments of samples using sand with SSD condition after 1 day to 7 days
350 were 17% for GFA and 31% for SFA. As free moisture is eliminated from sand with SSD
351 condition, the actual concentration of alkaline solution is completely used for the
352 geopolymerisation thus higher compressive strength can be obtained.

353 The strength increments for samples using sand with SSD condition from those samples
354 without SSD condition after 1 day were 15% and 14% for GFA and SFA samples whereas it
355 was found to be 29% and 45% after 7 days respectively. The strength increment of SFA
356 samples was approximately 16% higher than GFA sample after 7 days.



357

358 **Fig. 6.** Strength increment of geopolymer using sand with and without SSD condition

359 Both compressive strengths of GFA and SFA have been observed to increase with curing
 360 age. This observation could be due to some unreacted fly ash particles, which earlier on did
 361 not manage to undergo geopolymerisation during heat curing in oven that continued to react
 362 with the alkaline solution when being cured at room temperature. Also, it could be due to the
 363 reaction on the reactive fine particle size of fly ash which consequently improved the bonds
 364 in geopolymer over the curing age [38].

365 The strength increments of SFA over the curing age were observed to be 14% higher than
 366 GFA. This suggested that the rate of geopolymerisation for SFA samples was initially slower
 367 and mainly developed its strength with the curing age. SFA which has relatively larger
 368 particle size may need longer period for dissolution of fly ash particles to build up the
 369 strength. Therefore, better strength development was observed at later stage.

370

371 **5. Conclusions**

372 Fly ash from Sarawak (SFA) was used to assess its potential in making geopolymer and
 373 the results were compared with Gladstone fly ash (GFA) from Australia. The results obtained
 374 from the experiments showed that SFA-based geopolymer developed lower compressive

375 strength (55 MPa) than geopolymer using GFA (62 MPa) after 7 days. The factors affecting
376 SFA geopolymer obtained lower compressive strength than GFA are listed as below:

- 377 1. SFA and GFA behave very differently in nature. Besides the different color (brownish on
378 GFA whereas grey on SFA), all the chemical elements on GFA are higher than SFA with
379 the exception of K_2O and MnO . The large quantities of MnO content on SFA reduce the
380 quality index of fly ash by 2 times as compared to GFA, *i.e.* 0.6 for GFA and 0.3 for
381 SFA.
- 382 2. The effect of LOI on geopolymer strength can be neglected due to low LOI content on
383 both fly ashes (<1%).
- 384 3. GFA shows better amorphous phase than SFA. The faster flow rate of cooling system at
385 Gladstone Power Plant than Sejingkat Power Station (approx. 5 times) enhances the
386 amorphous nature of GFA.
- 387 4. The PSD of GFA is predominantly smaller than $24\ \mu m$ whereas SFA is predominantly
388 smaller than $40\ \mu m$, both comprising 90% of the total fly ashes. The finer particles on
389 GFA have better ability to have more closely packed particles to enhance the filler effect
390 and better reactivity. It also acts as 'nucleation sites' to develop strength.
- 391 5. The morphology studies show that SFA has been observed to be less rounded in shape
392 and appear with some angularities whereas GFA has been observed to consist mainly of
393 smooth spherical particles and less irregular grains. The former type increases the liquid
394 demand for workability but reduces the strength development; inversely, the latter type
395 increases the ball bearing and lubricant effect for workability also enhancing the strength
396 development.
- 397 6. SFA samples have higher strength increment of approx. 14% than GFA samples over the
398 curing age. It has suggested that the geopolymerisation of SFA was initially slower and
399 the strength development was built up at later stage.

400 This paper shows that Sarawak fly ash can be a potential construction material for
401 producing Sarawak fly ash-based geopolymer. With benchmarking against the well-
402 developed Gladstone fly ash-based geopolymer, factors that influence the strength capability
403 of Sarawak fly ash-based geopolymer can be effectively improved to achieve the desired
404 strength performance.

405

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414

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486

487 **Highlights**

- 488 • Geopolymer produces lower carbon footprint and this leads to cleaner production.
- 489 • Strength of geopolymer made using fly ash from Malaysia and Australia are studied.
- 490 • Both types of fly ash vary in chemical contents, mineral phase and particle size.
- 491 • Geopolymer made using Malaysia fly ash show lower strength but has great potential.