

Influence of different monomer ratios and recycled concrete aggregate on mechanical properties and durability of geopolymer concretes

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Abstract

Properties of geopolymer concrete using metakaolin (MK) as the aluminosilicate source and recycled concrete aggregate (RCA) as partial replacement of natural aggregate are presented in this paper. The effects of sodium silicate (SS) to sodium hydroxide (SH) ratios, and RCA in different percentages on the mechanical and durability properties of geopolymer concrete were determined. Microstructural changes of geopolymers as a result of using RCA were evaluated via scanning electron microscopic (SEM) images. Test results showed that compressive strength of geopolymer concrete improved with increasing the SS/SH ratio. Although the use of RCA reduced compressive strength by up to 28%, the strength was still high enough for structural applications. In addition, increasing the SS/SH ratio reduced the chloride ion permeability and absorption of geopolymer concrete. The morphology results showed that the debonding width at the interfacial transition zone (ITZ) between RCA and binder decreased with the increase of SS/SH ratio. The polymer products in the proximity of adequate monomer ratios became more uniform and

27 homogenous. Results also showed that the binder with a SS/SH ratio of 3 exhibited higher density
28 and less porosity than that with smaller ratios. Therefore, the recycled construction and demolition
29 waste can significantly contribute to the sustainability of construction industry from technical,
30 economic and environmental points of view.

31

32 **Keywords:** Geopolymer Concrete; Alkali Activator Ratio; Recycled Concrete Aggregate;
33 Compressive Strength; Durability; Microstructure Properties.

34

35 **1. Introduction**

36 Currently, concrete is the most used building material because of its low price, availability of raw
37 materials and suitable mechanical and durability properties for various applications [1]. The
38 demand for concrete and cement-based materials is increasing with the growing trend of
39 industrialization and population growth. Consequently, concrete industry faces two serious
40 concerns: a) uncontrolled harvesting of natural resources, for instance rock mines, and b) pollution
41 and wastes derived out of the activities of cement plants. The process of cement production is
42 associated with emissions of greenhouse gases which are harmful to people and the nature. It is
43 estimated that about 6% of the total CO₂ emission is produced by cement industry. With the current
44 unfavorably increasing trend, its value will rise to more than 7% [1, 2]. This is why numerous
45 studies have been carried out over the last decades on development of suitable cement substitutes
46 and solutions to reduce the use of cement [3]. Various cement replacing materials are currently
47 used in production of concrete [4-6]. These materials are divided into two main groups. The first
48 group is natural pozzolans, which are a combination of silica/silica-alumina based- amorphous
49 glasses and mainly derived from volcanic magma material [7]. The second group is organic

50 materials, which usually are industrial by-products such as metakaolin, fly ash, slag, red mud and
51 rice husk ash [8-10]. In this regard, over the recent years, alkali activated binder or geopolymer
52 has been introduced as a substitute of cement.

53 Geopolymer concrete with high performance has been proposed as an eco-friendly material to
54 substitute conventional cement concrete [11]. Alkali activation of a wide range of industrial
55 wastes, such as pozzolans and other materials that contain non-crystalline (amorphous)
56 aluminosilicate can transform into geopolymer [12]. Theoretically, any material that contains silica
57 and aluminum in its composition can be activated by alkali and used as solid raw material in
58 geopolymerization technology [12, 13].

59 Geopolymer is produced by mixing materials containing silica and aluminum with a silicate alkali
60 solvent. The silicate alkali solvent interrupts the bonds of aluminum oxide and silica oxide in the
61 aluminosilicate powder. After dissolving and activating the Si and Al particles in a rapid
62 polymerization reaction, the main inorganic polymer chain is formed [14, 15]. The alkali-silicate
63 activator solution contains an alkaline hydroxide solution such as sodium hydroxide (NaOH),
64 potassium hydroxide (KOH), calcium hydroxide (Ca(OH)₂) alone, or a combination of these
65 solutions, including a silicate solution such as sodium silicate (Na₂SiO₃) or potassium silicate
66 (K₂SiO) [16].

67 Various studies have been conducted to investigate the properties of geopolymer concrete [17, 18].
68 Previous researchers [19, 20] reported that increasing the specific area of aluminosilicate source,
69 such as metakaolin (MK) improves properties of the product due to increased degree of
70 geopolymerization or reactivity. Nguyen et al. [21] showed that although MK-based geopolymer
71 has higher cost in North America because of limited availability of MK and the associated
72 transportation cost, it could be competitive with ordinary Portland cement in places the raw

73 material is available in more abundance. Additionally, it was indicated that the MK-based
74 geopolymer produces 40% less greenhouse gas emissions in comparison to its conventional
75 cement counterpart, which is in agreement with the analysis of McGuire et al. [22] and Davidovits
76 [23]. Moreover, geopolymer may be produced using low-grade calcined clay. However, some
77 studies [24-26] have shown that dissolution of calcium occurs in low alkali when calcium silicate-
78 rich sources are added to geopolymer mixtures and, consequently calcium silicate hydrate (C-S-
79 H) gel is created together with geopolymer gel. Furthermore, not only does the presence of these
80 two gels in the binder phase increase the mechanical strength, but the type and concentration of
81 the activator as well as the ratios in its compounds can also have a significant effect on the
82 mechanical and chemical properties of the geopolymers [27, 28]. It was shown in previous studies
83 that the use of a mixture of sodium hydroxide and sodium silicate as the alkaline activator resulted
84 in higher mechanical strength of geopolymer than using sodium hydroxide only as the activator
85 [29]. Hardjito et al. [30] studied the geopolymer concretes with a compressive strength range of
86 30 MPa to 80 MPa. It was shown that optimum strength was obtained when the water to
87 geopolymer solids ratio was 0.18 and the specimen was cured at 90°C. In another study [31],
88 slag/rice husk ash based geopolymer concretes were evaluated with a compressive strength range
89 of 15 MPa to 72 MPa under three curing conditions of ambient temperature, 60 °C and 90 °C. It
90 was shown that an increase in compressive strength of up to 1.45 and nearly 3 times for black rice
91 husk ash replacement (BRHA) levels of 10% and 20%, respectively could be achieved when the
92 samples were cured at 60°C. They found that BRHA replacement would increase the amount of
93 unreactive silica, which would lead to an inappropriate increase in the ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ in
94 geopolymer mixture and consequently has a negative effect on the compressive strength. In
95 addition, the chloride ion penetration for these mixes was classified as low to moderate.

96 The limitation on useful life of structures and their exposure to natural factors such as earthquakes,
97 storms, corrosive or severe environments and rapid urban development programs cause destruction
98 of structures. Due to these destructions, the amount of construction and demolition (C&D) waste
99 produced from concrete and masonry structures is increasing day-by-day. Efficient management
100 of the C&D wastes has been a significant issue from the environmental perspective. The reuse and
101 recycling of these materials lead to economic efficiency and have positive effect on the
102 environment, which has attracted the interest of researchers and structural engineers [32-34].
103 Following the reduction of using natural resources and construction of environment-friendly
104 concretes, the use recycled concrete aggregates would be a sustainable remedy to reduce the
105 exploitation of natural resources and the negative impacts of concrete industry to the environment.
106 Different amounts of recycling or landfill of these materials have been reported in different
107 countries [35]. In 2004, about 20 million tons of C&D waste was produced in Hong Kong out of
108 which about 88% was recycled. In Europe, nearly 180 million tons of C&D waste is produced
109 annually, however, only 28% of it is recycled [36]. In 2014, about 534 million tons of C&D debris
110 was produced in the United States, which was more than twice the amount of urban solid waste.
111 Contrary to the benefits mentioned above, a small amount of recycled concrete aggregate (RCA)
112 has been used in structural engineering projects in the US. However, it has been accepted that the
113 recycling of old concrete and the conversion of these into materials for structural applications is
114 suitable and cost-effective [37, 38] RCA can be used as substitutes of natural fine and coarse
115 aggregates partially or completely [39]. RCA have mainly higher water absorption and lower
116 strength as compared to natural aggregates. This is because the strength of hardened mortar is
117 usually less than the strength of natural aggregate [40, 41].

118 Nuaklong et al. [42] studied the mechanical properties and durability of the metakaolin-fly ash
119 based geopolymer concrete containing RCA. The results indicated that the partial replacement of
120 metakaolin with fly ash could significantly improve the mechanical properties, abrasion resistance,
121 permeability, and pore structure. Moreover, increasing the metakaolin up to 30% in RCA-
122 geopolymer concrete, resulted in improvement of compressive strength and reduction of porosity
123 and water absorption by up to 134%, 69% and 89%, respectively. Mohseni et al. [38] investigated
124 the engineering properties and microstructure of self-compacting concrete containing RCA. The
125 results showed adequacy of the concrete for structural applications despite the reduction of
126 compressive strength by RCA. It was also shown that the interfacial transition zone (ITZ) between
127 RCA and cement paste was weaker as compared to that in natural aggregate concrete.

128 Additionally, it can be inferred from literature that the cement paste-aggregate interface is the
129 weakest link and often it is the starting point of the development of micro or macro cracks in
130 cementitious composites [3, 43-46]. Although, the ITZ was shown to be denser and more compact
131 in geopolymer concrete [47, 48], it plays a crucial role in concretes utilizing RCA. Increasing
132 mechanical strength usually indicates the increase in density and uniformity of the ITZ at
133 geopolymer binder-aggregate interface, which eventually results in the improvements of bond
134 strength and durability. Zhang et al. [49] showed that the ITZ of fresh potassium (sialates)
135 geopolymer concrete is porous and somewhat non-uniform. However, with the increase of age and
136 increase of hydration reaction, the interface becomes denser by the secondary products of
137 geopolymerization that improves the bond between binder and aggregates.

138 The geopolymer composite, as a relatively new technology, with unique advantages requires more
139 research for better understanding and development for its extensive use in construction industry.

140 On the other hand, it is necessary to propagate and develop applications of recycled materials in
141 order to realize the sustainable development strategy of this new technology.

142 Limited works have evaluated the mechanical properties of geopolymer concrete containing RCA,
143 as discussed above. However, durability and microstructural properties of geopolymer composites
144 incorporating RCA have been reported rarely. The present study seeks to address this gap by using
145 sodium silicate (SS) and sodium hydroxide (SH) in different weight ratios as alkaline liquid to
146 activate the metakaolin. Moreover, RCA was used in three different weight ratios of 10%, 20%
147 and 30% as substitute of natural coarse aggregate. To evaluate the engineering and durability
148 properties of geopolymer concrete composites, compressive strength, water absorption, specific
149 electrical resistivity and chloride ion penetration (RCP) tests were carried out. Microscopic
150 analysis was utilized via scanning electron microscopic (SEM) images to provide a thorough
151 insight to the alterations in the alkali activator's influence and ITZ of aggregate-geopolymer paste.

152

153 **2. Experimental program**

154 **2.1 Materials**

155 In this study, metakaolin was used as a source of aluminosilicate to make geopolymer samples.
156 Table 1 indicates the chemical characteristics of metakaolin. For the manufacturing of geopolymer
157 concrete mixtures, sodium silicate (SS) and sodium hydroxide (SH) solution was used to activate
158 the source material.

159 Three types of aggregates including natural river sand, crushed gravel and demolished concrete
160 were used as fine, coarse and recycled coarse aggregates, respectively. The physical properties of
161 aggregates and the appearance of RCA have been shown in Table 2 and Figure 1-a, respectively.
162 Note that the natural aggregates were used in saturated surface dry (SSD) condition. The particle

163 size distributions of aggregates have been illustrated in Figure 1-b to have a comparison with the
 164 requirements specified in ASTM C33 [50].

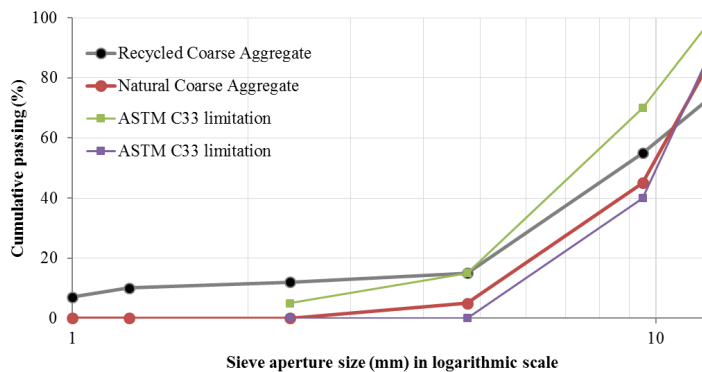
165 Table 1: Chemical composition and physical properties of metakaolin

Component	Mass (%)
SiO ₂	52.1
Al ₂ O ₃	43.8
Fe ₂ O ₃	2.6
CaO	0.2
MgO	0.21
SO ₃	0
K ₂ O	0.32
Na ₂ O	0.11
L.O.I	0.99
Surface area (m ² /kg)	2540
Specific gravity	2.6

166



(a)



(b)

167

168

169 Figure 1. a) Appearance of recycled coarse aggregate; and b) Sieve analysis of recycled and

170

natural aggregate.

171

172 Table 2: Recycled coarse and natural aggregate properties.

Properties measured	Fine aggregate	Natural coarse aggregate	Recycled coarse aggregate
Water absorption (%)	1.2	0.69	4.3
Specific gravity	2.0	2.3	1.9
Maximum particle size (mm)	4.75	12.5	12.5

173

174 2.2. Mix proportion

175 In this study, the binder content for each of the 12 mixtures was considered to be 400 kg/m³. Also,
176 RCA replaced with natural coarse aggregate in three ratios by weight of total aggregates, i.e. 10,
177 20 and 30%. High range water reducer (HRWR) with 1% of the weight of binder materials was
178 also added to the geopolymer concrete mixtures to increase the workability. Three SS/SH ratios,
179 i.e. 2, 2.5 and 3 were chosen to evaluate the effect of activator ratios on geopolymer concrete
180 containing recycled aggregates. Mix designs of the geopolymer concrete mixtures are shown in
181 Table 3.

182

183 Table 3. Mix proportions of geopolymer concrete (kg/m³).

Mix ID	Metakaolin	SS ^b	SH ^c	Aggregates		
				Fine aggregate	NCA	RCA
R2RCA0 ^a	400	90	45	600	1250	0
R2RCA10	400	90	45	600	1025	115
R2RCA20	400	90	45	600	910	230
R2RCA30	400	90	45	600	800	340
R2.5RCA0	400	115	45	600	1250	0
R2.5RCA10	400	115	45	600	1025	115
R2.5RCA20	400	115	45	600	910	230
R2.5RCA30	400	115	45	600	800	340
R3RCA0	400	135	45	600	1250	0
R3RCA10	400	135	45	600	1025	115
R3RCA20	400	135	45	600	910	230
R3RCA30	400	135	45	600	800	340

184 ^a: Numbers after R is SS/SH ratio and after RCA is recycled aggregate percentage; ^b: SS is
185 sodium silicate; ^c: SH is sodium hydroxide (SH).

186

187 **2.3. Mixing and testing procedure**

188 For mixing, dry materials including metakaolin, fine and coarse aggregates mixed together in a
189 mixer for 2 minutes. Then, the prepared alkaline activator solution was added to the pre-
190 metakaolinite to form the required gel and then the mixing continued for about another 3 minutes.
191 Later on, the obtained geopolymer concrete mixture was cast into the molds before hardening. The
192 hardened specimens were demolded after 24 hours and cured in ambient temperature of 21±3°C
193 until the testing day. Figure 2 shows the manufacturing process of geopolymer concrete specimens.
194 After 90 days of ambient curing, the compressive strength and water absorption of the hardened
195 geopolymer concrete was evaluated on 150 mm cubic specimens according to BS EN 12390-3
196 [51] and ASTM C-642 [52], respectively.

197 The electrical resistivity is one of the inherent properties of concrete that depends greatly on the
198 moisture content and pore solution composition of concrete. It was found that, there is a
199 meaningful relationship between the electrical resistance of the concrete and the penetration of the
200 chloride ions [53, 54]. For this purpose, the specific electrical resistivity test was carried out on
201 saturated 150 mm cubic specimens at the age of 90 days. The testing procedure was adopted
202 according to test set up used by the previous studies and ACI committee 222 [55, 56]. Ohm's law
203 was used to evaluate the electrical resistivity. Electrical specific resistivity was calculated using
204 Equation 1:

$$205 \quad \rho = RA/L \quad (1)$$

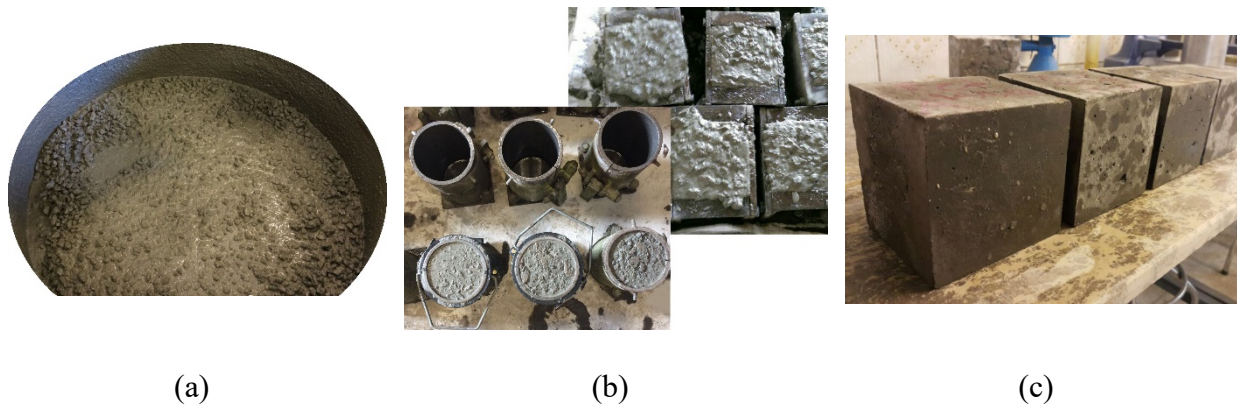
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207 Where, ρ is the specific electrical resistivity ($\Omega.m$), R is the electrical resistivity (Ω), L is the
208 distance between the two electrodes (m) and A is the contact surface area of the electrodes with
209 the specimens (m^2).

210 Rapid chloride permeability (RCP) test is a common method for rapid assessment of the concrete
211 permeability against chloride ion. In this test, the total charge passing through a concrete disc after
212 90 days of ambient curing with 50 mm in thickness and 100 mm in diameter was evaluated in 6
213 hours by 60 volts' potential in accordance with ASTM C1202. Eventually, SEM images were used
214 to study of morphology, microstructure and ITZ of the geopolymer concretes containing RCA.

215 It should be noted that, the presented results are the mean value from at least three identical
216 specimens.

217



218 Figure 2. Manufacturing process of geopolymer concrete specimens; (a) Fresh mixture, (b)
219 Casting, (c) Demolded specimens.

220

221 3. Results and discussion

222 3.1 Compressive strength

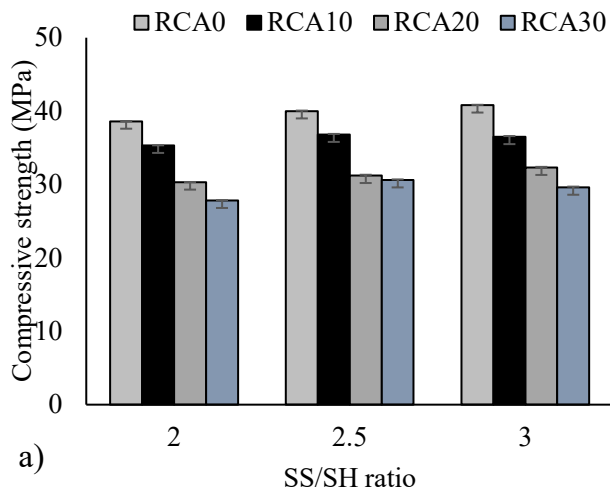
223 Figure 3 shows the effect of sodium silicate to sodium hydroxide ratio and RCA replacement
224 percentage on the compressive strength of concrete. According to Figure 3-a, the lowest value of

225 compressive strengths was found for SS/SH=2 and the highest strength was found for SS/SH=2.5.
226 $\text{Na}_2\text{O}/\text{Al}_2\text{O}_3$ and $\text{SiO}_2/\text{Na}_2\text{O}$ ratios are the major parameters affecting on geopolymer paste
227 properties [27, 57]. The results indicated that although the compressive strength decreased slightly
228 in samples with 30% RCA, the compressive strength increased in almost all cases with the increase
229 of SS/SH ratio.

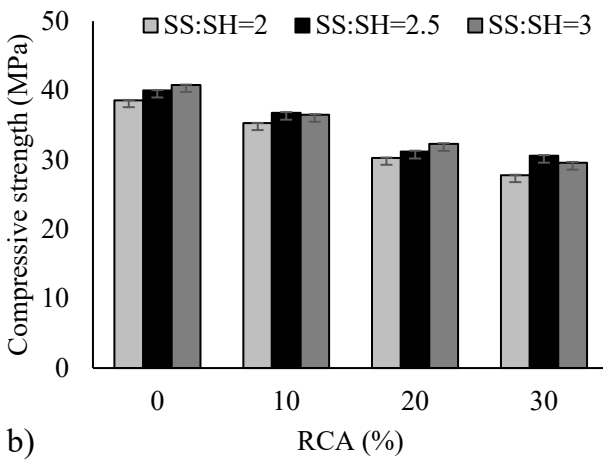
230 In NaOH activated-geopolymer with Si/Na ratio more than 4.4, nano-sized crystals of another
231 zeolite ($\text{Na}_6[\text{AlSiO}_4]_6\cdot 4\text{H}_2\text{O}$) is formed [58]. It has been found that the released aluminum reacts
232 with silicates in the early stages and produces an aluminosilicate oligomer and then the sodium
233 aluminosilicate hydrate (N-A-S-H) gel grows and ultimately takes on a zeolite-like structure [59].
234 Changes of SS/SH ratio mostly affected the non-RCA mixture (strength increase of about 6%),
235 and with the increasing RCA replacement, the effect of SS/SH ratio became negligible. Moreover,
236 when the RCA percentage increased, the geopolymer concretes showed approximately 23–28%
237 reduction in the compressive strength Figure 3-b. Although, compressive strength decreased with
238 the increase of RCA content, the achieved concretes are still appropriate for use as structural
239 concrete. The presence of porous cementitious mortar that adhered to RCA reduces the stiffness
240 of recycled aggregates and consequently replacement of conventional aggregates by RCA have
241 directly negative effect on the compressive strength of concrete. Consequently, the weaker bond
242 at binder-recycled aggregate interface and the porous microstructure of the ITZ caused the
243 reduction of compressive strength. Photograph of a specimen failed in compressive strength test
244 can be seen in Figure 4.

245 According to Allahverdi et al. [60], increasing sodium silicate concentration improves the
246 compressive strength of geopolymer matrix (up to 2 times) by the increased geopolymerization
247 reaction. Also, some researchers [14, 30, 61] reported that increasing sodium silicate to sodium

248 hydroxide ratio results in the development of mechanical properties due to the prevention of water
 249 vapor and structural formation. This supports the results obtained in this study. However, in a
 250 recent research by Alanazi et al. [62], it has been reported that increasing sodium silicate to sodium
 251 hydroxide ratio has a different effect on the mechanical performance of geopolymers that depends
 252 on the type of base material and curing conditions.



253



254

255 Figure 3. Effect of a) sodium silicate to sodium hydroxide ratio and b) RCA replacement on the
 256 compressive strength.



257

258

Figure 4. Geopolymer sample containing RCA after testing.

259

260 3.2 Water absorption

261 Permeability of cementitious composite indicates the porosity and reflects the volume and

262 extensiveness of capillary pores in it. The absorption behavior of concrete plays a vital role in its

263 resistance to ion dispersion and, in general, to the long-term performance. Figure 5 shows the effect

264 of sodium silicate to sodium hydroxide ratio and the percentage of RCA on water absorption of

265 geopolymer concrete. It can be seen from Figure 5-a that the water absorption of geopolymer

266 concrete decreased by about 5.5% to 10%, by increasing the SS/SH ratio from 2 to 3. The decrease

267 of water absorption is attributed to the formation of denser sodium-aluminasilicate gel in the

268 geopolymer matrix by increasing the Na_2SiO_3 content. This is also in agreement with the results

269 reported by previous researchers [63]. Furthermore, the sodium silicate contributes to solubility of

270 metakaolin particles during geopolymerization process. It is noteworthy to mention that there will

271 be stronger bond between unreacted or partially reacted metakaolin particles and geopolymer

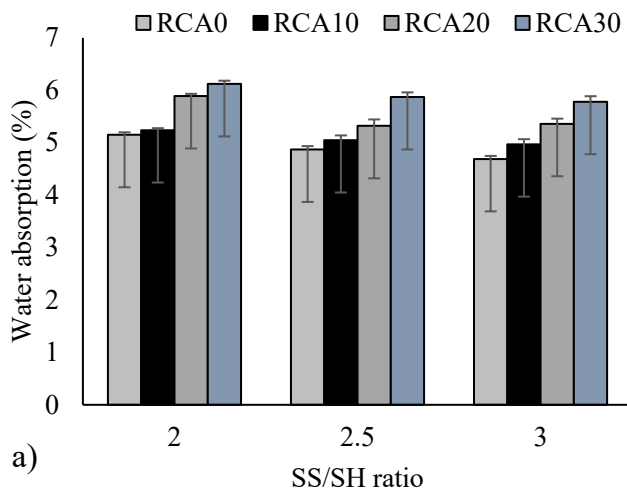
272 matrix when the sodium silicate content is increased. Singh et al. [16], showed that the increase of

273 activator dosage decreased the pores volume and improved the micro structure. The pores that

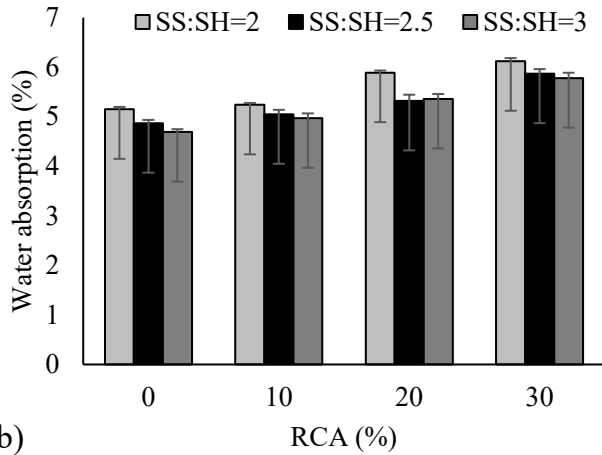
274 showed a narrow distribution, mainly derived their refining due to the more dissolution of

275 aluminosilicate particles and formation of reaction products. Bernal et al. [64] showed that by
276 increasing the ratio of sodium silicate to potassium hydroxide, the pores volume and water
277 absorption of samples are reduced (63% and 73%, respectively) due to the formation of stable
278 products by geopolymerization reaction.

279 According to Figure 5-b, the water absorption of geopolymer concrete increased by up to 23% by
280 increasing the RCA percentage from 0% to 30%. This occurred because the volume of permeable
281 voids strongly affects the water penetration or absorption in recycled aggregate geopolymer
282 concrete. Additionally, attached porous mortar onto the surface of recycled aggregate and presence
283 of masonry products caused an increase of water absorption. As discussed, all the geopolymer
284 concretes except for R2RCA30 had water absorption values less than 6%. It should be also noted
285 that the highest and lowest water absorption belong to the mixtures R2RCA30 and R3RCA0 which
286 were measured to be 6.12% and 4.69%, respectively.



287



288

289 Figure 5. Effect of a) sodium silicate to sodium hydroxide ratio and b) RCA replacement on the
 290 water absorption of concretes.

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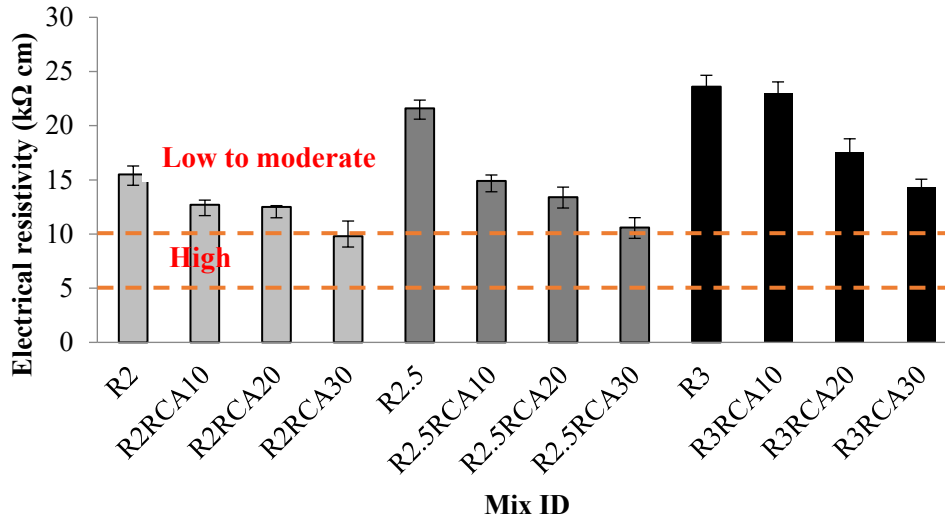
292 **3.3. Specific electrical resistivity**

293 The experimentally obtained electrical resistivity values of geopolymer concretes are plotted in
 294 Figure. 6 which are compared to the limits given by ACI 222 [55] to indicate the probability of
 295 corrosion through the mixes with different sodium silicate to sodium hydroxide ratio and RCA
 296 percentage. It was found that increasing the SS/SH ratio improved electrical resistivity of the
 297 mixtures by about 40 to 81%. The resistivity has been dramatically developed up to 54% when
 298 SS/SH ratio increased from 2.5 to 3. While this amount was about 30% for SS/SH rates 2 to 2.5.
 299 The electrical resistivity measures the quantity of ion transport through the concrete. High
 300 concentrations of ions increase the electrical conductivity of concrete, and hence, decrease of the
 301 electrical specific resistivity. Hydration in cementitious composites due to the pozzolanic reaction
 302 and polymerization in geopolymer composites due to the polymeric reaction are the reasons for
 303 the formation of C-S-H/ N-A-S-H gels. Consequently, the matrix is compressed and the free ion
 304 content is reduced, which result in increase of the electrical resistivity of the matrix [65]. However,

305 the mechanism and performance of the presence of alkali ions in the molecular structure of
306 geopolymer-based materials is not clearly known at present [66]. In previous studies [67-69], it
307 was shown that the test results of the electrical resistivity of geopolymer concrete may be
308 overestimated and appeared unrealistic. This is due to the high electrical conductivity of this type
309 of concrete. It seems that the reason is the presence of more free chemical ions around natural
310 aggregates (binder-aggregate interface) compared to RCA- based mixtures. In RCA-containing
311 mixtures, the effective value of SS/SH ratio is more than non-RCA mixtures. The free Na or alkali
312 metal, OH^- ions and water molecules play a major role in determining the conductivity and
313 electrical properties of geopolymer-based materials.

314 Cui et al. [66] reported that in a $\text{H}_2\text{O}/\text{Na}$ and various Na/Al ratios, the free Na ions fraction of
315 geopolymer materials hold approximately constant after initial curing. Also, their results showed
316 that the electrical conductivity decreased with the increment of $\text{H}_2\text{O}/\text{Na}$ ratio. In the current
317 research, the result also showed that increasing the $\text{H}_2\text{O}/\text{Na}$ ratio resulted in reduction of electrical
318 conductivity which means improvement of electrical resistivity.

319 The electrical resistivity of the geopolymer concrete decreased when the RCA content was
320 increased, as shown in Figure 6. The reduction in resistivity for mixtures with SS/SH ratio of 2,
321 2.5, 3 was 36, 50, and 40% when the use of RCA increases by 30%, respectively. According to the
322 compressive strength and water absorption test results, the presence of RCA in mixtures increases
323 the pore volume and porosity at the old/new aggregates-binder interface or ITZ, which helps the
324 ions transport. Consequently, the reduction of electrical resistivity would be expected. It was seen
325 that the mixtures R2.5RCA0, R3RCA0 and R3RCA10 are categorized as “low” for possibility of
326 corrosion and the other mixtures were in the range where the possibility of corrosion were “low to
327 moderate”.



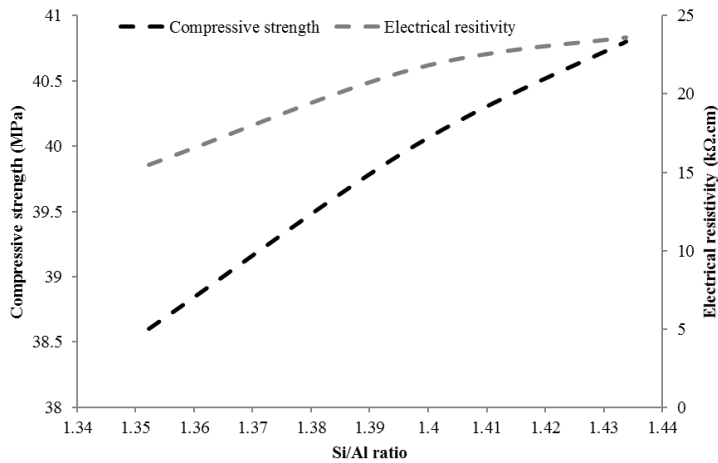
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329 Figure 6. Effect of sodium silicate to sodium hydroxide ratio and RCA replacement on the
 330 electrical resistivity of geopolymer concretes (as per ACI 222 [55]).

331

332 The influence of Si/Al ratio on compressive and electrical resistivity of the specimens with
 333 different monomer ratios has been illustrated in Figure 7, which was achieved based on the data
 334 associated with their percentages in each mixture and the chemical composition of components
 335 presented in Table 1. It can be seen that the compressive strength and electrical resistivity increased
 336 with the increase of Si/Al ratio. The strengths of the mixture with Si/Al ratio of 1.43 were higher
 337 than those with the other two ratios (1.35 and 1.4). It can be also inferred from this figure that the
 338 impact of Si/Al ratio was more significant on the compressive strength test results in comparison
 339 to that of for electrical resistivity.

340



341

342 Figure 7. The impact of Si/Al ratio on compressive and electrical resistivity.

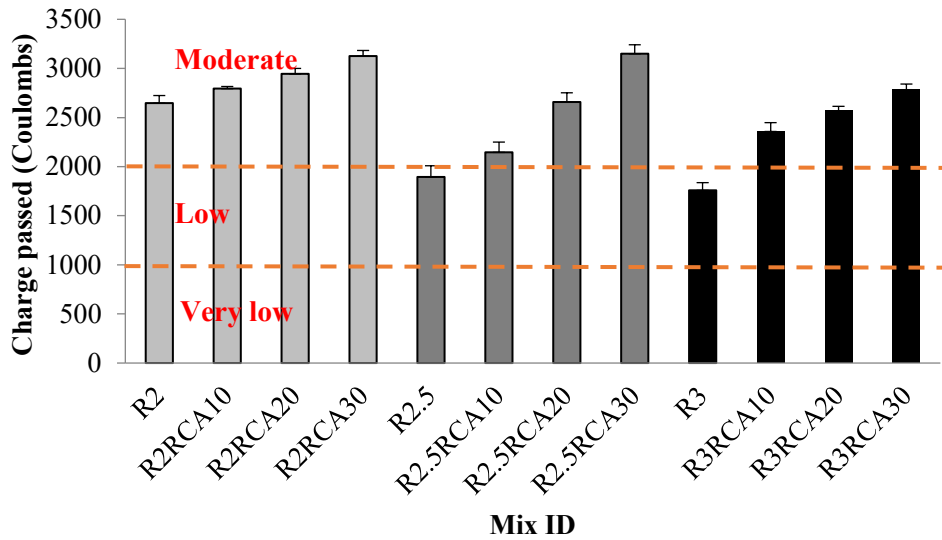
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344 3.4 Rapid chloride permeability (RCP) test

345 The chloride permeability of geopolymer concretes was measured by RCP test as acid-soluble
 346 chloride (total chloride), according to ASTM C-1202 [56]. Test results showed that the increasing
 347 SS/SH ratio reduced the charge passed through concrete, as shown in Figure 8. By increasing
 348 SS/SH ratio to 3, the maximum reduction of charge passed for concrete specimens of the mixtures
 349 RCA0, RCA10, RCA20 and RCA30 concretes was 33, 23, 13 and 11%, respectively. It is well
 350 known that the porosity and concentration of ionized solution directly influence the chloride
 351 permeability of cementitious materials. Consequently, it is clear that by increasing the ratio of
 352 sodium silicate to sodium hydroxide, a significant change in the resistance of the specimens can
 353 be seen. In fact, using a higher monomer ratio reduces the porosity and increases resistance, and
 354 this creates a delay in chloride ion penetration and increases durability. The reduction of chloride
 355 permeability will make it difficult to move the ions into the specimen, which means it will be more
 356 resistant to corrosion.

357 According to Figure 8, increasing the percentage of RCA increased the chloride ion penetration.
358 The main reasons behind this result can be attributed to the high water absorption and porous
359 structure of RCA. Also, it may be due to the presence of attached porous materials or masonry
360 content in RCA which increased the value of chloride ions transport in continuous pores through
361 the geopolymer concrete. It can be seen that in the mixture with high percentage of RCA, except
362 for SS/SH=2.5, the differences in charge passed have increased. It may be due to the chemical
363 reaction of the chloride ion with un-hydrated tricalcium aluminate (C_3A), which formed calcium
364 chloroaluminate hydrate, and consequently, chloride ion is transferred at a lower rate due to the
365 reduction of pores in microstructure [70, 71]. In a research by Ramezani pour et al. [65], it has
366 been shown that high sodium silicate matrices exhibit relatively less chloride ion penetration (up
367 to 40% compared to OPC). In another study [64], it was shown that in mixtures with the same
368 base of aluminosilicate, increasing the ratio of sodium silicate to sodium hydroxide increases up
369 to 37% the charge passed through the samples. The authors stated that this behavior can be
370 attributed to the penetration of Na^+ ions into the matrix, which increases alkalinity of the pore
371 solution.

372 The charge passed of almost all mixtures are higher than 2000 Coulombs which can be categorized
373 as “moderate” chloride ion penetration. Increasing the SS/SH ratio showed improvement of the
374 chloride ion penetration, as shown in Figure 8. However, specimens of mixtures R2.5RCA0 and
375 R3RCA0 were classified as “low” level of chloride ion penetration in the RCP test.



376

377 Figure 8. Effect of sodium silicate to sodium hydroxide ratio and RCA replacement on the chloride
 378 permeability of geopolymer concretes (as per ASTM C 1202 [56]).

379

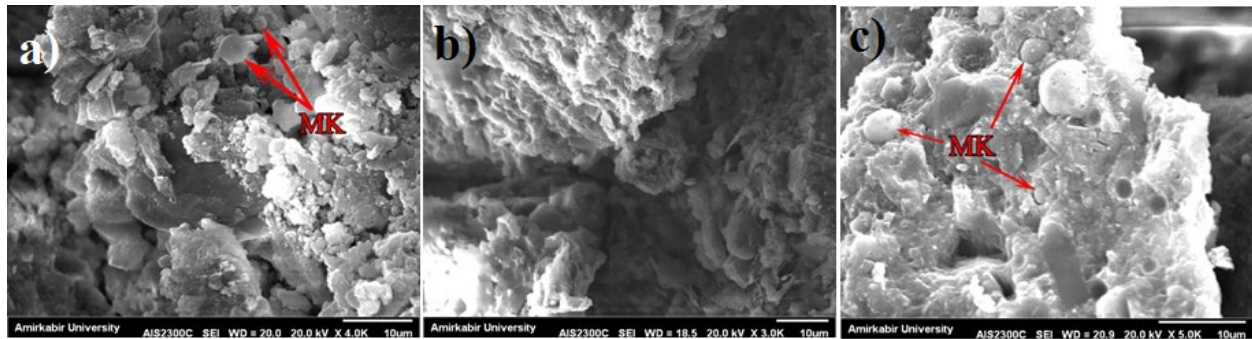
380 3.5. Morphological analysis

381 The microstructure images of geopolymer concretes with different alkali ratios without RCA after
 382 28 days of curing are shown in Figure 9. It can be seen that there are both unreacted MK and silica
 383 in the microstructure of geopolymer concretes. Improvement of the pore structure and density
 384 development are most distinctive differences in the microstructure of the mixtures, as the Na
 385 content increases [72]. The microstructure of the geopolymer paste image of the R2RCA0 mixture
 386 indicates the multiplicity of porosity and non-uniform structure that can be attributed to the
 387 unreacted MK particles. Despite the improvement of the microstructure in higher SS/SH ratios,
 388 number of partially reacted or unreacted particles are observed in the matrix. Since unreacted
 389 particles are composite components, they significantly affect the properties of the transfer,
 390 absorption and mechanical properties of geopolymer materials [73]. It also seems that, some
 391 particles of MK are partially dissolved by alkali, as shown in Figure 9-c. However, its components

392 have been deployed with good homogeneity. According to previous research [74, 75], it could be
393 attributed to the fact that by increasing of Na or K content in activating solutions, the process of
394 polymerization becomes faster and the entrapped air bubbles decreases. Moreover, it has been
395 argued that dimers and smaller silicate monomers help ion pairing with smaller sodium cations,
396 while potassium cations contribute to ion pairing with larger silicate oligomers. This can be an
397 important factor in the apparent density of geopolymer concretes, but on the other hand, it
398 contributes to the difference in microstructure, as shown in Figure 9.

399 The SEM micrographs of the ITZ between aggregates and geopolymer paste are shown in Figure
400 10. By comparison of the Figures 10-a and 10-b, it can be seen that the aggregate-paste interface
401 of specimen R2RCA0 which consists of normal coarse aggregate benefits from a denser and
402 compacted microstructure. On the other hand, a higher porosity can be observed at the interface
403 between the RCA and paste in micrograph of the specimen containing RCA (Figure 10-b). This
404 particular gap was formed because of the masonry debris adhered on the surface of RCA (as
405 shown). Such observation can explain the reduction in compressive strength and increase of
406 permeability and transport properties, as noted in the previous sections. It can be also inferred from
407 Figure 10-c that the hydration products overlapped and interlocked with each other and coated the
408 metakaolin particles tightly, indicating an outstanding geopolymer paste formation adjacent to the
409 RCA. Despite a weak bonding at binder-RCA interface, the geopolymer matrix of the specimen
410 R3RCA30 showed relatively less porosity and more uniform structure, indicating a positive effect
411 of high sodium content. Moreover, a couple of secondary polymerization products including C-S-
412 H or N-A-S-H gels, ettringite and mono/tri sulfate with elongated and needle-like shape, due to
413 chemical reactions can be observed in Figure 10-c, as others researchers [13, 74, 76, 77] showed
414 in their studies . Possibly, the activator solutions or pozzolanic materials coating the surface around

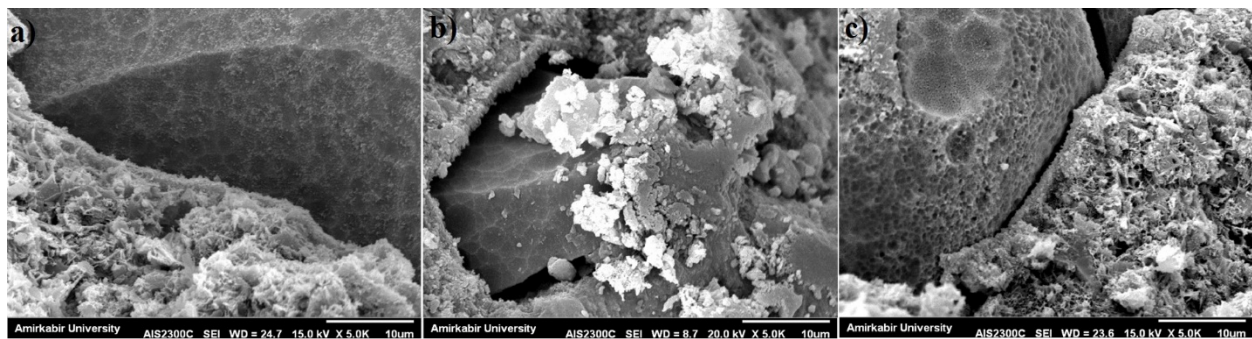
415 of RCA during mixing and curing, which led to their penetration into RCA surface pores
416 consequently, improved the transport properties [73, 78].



417

418

Figure 9. SEM images of a) R2RCA0, b) R2.5RCA0 and c) R3RCA0 specimens



419

420

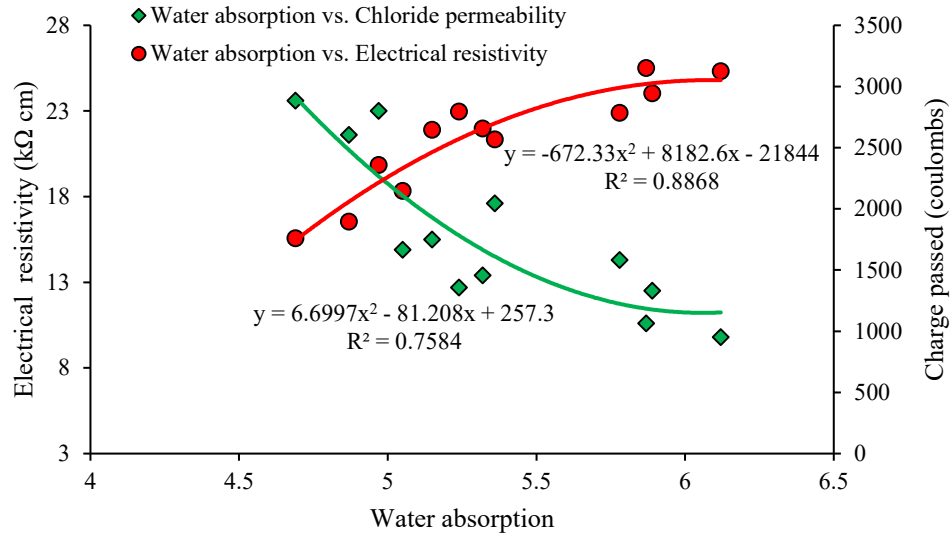
Figure 10. SEM images of a) R2RCA0, b) R2RCA30 and c) R3RCA30 specimens

421

422 3.6. Correlation between the durability test results

423 In order to understand the interdependence between the geopolymer concrete characteristics, the
424 electrical resistivity and RCP test results are plotted against water absorption in Figure 11. It was
425 found that the coefficient of correlation between water absorption and electrical resistivity test
426 results is relatively moderate with R^2 of 0.76. The coefficient was 0.88 for correlation between
427 water absorption and permeability. A high correlation coefficient reflects a stronger relationship
428 between these different test results.

429



430

431 Figure 11. Correlation between electrical resistivity vs absorption and chloride permeability vs
 432 absorption results

433

434 **4. Conclusion**

435 In this study, the effect of different Na₂SiO₃/NaOH ratios on mechanical strength and durability
 436 of alkali-activated metakaolin concrete containing recycled concrete aggregate are evaluated. The
 437 following important conclusions can be drawn based on the obtained results:

438

- 439 **1.** Increasing the ratio of sodium silicate to sodium hydroxide from 2 to 3 increased compressive
 440 strength of the specimens. The greatest change was observed with a 6% increase in compressive
 441 strength. In addition, the higher the ratio of sodium silicate to sodium hydroxide, the lower was
 442 the water absorption. The water absorption decreased by 9% when the monomer ratio was
 443 increased from 2 to 3.
- 444 **2.** The electrical resistance of specimens increased by 39% and 52%, when the monomer ratio was
 445 increased from 2 to 2.5 and 3, respectively. The probability of occurrence of corrosion of

446 specimens with a monomer ratio of 2 was in the range of low to medium and that with increasing
447 the proportion of sodium silicate to sodium hydroxide was in the low range.

448 **3.** Images of electron microscopy of hardened geopolymer paste confirmed the significant increase
449 in density and uniformity of polymer products in the presence of suitable monomer ratios.

450 **4.** It was confirmed that the geopolymer concretes incorporating RCA indicated a reduction in
451 compressive strength of around 8% to 28%, in comparison to the mixtures with the same
452 monomer ratio. It is noteworthy to mention that the compressive strength achieved was still
453 high enough for structural applications.

454 **5.** The microstructure of geopolymer concretes containing RCA indicated a relatively weak
455 aggregate-paste interface which is attributed to the old mortar attached on the surface of RCA.
456 Nevertheless, the hydration products overlapped and interwoven with each other to result in
457 good geopolymer paste formation around the recycled aggregate particles.

458 **6.** The results of this study showed the feasibility of producing sustainable geopolymer concrete
459 using available local materials such as metakaolin as binder and recycle concrete aggregates as
460 partial replacement of virgin coarse aggregate.

461

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465

466 **References**

467

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