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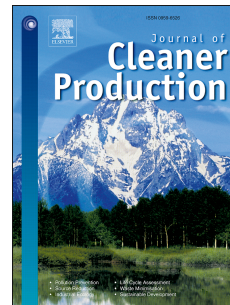


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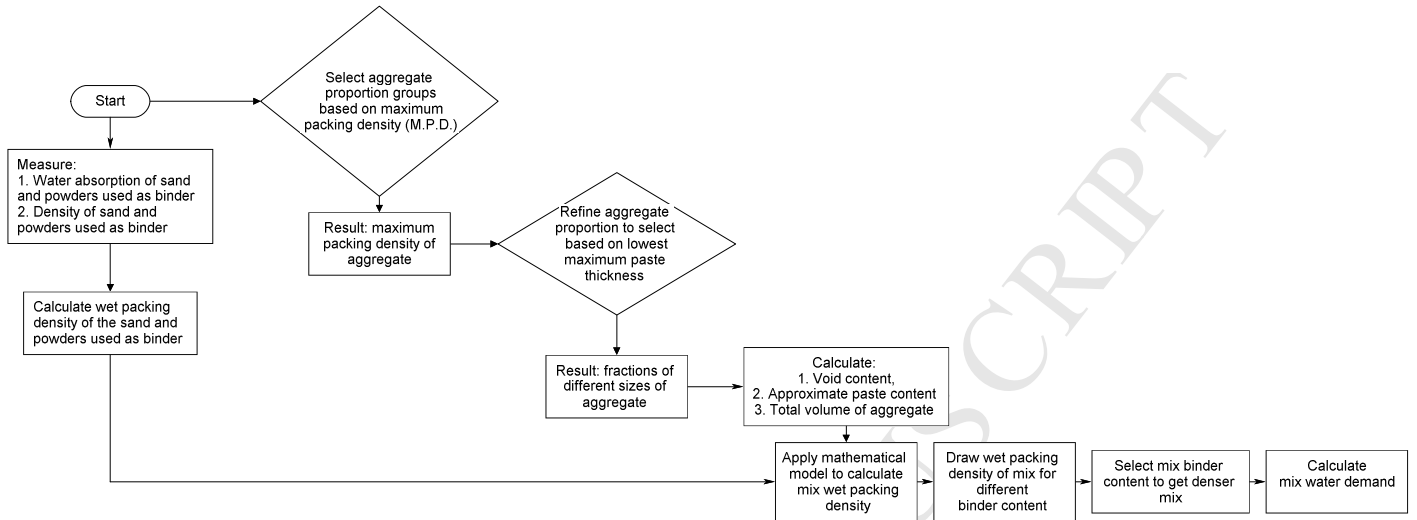
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Efficient mix design of alkali activated slag concretes based on packing fraction of ingredients and paste thickness

1 *Efficient mix design of alkali activated slag*
2 *concretes based on packing fraction of*
3 *ingredients and paste thickness*

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9 **Abstract**

10 Many studies have been dedicated to the properties of alkali-activated slag concretes as a form of low-
11 carbon high performance concrete, but less work has been focused on the application of mix design
12 procedures to have a dense, durable and cost-efficient alkali-activated concrete. This study proposes a
13 method for selecting the mix proportions of alkali-activated concretes based on the packing fraction of
14 materials. The design method is based on the selection of the volumetric proportions of sand and
15 coarse aggregate according to an ideal particle gradation curve. To validate this method, trial castings
16 were carried out for concrete mixes containing alkali activated slag (AAS) with different paste
17 contents to suggest the most cost-efficient concrete for different classes of workability and
18 applications. Compaction and pore structure of these mixes studied by optical microscopy have shown
19 that the design of AAS concretes based on the proposed method resulted in a dense and workable mix.

20
21 **Keywords:** Alkali activated slag concrete; efficient mix design; packing fraction

22
23 **1 Introduction**

24 The use of alkali activated materials as binders to produce concretes has gained increasing attention
25 due to the need for emissions reduction, energy conservation and environmental considerations in the
26 cement and concrete industry. Much work has been undertaken in optimising the chemistry of alkali-
27 activated concretes made from different mineral resources and industrial by-products (Shi et al., 2006;
28 Provis and Deventer, 2014; Bondar et al., 2011; Lloyd and Rangan, 2010; Bernal et al., 2011, 2012;

29 Pavithra et al., 2016; Yost et al., 2013; Ng and foster, 2013; Rafeet et al., 2017), but there is still the
30 need for a rational mix design methodology which can be followed for fully efficient production of
31 these concretes. In 2008, Lloyd and Rangan (2010) proposed a mix design method for alkali activated
32 fly ash-based concrete but did not discuss how to deal with the effects of the ingredients' specific
33 gravities, while assuming a constant concrete density of 2400 kg/m^3 . As Shi et al. (2006) reported, the
34 mix design methods presented for alkali activated slag concretes since 1967 are all based on
35 experimental and empirical methods and need large amount of experiments and statistics to determine
36 the relationship between the proportion of concrete constituents and the properties of concretes.
37 Rafeet et al. (2017) published guidelines for mix proportioning of alkali activated fly ash/slag
38 concrete, but there was no discussion on the density of concrete or how the fractions of aggregates
39 with different particle size distributions affect the mix design. Li et al. (2018) recently reported a
40 general mix proportioning method for alkali activated slag-based concretes considering highest bulk
41 density of combined aggregates while the binder content was determined based on experimental
42 method. The effect of compaction of wet ingredients and binder content was not considered in
43 designing the mixture to get the possible highest density and minimum porosity.

44 The use of chemical activators in alkali-activated concretes also causes cost to be an issue when
45 making alkali-activated concretes competitive with Portland cement-based concrete. McLellan et al.
46 (2011) have shown that there is a wide variation in the calculated financial and environmental cost of
47 geopolymers. Their study indicated potential for a 44-64% reduction in greenhouse gas emissions at
48 financial costs between -7% and 39% higher than Portland cement, in the Australian context,
49 depending on mix design. Thus, it is important to find the optimum paste content, based on optimal
50 binder/aggregate and water/binder ratios, in alkali-activated concretes that can make workable
51 concrete while meeting criteria for minimum strength grade and good durability.

52 Particle packing fraction measurements and particle packing models can help optimise the water
53 demand and/or the paste content of concretes, while achieving a constant workability. In optimising
54 the concrete composition by targeting a maximum particle packing fraction, the particle size
55 distribution should be selected so as to fill the voids between large particles with smaller particles and
56 to obtain a dense and interlocking particle skeleton in the resulting hardened concrete. In general, the
57 higher the packing fraction of the aggregates, the smaller will be the volume of the voids to be filled
58 by paste, and so less paste will be needed to provide sufficient excess over the void-filling material
59 (Rafeet et al., 2017). It is this excess paste (above the paste needed to fill the voids between the
60 aggregate particles) that lubricates the aggregate particles and enables the concrete to flow. Therefore,
61 a higher packing fraction of the aggregates would at the same paste volume lead to a higher
62 workability, or at the same workability allow the use of a smaller paste volume to increase the
63 dimensional stability, paste consumption and carbon footprint (Li and Kwan, 2014; Kwan and Wang
64 2008; Domone and Soutsos, 1994; de Larrard and Sedran, 1994; Lange et al., 1997).

65 Moini et al. (2015) reported that the aggregate blends have considerable effect on concrete
66 performance and as a result of aggregate optimisation, the concrete compressive strengths in their
67 study increased by up to 37%; the improvement was slightly more pronounced at earlier age when the
68 strength of the cementitious matrix is still low and concrete strength depends more on the load
69 carrying capacity of the aggregates.

70 Zou et al. (2003) and Sutcu and Akkurt (2007) have also worked on packing of mono-sized and multi-
71 sized mixtures and indicated that porosity of the assembly is strongly affected by particle size, their
72 distribution and moisture content.

73 Recent work by Miller et al. (2016) has shown that concrete with a high water to binder ratio
74 containing only cement as the binder provided a lower ratio of global warming potential (GWP) to
75 compressive strength than some of the mixtures containing large quantities of replacement binder.
76 This shows that large volume replacement of cement as a binder may not always be the most
77 sustainable solution if mix designs are not optimised.

78 The aim of this paper is to build a consistent, rational and scientifically based approach for designing
79 alkali activated slag concrete mixtures. Modern concretes for general use in engineering applications
80 must meet a comprehensive list of requirements, which are not limited to the final compressive
81 strength, but also include rheological properties, early age characteristics, deformability properties
82 and durability aspects. Workability and compressive strength evolution were studied for mixes with
83 different binder contents and water/binder ratios. Furthermore, the pore structures and microstructures
84 of mixes were studied by optical microscopy to support the mix design approach.

85

86 **2 Experimental programmes**

87 2.1 Materials and methods of characterisation

88 The primary raw material used in this study is a granulated blast furnace slag which was supplied by
89 ECOCEM, France (Table 1). The particle size distribution of slag was determined by laser diffraction,
90 and the particle density was measured using a Le Chatelier flask; Fig. 1 and Table 2.

91 A centrifugal consolidation method was used to determine the actual water demand of slag (water
92 absorption) according to the method Miller et al. (1996) suggested for measuring water demand of
93 powders and fine particles based on achieving maximum packing density. In this method, 300 g of dry
94 powder was mixed with a known content of water, in a 3 litre Hobart mixer, for 2 minutes at low
95 speed. Then 50 g of this mix is poured into 90 mm long test tubes with internal diameter of 22 mm.
96 By determining the mass of the paste in the test tube, the amount of the powder and water in the test
97 tube at the beginning of the test were known. The test tube was then centrifuged for ten minutes at
98 4000 rpm in a Dumeé Jouan E82N centrifuge with an internal diameter of 300 mm. The excess
99 amount of water which came out as a supernatant layer on the top of the paste was removed with a

100 pipette after centrifuging. By determining the amount of water removed, the amount of water
101 absorbed was calculated and reported in Table 2.

102 Sodium hydroxide (NaOH) powder was dissolved in tap water and used along with sodium silicate
103 solution (WG) to act as alkaline activators in concrete production at specified concentrations and
104 compositions, as shown in Table 6. The chemical composition of the as-received sodium silicate
105 solution was 15.5% sodium oxide (Na₂O), 30.5% silicon dioxide (SiO₂), and 54% water.

106 The aggregates used in this study were crushed basalt from local sources in Northern Ireland and
107 comprised of 10 mm and 16.5 mm crushed fine and coarse aggregates and 4 mm sand complying with
108 BS EN 12620 (2013). Sieve analysis of the aggregates is shown in Fig. 2. The bulk specific gravity
109 and water absorption of these materials were measured based on BS EN 1097-1 and are presented in
110 Table 3.

111 All size fractions of aggregates for the different mixes are presented in Fig. 3. The ingredients were
112 mixed and filled into a 150mm diameter x 300 mm height cylindrical steel container in three equal
113 layers. After filling each layer, the mixed aggregates in the container were compacted by applying 20
114 compaction blows with a metal tamper rod with diameter of 16 mm. The mass of the compacted
115 mixed aggregates was measured, and the bulk dry packing fraction (refer to Table 4) was determined
116 for the compacted mixed aggregates using the following equation:

$$117 \quad \gamma = \frac{4W}{\pi D^2(H-\delta H)} \quad (1)$$

118 Where γ is the bulk dry packing density of compacted mixed aggregates (kg/m³), W is the mass of
119 compacted mixed aggregates (kg), H and D are the height and diameter of the container (m), and δH
120 is the height reduction due to compaction of materials (includes the three individual differences in
121 height), m.

122 Compacted dry packing fraction of mixed aggregate (φ) were determined by the following equation:

$$123 \quad \varphi = \gamma \cdot \sum_{i=1}^n \frac{A_i}{P_i} \quad (2)$$

124 Where P_i is the grain density (bulk specific gravity) of the aggregate fraction (kg/m³), A_i is the fraction
125 of the aggregate (mass %) and n is the number of aggregate fractions. The results of this analysis are
126 presented in Table 4.

127 For measuring the wet packing fraction of crushed aggregate fractions individually (i.e., 5-10 mm and
128 10-16.5 mm), the wet aggregates (saturated aggregates with moisture on the surface) were filled
129 separately into a steel container in three equal layers. Each layer was compacted by applying 20
130 compaction blows with a metal tamping rod before progressing to the next layer. The weight of the

131 container with and without the aggregates and the volume of space filled by the aggregate was
132 measured and was used to determine the bulk wet packing fraction.

133 2.2 Mixing

134 All the concrete ingredients were mixed in a laboratory pan-mixer according to the formulations in
135 Tables 5 and 6. Crushed basalt aggregates and sand were dry-mixed together for a minute; after
136 adding the granulated blast furnace slag (GGBS) powder, mixing continued for 2 minutes and then the
137 sodium hydroxide solution was added and after 2 minutes further mixing, sodium silicate solution was
138 added and mixing continued for a minute.

139 2.3 Measuring fresh properties

140 The slump value and flow test were measured according to Part 2 and Part 5 of BS EN 12350 (2009),
141 respectively. The mixes studied here span from highly fluid to very stiff, both tests were conducted
142 for each mix, but the slump data are more instructive for moderate to stiff concretes, while slump flow
143 is more useful for highly flowable mixes where the measured slump values are very high.

144 The wet (fresh) density of each mix, was measured based on the weight and volume of fresh concrete
145 immediately after filling the two cube moulds in three equal layers and vibrating them at each time.

146

147 2.4 Casting and curing of the specimens

148 From each concrete mix, nine 100 mm cubes were cast, and used for the determination of
149 compressive strength according to BS EN 12390-3 (2009) at 2, 7, 28 and 90 days of age, with two
150 replicate samples tested per age; the remaining cube sample from each mix was used for preparing
151 thin section samples as described below. The density of each mix was measured based on the weight
152 and volume of the cubes cast for measuring compressive strength before crushing them. The concrete
153 specimens were cast in three equal layers and compacted on a vibrating table. After casting, all the
154 specimens in mould were covered with plastic sheets and left in the casting room for 24 hrs.
155 Thereafter, the samples were removed from the mould and kept in plastic zip bags at 20°C until the
156 test date.

157 The cube from each set that was not used in compressive strength testing was cured for 90 days, at
158 which time material was sampled from its centre to be prepared for thin section samples. For
159 preparing thin section samples, following impregnation with blue dye resin and drying, the back of the
160 specimen was glued to a glass plate. Then the selected face of the concrete specimen was prepared by
161 dry grinding and mounted on the glass slide. The thin section optical microscopy was used to study
162 the pore structure of all of these mixes.

163 3 Results and discussions

164 The basis of the mix designs used in this study is a standard concrete mix being used in the round-
 165 robin testing programme of RILEM Technical Committee 247-DTA¹ (2014), which was designed to
 166 target a relatively low activator content for making structural concrete from alkali activated slag. In
 167 the RILEM TC 247-DTA mix design, oven-dried rounded quartz aggregates had been used, yielding
 168 flowable concretes, while crushed basalt aggregates were available for use in this work. The first trial
 169 mixes (Table 5) showed that even by increasing the water/binder (W/B) ratio from 0.45 to 0.5, no
 170 workability could be achieved with crushed basalt aggregates. So, workability of the mixes appeared
 171 to be an issue which was addressed through the following steps:

- 172 • Increasing the ratio of coarse to fine aggregates
- 173 • Decreasing the aggregate to binder (A/B) ratio.
- 174 • Considering the water absorption of materials based on saturated surface dry conditions, to
 175 add extra water required for both hydration and workability.
- 176 • Increasing the sodium silicate dose by 1% (relative to the mass of slag) and comparing the
 177 workability and strength results.

178 The first three of these items can be met reasonably by considering the packing fraction of the
 179 particles and the paste thickness needed to reach to a cost efficient dense workable mix design. The
 180 packing fraction of the mixture is defined as the solid volume of (aggregate) particles in a unit
 181 volume, which is optimised by mixing different fractions of particles with a view to minimise the
 182 porosity, which allows the use of the least possible amount of paste. However, this must be balanced
 183 with the provision of sufficient paste thickness to give suitable flow characteristics.

184

185 *3.1 Selection of aggregate proportioning based on packing fraction and paste thickness*

186 For considering aggregate proportions with maximum packing fraction in the mix design, different
 187 fractions of sand and crushed aggregates were selected, optimising away from the simple blend
 188 fractions used in the baseline mix design (Fig. 3). The results are presented in Fig. 4 and show that the
 189 maximum packing fraction (resulted from equation 2 for different fractions of aggregates mixes) is
 190 0.68 for the three aggregate proportions: 40:12:48, 40:18:42, and 50:25:25 (Mass fractions reported as
 191 sand:10 mm:16.5 mm). Based on these results, void content (VC) will be $(1 - \phi) = (1 - 0.68) =$
 192 0.32. The volume of the cement paste must be more than the void space between particles to overfill
 193 them after compaction. Thus paste content (PC) will be 10% in excess of void content (Raj et al.,
 194 2014) $(1.1 \times VC) = (1.1 \times 0.32) = 0.352$. The total solid volume content (TSVC) of aggregate for the
 195 above three aggregate proportions was calculated by the equation below:

196

$$TSVC = \sum_{i=1}^n A_i / P_i \quad (3)$$

¹<https://www.rilem.net/groupe/247-dta-durability-testing-of-alkali-activated-materials-290>

197 and the mass of each type of aggregate per cubic meter was thus calculated as $\left[\frac{(1-PC)}{TSVC} \cdot A_i \cdot 1000\right]$. The
 198 minimum required binder content was calculated for different water/binder (W/B) ratios
 199 $(1000PC/(W/B+1/\rho_{\text{binder}}))$, Table 7.

200 De Larrard (1994) has investigated in more detail the variations of concrete compressive strength,
 201 with the topology of the aggregate skeleton as the primary parameter and showed that the second key
 202 parameter governing the concrete compressive strength is the maximum paste thickness (MPT). He
 203 mentioned that this physical parameter represents the mean distance between two aggregates, if each
 204 aggregate is surrounded by a paste layer whose thickness is proportional to the aggregate diameter,
 205 equation 4:

$$206 \quad e_M = D(\sqrt[3]{g^*/g} - 1) \quad (4)$$

207 Where e_M is the MPT, D is the maximum size of aggregate, g^* is the packing fraction of the aggregate
 208 and g is the actual volume of the aggregate in the mix (de Larrard and Sedran, 1994). For the above
 209 aggregate mix proportions with maximum packing fraction (= 0.68), the maximum paste thicknesses
 210 are 3.711, 3.729 and 3.747 mm for aggregate proportion 40:12:48, 40:18:42, and 50:25:25,
 211 respectively. Due to the use of a crushed basalt aggregate in this study, these values are selected to be
 212 at the top end of the range 0.61 to 3.83 mm which Torres et al. (2015) have presented as maximum
 213 paste thicknesses for normal concrete at the highest compaction level and the lowest paste content.

214 The lowest maximum paste thickness belongs to aggregate proportion 40:12:48 (Sand: 10 mm: 16.5
 215 mm).

216 Several particle packing models have been developed over the past decades and applied to concrete
 217 mix design. Andreasen (1930) developed a packing theory based on continuous particle distributions.
 218 The equation he derived, the Andreasen equation for dense packing, is $F_v(a) = (a/a_{\max})^n$ where $F_v(a)$
 219 is the cumulative finer volume distribution, and a_{\max} is the maximum particle size. Andreasen's
 220 experiments indicated that to obtain a dense packing, n must be 0.33 to 0.50. The Andreasen equation
 221 indicates that infinitely small particles are required to achieve the theoretically denser packing. For
 222 real particle system, the minimum particle size is limited. Dinger and Funk recognized that real
 223 particle systems must have some minimum particle size (a_{\min}) (Andreasen, 1930; Zheng et al., 1990;
 224 Jones and Zheng, 2002). They modified the Andreasen equation to indicate a size range a_{\max} to a_{\min}
 225 and a cumulative finer fraction of zero when $a = a_{\min}$:

$$226 \quad F_v(a) = (a^n - a_{\min}^n) / (a_{\max}^n - a_{\min}^n) \quad (5)$$

227 A study on four particle packing models used to proportion the mix constituents (solid particles) of
 228 concrete to produce a minimum voids ratio (or maximum packing fraction) was published by Jones et
 229 al. (Jones et al., 2002). It was found that the models give broadly the same output and suggest similar
 230 combinations of materials to give the minimum voids ratio. It was noted that proportioning concrete

231 mix constituents to minimise voids ratio did tend to produce a harsher and unworkable mix than
 232 normal (Jones et al., 2002).

233 Validation was performed with Elkem Emma[®] software incorporating the modified Andreasen model.
 234 For each aggregate proportion i.e. 40:12:48, 40:18:42, and 50:25:25 and a constant paste volume, by
 235 decreasing the binder content and increasing the water to binder ratio the density has been reduced
 236 (Table 7). By using finer crushed aggregates and more sand in the aggregate mixtures the density has
 237 been increased a little, but on the other hand there is a certain probability that voids may be trapped
 238 under an aggregate. More precisely, the fractions which may lead to entrapment of the air bubbles are
 239 the ones that have simultaneously a sufficient particle size and a high specific surface (de Larrard,
 240 1999). Therefore, the sand fractions are the most efficient at retaining air, while the cement paste
 241 cannot fix a significant amount of entrapped air in the absence of air entraining admixtures, which
 242 were not used here. Furthermore, the water content of sand at saturated surface dry conditions is
 243 generally much higher than that of crushed aggregates, so the likelihood of changes in concrete water
 244 content will be higher. Finally, the more sandy the mixture, the more sensitive will be the strength to
 245 changes in workability (de Larrard, 1999). The effect of air content on slump greatly increases with
 246 the sand content of the mixture and this air content will in turn affect the compressive strength. Thus,
 247 considering the above description and the least maximum paste thickness, the aggregate proportion
 248 40:12:48 was used for the mixes in this study.

249

250 *3.2 Selection of mix binder content based on wet packing fraction (WPF)*

251 The wet packing fraction (WPF) of slag, sand, crushed fine and coarse aggregates were measured
 252 separately, and these values were used to calculate the total WPF of the mixture with different binder
 253 (slag) contents based on Goltermann et al. (1997), these are presented in Table 8. It is known that the
 254 WPF of fine particles such as slag and sand is inversely proportional to water/powder ($\frac{w}{p}$) at the same
 255 fluidity (Zou et al., 2001; Ye et al., 2008). The higher the WPF, the smaller will be the water to
 256 powder (or water to fine particle) ratio. This is because higher packing fraction leads to less pores, so
 257 smaller amount of paste is needed and cause smaller water to powder ratio which is direct
 258 proportional to the paste content. In equation 6, ρ' and $(1-\rho')$ are the volume fractions occupied
 259 respectively by powder (or fine particle) and water in the paste (or mix), which represent the WPF and
 260 the void content (Ye et al., 2008). If the true densities of water and particles are ρ_w and ρ_p , the
 261 relationship between water to powder (or water to particle) ratio and wet packing fraction
 262 (WPF= ρ') can be written:

$$263 \quad \frac{w}{p} = \rho_w(1 - \rho')/(\rho_p \rho') \quad (6)$$

264 and WPF could be obtained from the water to particle ratio which is measured at the least fluidity for
 265 samples with different particle size distribution (Ye et al., 2008; Miller et al., 1996). Using equation

266 (6) and $(\frac{w}{p})$ ratio measured by centrifugal consolidation method (0.35 for slag and 0.178 for sand), the
267 WPF of slag and sand was found to be 0.499 and 0.674 respectively. For the two different size
268 fractions of crushed aggregates (5-10 mm and 10-16.5 mm), WPF was measured 0.536 and 0.548.
269 Then using Golterman et al. (1997), WPF was calculated for mixed coarse crushed aggregates without
270 and with sand, to be equal to 0.565 and 0.636 respectively. Further, the total WPF for mixes with
271 different binder contents was calculated, Table 8. Fig. 5 shows that maximum packing fraction is
272 obtained for 350 kg/m³ of GGBS and the value remains constant up to 450 kg/m³. The WPF decreased
273 for mixes with binder content greater than 450 kg/m³.

274 The total volume of aggregates and binder of the mixes, calculated as described above, is used to find
275 the mix water demand. The steps required to select mix proportions for a mix are shown in Fig. 6 as a
276 flow chart.

277 3.3 Density, slump, flow and compressive strength

278 Slump, flow, density and compressive strength were measured for all the five groups of mixes
279 presented in Table 6. The test variables were binder content and water content, and their influence on
280 slump, flow, density and compressive strength was assessed in order to validate the mix design
281 procedure based on a particle packing approach. Testing these mixes shows the best combination of
282 binder and water content at the same paste volume for a strong and workable mix.

283

284 3.3.1 Density

285 Fig. 7 shows that for mixes with W/B of 0.45, the highest wet concrete density correspond to the
286 mixes with binder content from 437.7 to 450 kg/m³, with a maximum of 2482.7 kg/m³ at a binder
287 content of 447.6 kg/m³. At W/B = 0.55 this will happen for mixes with binder content from 365.1
288 kg/m³ to 414.6 kg/m³ with a peak of 2439.5 kg/m³ at 398.1 kg/m³. Increasing water binder ratio to
289 0.65, this is resulted for mixes with binder content from 315.6 kg/m³ to 375 kg/m³ with a maximum of
290 2436.6 kg/m³ at a binder content of 351.9 kg/m³. These results confirm previous calculation results
291 presented in Table 7 where the calculated binder content based on packing fraction and paste
292 thickness method for mixes with water/binder equal to 0.45, 0.55 and 0.65 was resulted 440, 391.11
293 and 352 kg/m³ respectively. Density measurement results show that at W/B of 0.65 mixes with a
294 binder content of 350 kg/m³ has shown the highest density at different ages up to 28days (Fig. 8c).
295 Furthermore, with regarding to the results is shown in Fig. 8a&b, at W/B of 0.45 and 0.55, mixes with
296 a binder content of 450 kg/m³ and 400 kg/m³ has shown the highest wet concrete density.

297 Fig. 8 depicts that density reduces with increase in W/B, this is to be expected as more water is
298 present in the matrix. It is also obvious that density reduces with age because of water evaporation
299 due to the semi-dry (sealed) curing condition which was used to prevent leaching of alkalis due to
300 water curing. This reduction is more obvious for mixes with higher binder content due to the fact that

301 they have a higher quantity of water that can evaporate. This evaporation is likely to result in a more
302 porous concrete. In addition, minimum variability as the difference between the three replicates
303 occurred for mixes with W/B of 0.55, and as binder content reduces the wet density decreases for low
304 W/B concrete, but this trend reverses for high W/B concrete.

305 The lowest density is observed for mixes with binder content equal to 400 kg/m^3 and 450 kg/m^3 and
306 water to binder ratio equal to 0.65 for all ages (Fig. 8). When the fraction of aggregate and paste are
307 considered based on packing fraction (lower void content) then less difference can be observed
308 between the density of these mixes compare to the mixes with lower binder content.

309 *3.3.2 Slump and flow*

310 AASC has often been known for its relatively low slump value and rapid setting behaviour. Slump
311 values of 60-120 mm have been reported in the literature (Collins and Sanjayan, 1999). The purpose
312 of this testing programme was to demonstrate the effect of mix design based on packing fraction on
313 the slump value that AASC is capable of producing, and to identify the governing W/B and binder
314 content that are necessary to achieve high slump. As is evident from Fig. 9 and 10, for a given W/B,
315 the drastic change of slump and flowability happens at the binder content calculated based on the mix
316 design with highest packing fraction method (i.e. for W/B=0.55, is at 400 kg/m^3). For mixes with
317 water to binder ratios of 0.45 and 0.55, the minimum binder content required to produce a workable
318 mix is 450 and 400 kg/m^3 , respectively.

319 The use of sodium silicate as a dispersant is reported to reduce the yield stress of a paste considerably,
320 to obtain good workability (Landrou et al., 2016). Fig. 9 shows that adding 1% more sodium silicate
321 to increase the slump is not effective when water to binder ratio is equal to 0.45, but at higher W/B, it
322 is more effective and increases the slump from 100 mm to 230 mm and 225 mm to 250 mm, for the
323 mixes with water to binder ratio of 0.55 and 0.65, respectively.

324 The contour map graphs in Fig. 14 (a) show slump as a function of binder content and water to binder
325 ratio, for different mixes with the same activator content. As it can be observed, a water to binder ratio
326 equal to 0.55 seems to be the minimum amount of W/B in order to achieve all classes of workability
327 by varying binder content from 350 to 450 kg/m^3 .

328 *3.3.3 Compressive strength*

329 The mechanical and durability properties of concrete are highly influenced by its density (Bondar et
330 al., 2018). A denser concrete provides higher strength and fewer amount of voids and porosity. Fig. 11
331 and 12 show the relation between the density of the mixes and their compressive strength at different
332 ages. Compressive strength of samples at different age has a good linear correlation with wet density
333 (Fig. 11) and with dry density (Fig. 12).

334 An overall comparison of the results for different W/B (Fig. 13) makes the effect of W/B very
335 apparent for all the mixes. Whereas mixes with W/B of 0.45, 0.55 and 0.65, offers the minimum value
336 of strength around 22, 11 and 10 MPa at 2 days and 42, 29 and 24 MPa at 90 days. The highest
337 compressive strengths are observed for mixes with water to binder ratio equal to 0.45: in the range of
338 22-26 MPa at 2 days and 42-49 MPa at 90 days. Increasing the sodium silicate dose by 1%, with the
339 aim of increasing the workability, decreases the compressive strength 29% at early ages and up to
340 17% at longer ages (Fig. 13(b)).

341 Wasserman et al. (2009) reported that strength of Portland cement (PC) based concrete is a function of
342 w/c and independent of the cement content. This is also true for AASC; comparing different mixes
343 tested at the same age and with same water to binder ratio shows that increasing the binder content
344 has no major effect on strength. For W/B = 0.55, increasing binder content to more than 350 kg/m³
345 cannot produce significantly higher compressive strength. Therefore, increasing the cementitious
346 material content will not in itself guarantee higher strength, although mixes with binder content less
347 than 400 kg/m³ do not offer good workability. For lower binder contents, the mix is not workable and
348 the voids content increases in concrete and makes them more porous. Fig. 14 can be used to design
349 alkali activated slag concrete mixtures using similar aggregates to those used in this study, with the
350 lowest content of chemical activators for practical applications. It shows that to achieve the minimum
351 value of the slump class of S2 (50 mm) specified in BS EN 206 (2013) the minimum water to binder
352 ratio and binder content are 0.5 and 400 kg/m³ respectively.

353 3.4 Pore structure

354 Thin sections were prepared from all twelve concrete samples as they represent alkali activated slag
355 concrete mixes with different binder to aggregate ratio (binder content) and water to binder ratio. Fig.
356 15 shows all twelve thin section images taken at a magnitude of 5^x from these samples. A narrow
357 zone of cement paste with a high porosity and/or cracks is occasionally seen at the interface between
358 the coarse aggregate particles and the surrounding hardened binder paste in some of them, which can
359 be due to a water concentration gradient based on heterogeneity close to the aggregate (San Nicolas
360 and Provis 2015) (Fig. 15 d, g, h & k). Furthermore, the coarse aggregate particles also exhibit signs
361 of internal cracking (Fig. 15 c & f). Concrete is a three-phase material, in which the gaseous phase can
362 never be totally removed. The images in the same row in Fig. 15 show samples with same binder
363 content and different water to binder ratios, while the images in each column present mixes with same
364 water to binder ratio and different binder content. It can be observed that mixes with water to binder
365 ratio of 0.45, 0.55 and 0.65, a corresponding binder content of 450, 400 and 350 kg/m³ result in a
366 dense matrix. Indeed, the density is a good indicator that reflects the durability of concrete. However,
367 the size distribution and the sinuosity of pores can also effect the durability of concrete. Image
368 analysis was performed by image analysis software (Image Pro) and the void contents in the concrete

369 including percentage of pores in the binder, were measured by counting pixels in area of interest with
 370 the same colour scale (or intensity) of that of the micro cracks observed in some of the specimens,
 371 divided by the whole pixels in the same area of interest and presented in Fig. 16. At W/B of 0.45, 0.55
 372 and 0.65, the least pore percentage was resulted for mixes with binder content of 450, 400 and 350,
 373 respectively. Therefore, these results confirm the mix design method and show that these mixes can
 374 present least porous matrixes which will have better durability properties for this new type of
 375 concrete.

376 **Conclusions**

377 This article shows that a rational mix design for alkali activated material (AAM) concretes can be
 378 achieved based on optimising packing fraction of the ingredient particles and giving consideration to
 379 the minimum required paste thickness to coat the aggregates for workability.

- 380 1. Alkali activated slag concretes designed in this way yielded higher slump for a given water
 381 content.
- 382 2. For each water to binder ratio there is an optimum amount of binder required; increasing the
 383 binder content beyond this point will not contribute to strength but will increase the
 384 workability, while mixes with less binder content are not workable.
- 385 3. AAS concretes with a minimum dosage of activators can be designed for different classes of
 386 workability and concrete strength grades from C16/20 to C32/40.
- 387 4. Typically, AAS concretes with a minimum dosage of activator require a binder content of 400
 388 kg/m³ and water/binder ratio of 0.55, to provide a cost-efficient workable normal C25/30
 389 concrete.
- 390 5. The binder contents selected in alkali activated slag concrete mixes designed based on wet
 391 packing fraction to get minimum water content are realistic, and do not cause high
 392 evaporation with the associated consequences of making large pores in the concrete.
 393 However, increasing the binder content can give a more porous structure for the product
 394 overall.

395 **Acknowledgements**

396 This work was supported by the United Kingdom Engineering and Physical Sciences Research
 397 Council (EPSRC) under grant EP/M003272/1, awarded jointly with NSFC (China).

398

399 Table 1: Oxide composition of GGBS used, from X-ray fluorescence analysis

Material	Component (mass% as oxide)
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	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	MgO	TiO ₂	Other	LOI*
GGBS	35.7	11.2	43.9	0.3	6.5	0.512	1.578	0.31

* LOI is loss on ignition at 1000°C.

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Table 2: Physical properties of GGBS

Fineness (particles $\geq 45 \mu\text{m}$)	7.74%
Particle density (tonnes/m ³)	2.86
Water absorption	35.14%

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Table 3: Physical properties of aggregates

Aggregates	Bulk specific gravity	Bulk Saturated surface dry (SSD) Specific gravity	Water Absorption (%)
Sand (0-4 mm)	2.72	2.73	0.75
Crushed Agg.(5-10 mm)	2.67	2.75	3.14
Crushed Agg.(10-16.5 mm)	2.60	2.67	2.60

Physical properties of sand and crushed aggregates were measured based on BS EN 1097-1.

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Table 4: Bulk and compacted packing fraction for various aggregate proportions

Aggregate proportions (Sand/10mm/16.5mm)	Bulk packing fraction (kg/m ³)	Compacted packing fraction
20/16/64	1.618	0.614
20/24/56	1.616	0.612
20/32/48	1.612	0.609
30/14/56	1.736	0.560
30/21/49	1.736	0.655
30/28/42	1.735	0.653
40/12/48	1.805	0.680
40/18/42	1.801	0.677
40/24/36	1.781	0.669

50/25/25	1.828	0.683
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Table 5: Baseline mix design and initial trial mixes for AASC

Mix No.	GGBS (kg/m ³)	NaOH (%)	WG (%)	Sand (kg/m ³)	Crushed Agg. (kg/m ³)	Blend ratios Sand/Fine Cr Agg. / Coarse Cr Agg.	W/B	Slump (mm)	Compressive strength (MPa)	
									7 days	28 days
Baseline-1*	320	4	6	756	1134	40-24-36	0.45	0	37.1	45.1
2*	320	4	6	756	1134	40-24-36	0.5	0	27.8	37.9

414 *For these mixes Na₂O=2.775%, SiO₂/Na₂O ratio (Ms) =0.324 and A/B=5.9

415

416 Table 6: Alkali activated slag concrete mixes with different binder content and water to binder ratio
417 for validation of mix design method

418

Mix Group No.	GGBS (kg/m ³)	Na(OH) (kg/m ³)	Na silicate (kg/m ³)	Sand (kg/m ³)	Crushed Agg. (5-10mm) (kg/m ³)	Crushed Agg. (10-16.5mm) (kg/m ³)	Water (kg/m ³)- W/B	Paste content	Excess of paste content to void content	Aggregate Mix proportion	Aggregate to binder ratio	90 days density (kg/m ³)
MG1	300	12	18	822	247	986	135-0.45	0.24	-0.25	40-12-48	6.85	2415
	300	12	18	789	237	947	165-0.55	0.27	-0.16	40-12-48	6.58	2410
	300	12	18	757	227	908	195-0.65	0.30	-0.06	40-12-48	6.31	2392
MG2	350	14	21	778	234	934	157.5-0.45	0.28	-0.12	40-12-48	5.56	2410
	350	14	21	741	222	889	192.5-0.55	0.32	-0.01	40-12-48	5.29	2400
	350	14	21	703	211	843	227.5-0.65	0.35	0.09	40-12-48	5.02	2367
MG3	400	16	24	735	221	882	180-0.45	0.32	0.00	40-12-48	4.6	2380
	400	16	24	692	208	830	220-0.55	0.36	0.13	40-12-48	4.33	2393
	400	16	24	649	195	779	260-0.65	0.40	0.25	40-12-48	4.05	2314.5
MG4	400	16	28	735	221	882	180-0.45	0.32	0.00	40-12-48	4.6	2365

	400	16	28	692	208	830	220-0.55	0.36	0.13	40-12-48	4.33	2377
	400	16	28	649	195	779	260-0.65	0.40	0.25	40-12-48	4.05	2354
MG5	450	18	27	692	208	830	202.5-0.45	0.36	0.13	40-12-48	3.84	2390
	450	18	27	643	193	772	247.5-0.55	0.41	0.27	40-12-48	3.57	2354
	450	18	27	595	178	714	292.5-0.65	0.45	0.41	40-12-48	3.30	2310

419

420 Table 7: Theoretical mix design based on maximum packing fraction of aggregates

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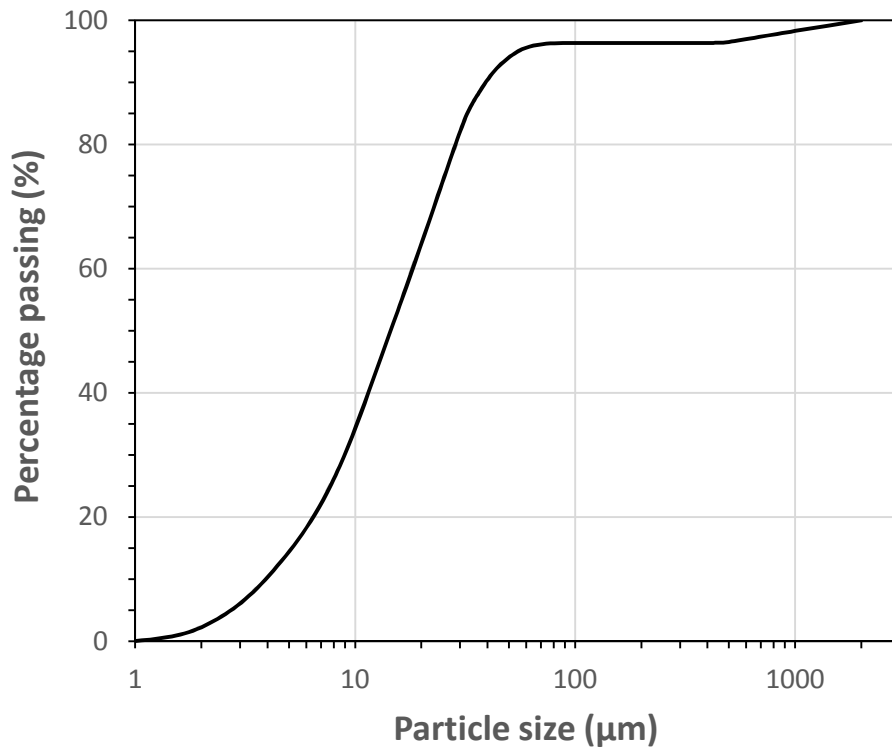
Mix No.	Aggregate proportion (Sand/10mm/16.5mm)	Aggregate packing fraction	Total solid volume of aggregate	Sand (kg/m ³)	Crushed Agg. (5-10mm) (kg/m ³)	Crushed Agg. (10-16.5mm) (kg/m ³)	W/B	Theoretical GGBS content (kg/m ³)	Density calculated by Emma Software (kg/m ³)
1	40-12-48	0.68	0.370	700.54	210.16	840.65	0.45	440	2.39
							0.55	391.11	2.36
							0.65	352	2.33
2	40-18-42	0.68	0.369	702.44	316.10	737.56	0.45	440	2.39
							0.55	391.11	2.36
							0.65	352	2.34
3	50-25-25	0.68	0.368	880.44	440.22	440.22	0.45	440	2.4
							0.55	391.11	2.37
							0.65	352	2.34

422

423 Table 8: Wet packing fraction of different mixes to get minimum water demand

Mix specification	Mixture ingredients properties	Slag	Sand	Fine Crushed	Coarse Crushed	Total wet packing fraction (WPF)
	Density (kg/m ³)					
	Wet packing fraction					
Mix 100GGBS	Mass (kg)	100	701	210	841	0.638
	Volume fraction	0.051	0.376	0.112	0.461	
Mix 200GGBS	Mass (kg)	200	701	210	841	0.646
	Volume fraction	0.097	0.358	0.106	0.439	
Mix 250GGBS	Mass (kg)	250	701	210	841	0.649
	Volume fraction	0.119	0.349	0.104	0.428	
Mix 300GGBS	Mass (kg)	300	701	210	841	0.651
	Volume fraction	0.139	0.341	0.101	0.418	
Mix 350GGBS	Mass (kg)	350	701	210	841	0.652

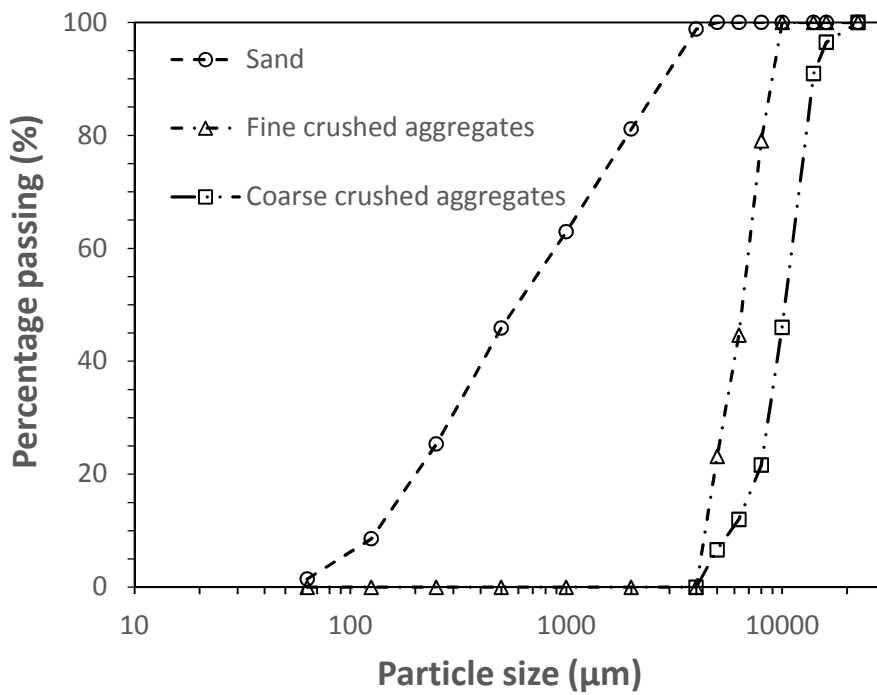
	Volume fraction	0.159	0.333	0.099	0.409	
Mix 400GGBS	Mass (kg)	400	701	210	841	0.652
	Volume fraction	0.177	0.326	0.097	0.400	
Mix 450GGBS	Mass (kg)	450	701	210	841	0.652
	Volume fraction	0.195	0.319	0.095	0.391	
Mix 500GGBS	Mass (kg)	500	701	210	841	0.651
	Volume fraction	0.212	0.312	0.093	0.383	



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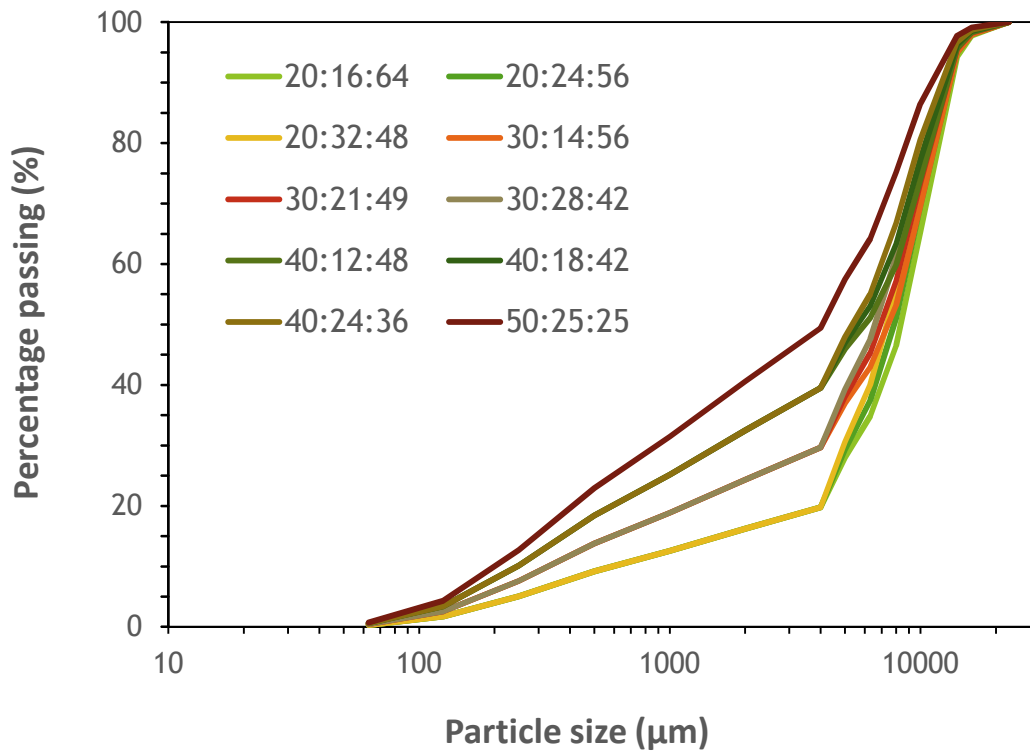
Fig. 1. Cumulative particle distribution of GGBS



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Fig. 2 Particle size distribution of sand and crushed aggregates

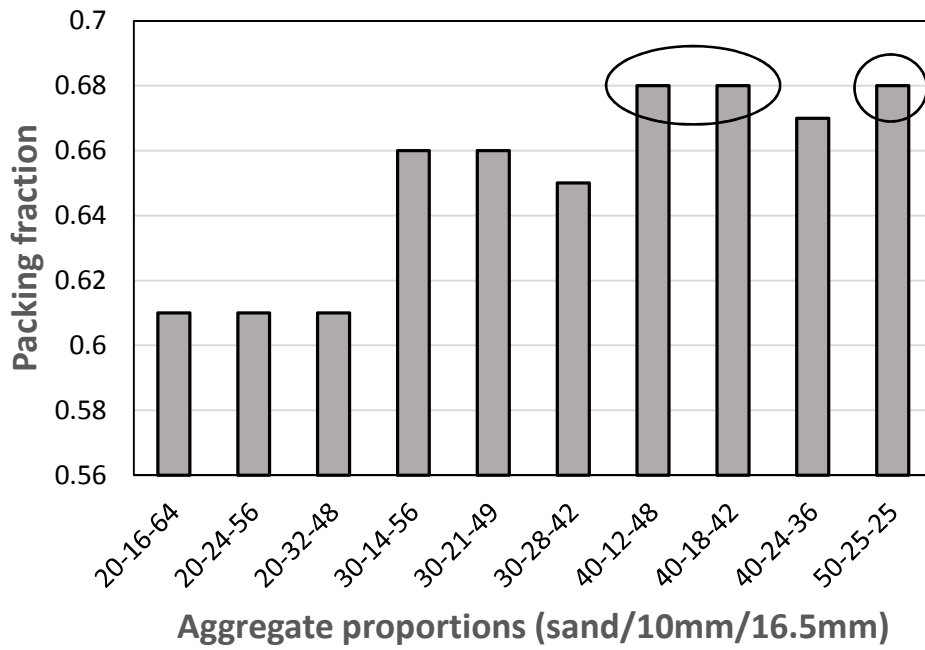


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Fig. 3 Particle size distributions for different mix proportions of aggregates (sand/10mm/16.5mm)

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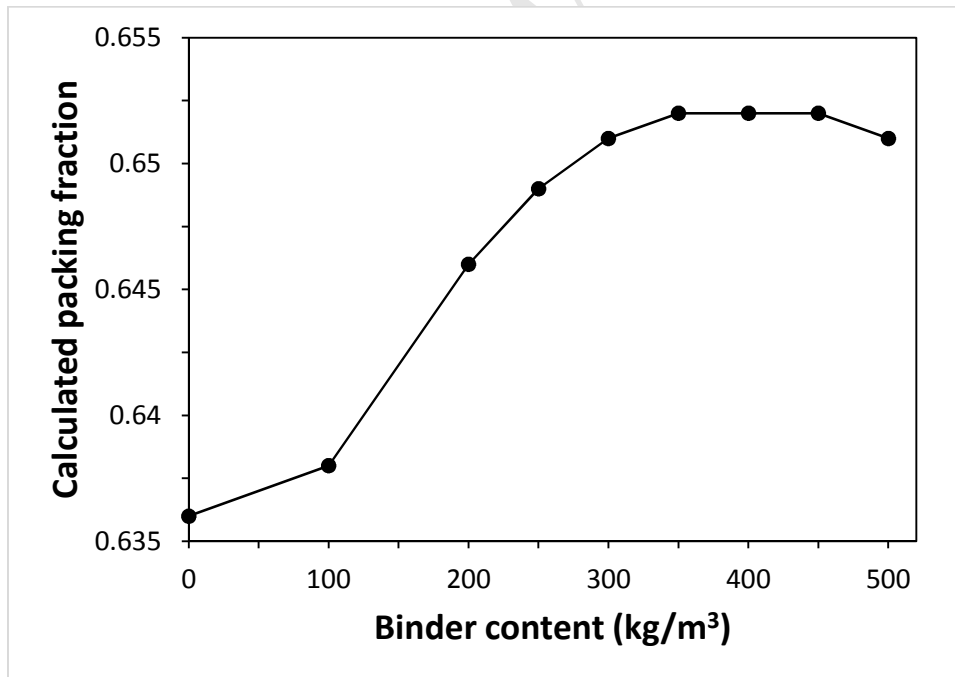


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Fig. 4 Packing fraction and void ratio resulted for different proportion of aggregates

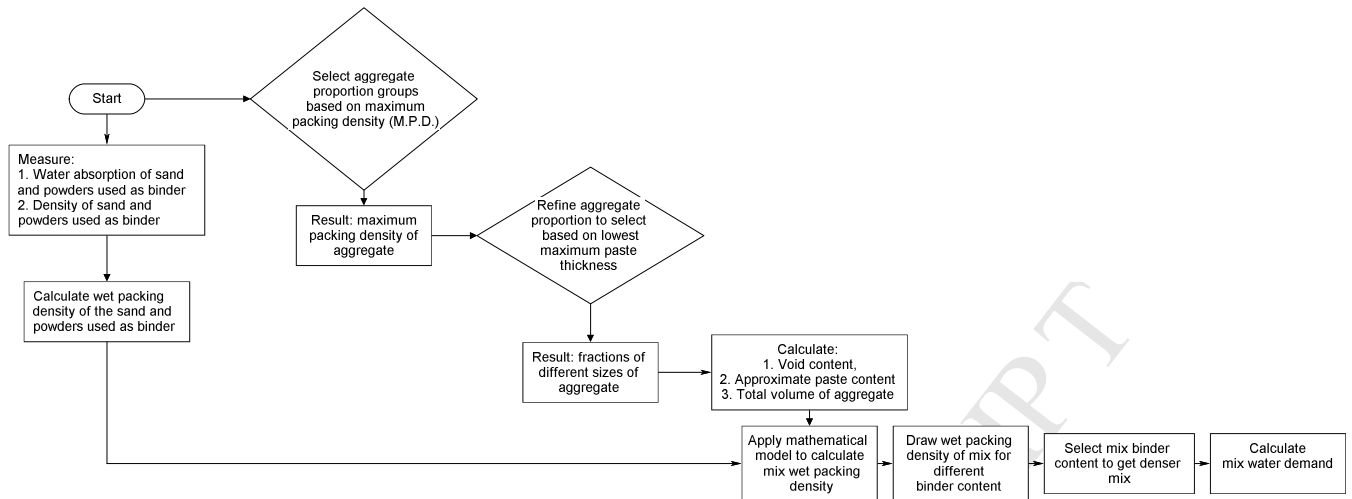
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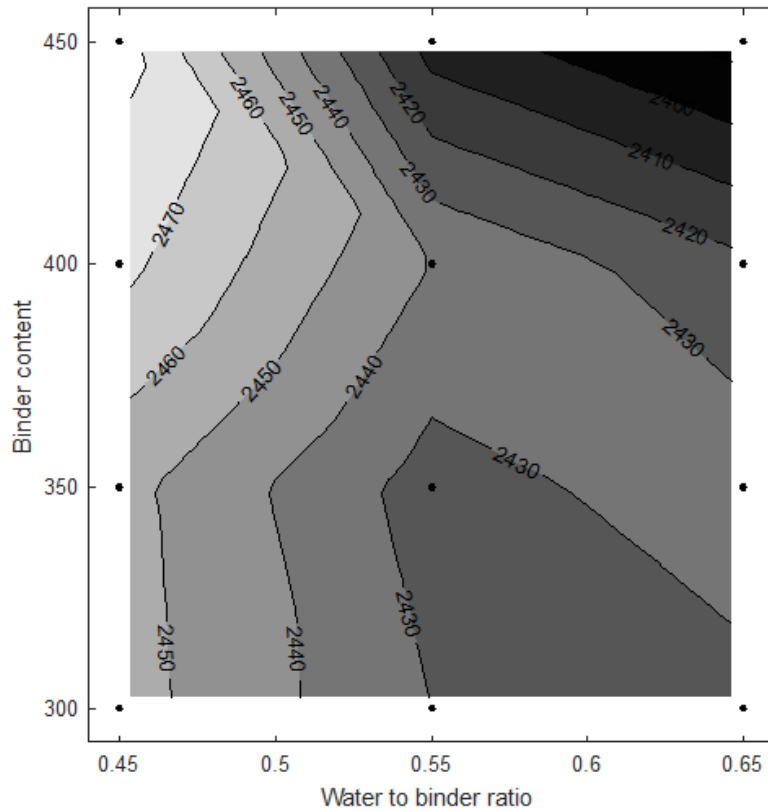
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Fig. 5 The relationship between the theoretical packing fraction and binder content of mixes



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Fig. 6 Mix design procedure for alkali activated concretes



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Fig. 7 Contour graph for wet density of mixes made with 4% NaOH & 6% WG, and different binder contents and water to binder ratios

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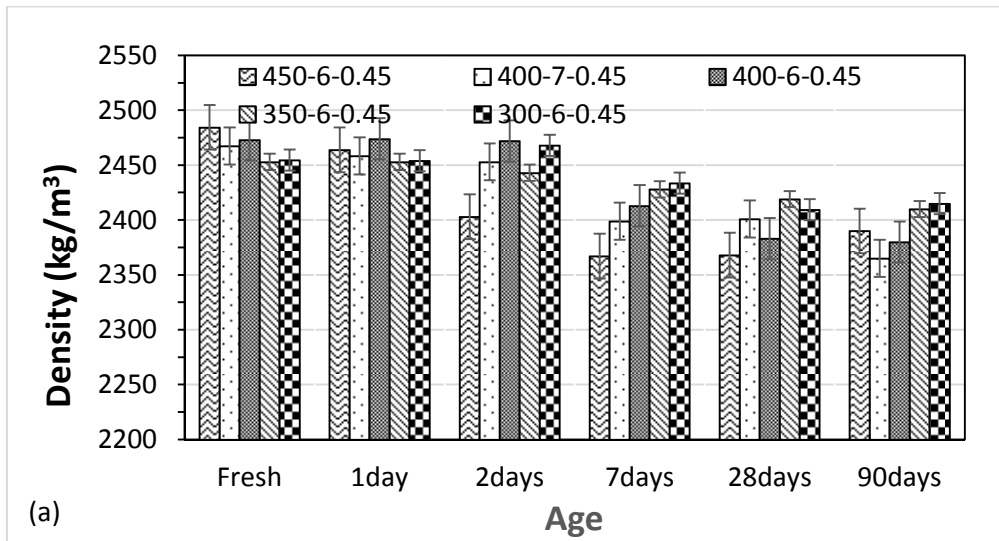
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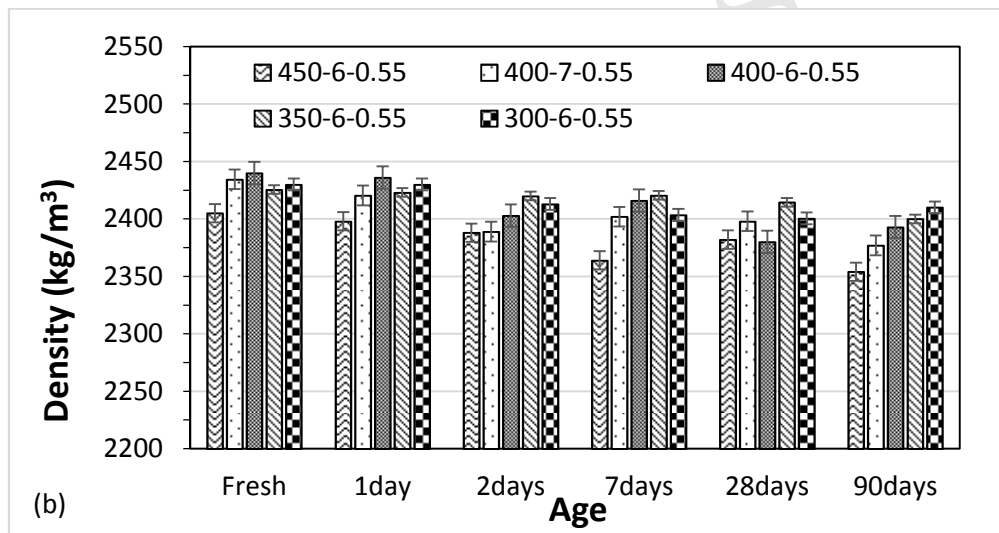
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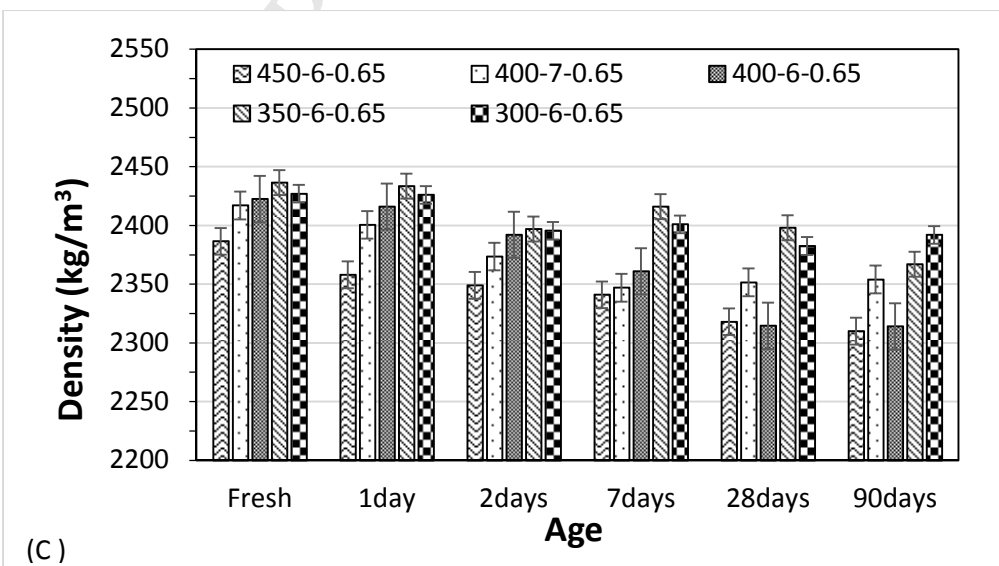
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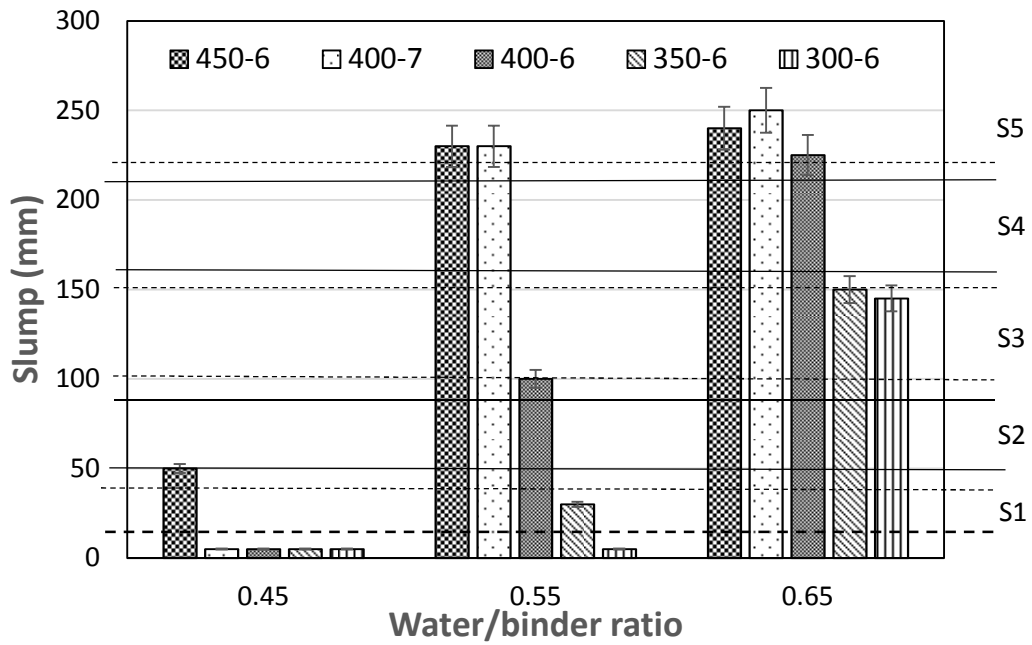


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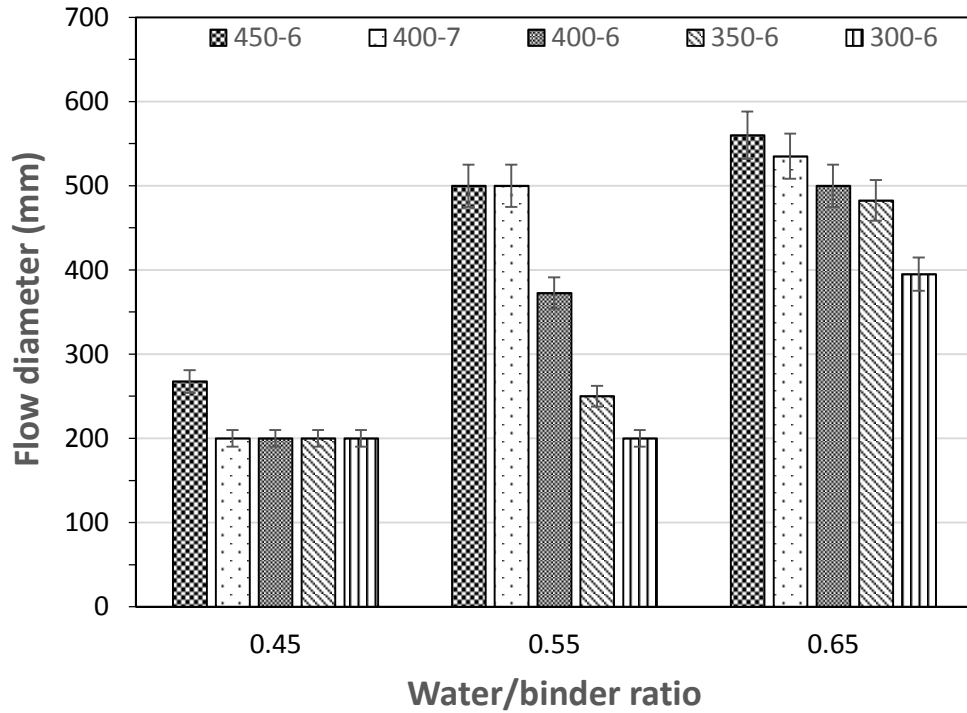
450 Fig. 8 Density of mixes at different ages (a) Water to binder ratio equal to 0.45; (b) Water to binder
 451 ratio equal to 0.55; (c) Water to binder ratio equal to 0.65



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453 Fig. 9 Slump of mixes with different binder contents and water/binder ratios (dashed and solid
 454 horizontal lines show minimum and maximum of slump for each classification)

455

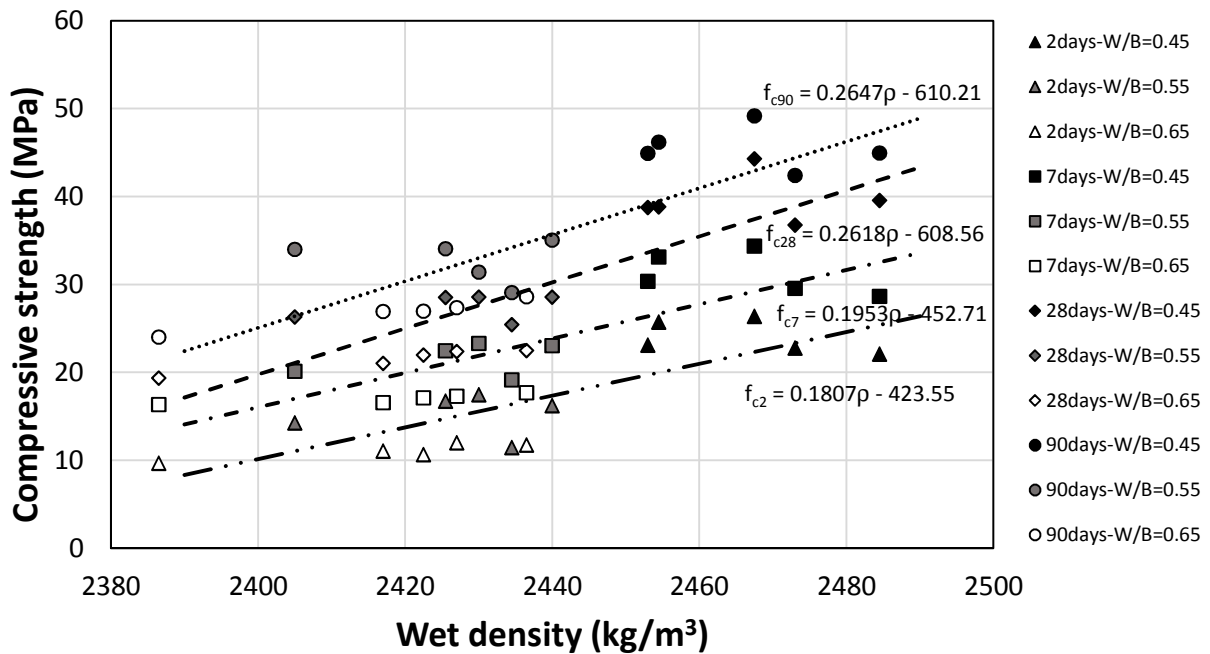


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Fig. 10 Flow results for mixes with different binder contents and water/binder ratios

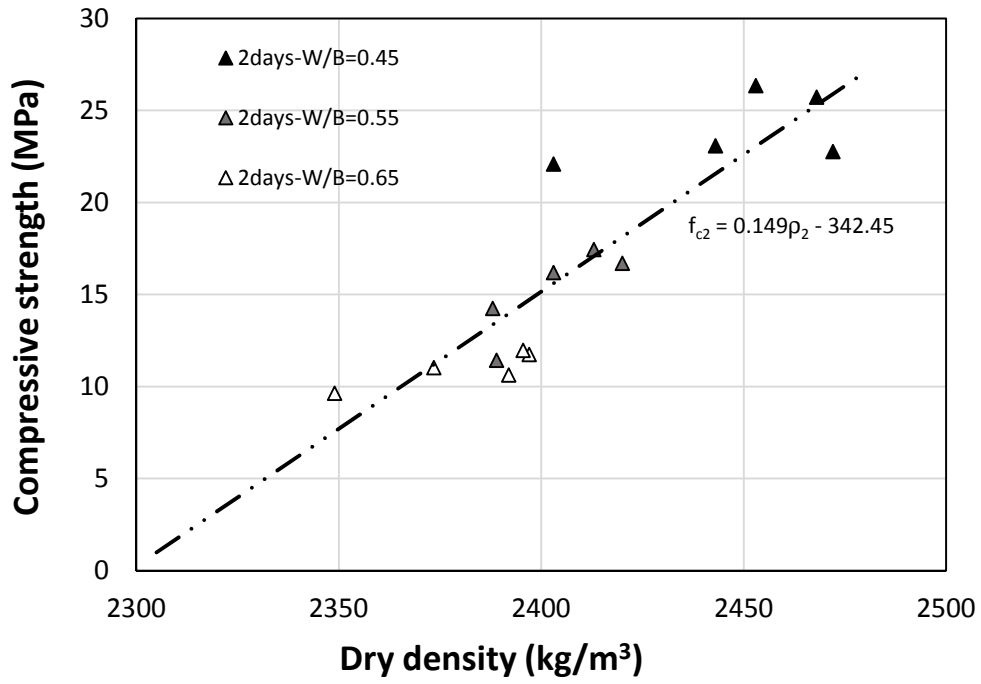
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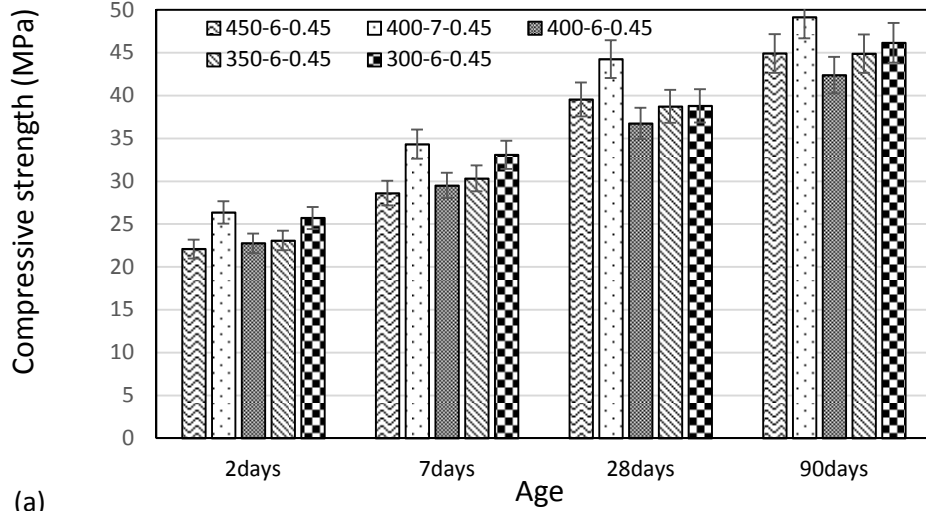
Fig. 11 Compressive strengths at different ages relative to wet density



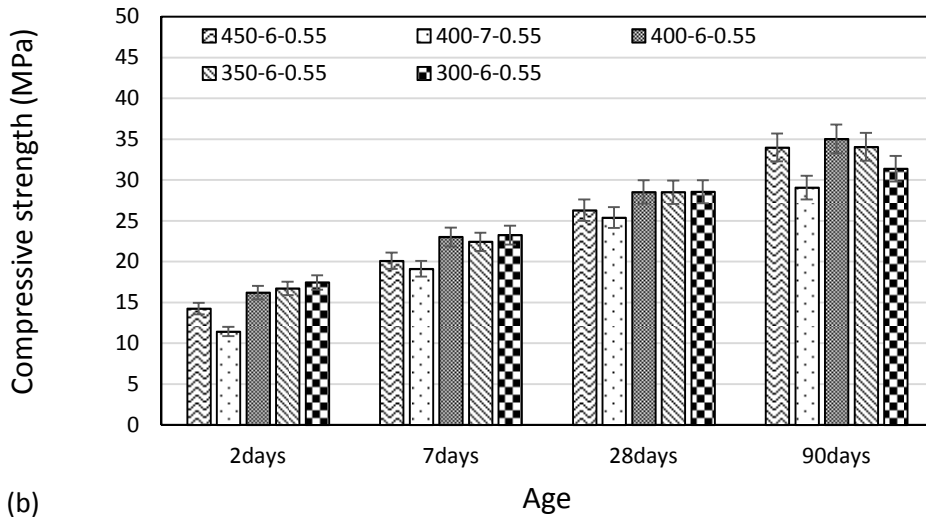
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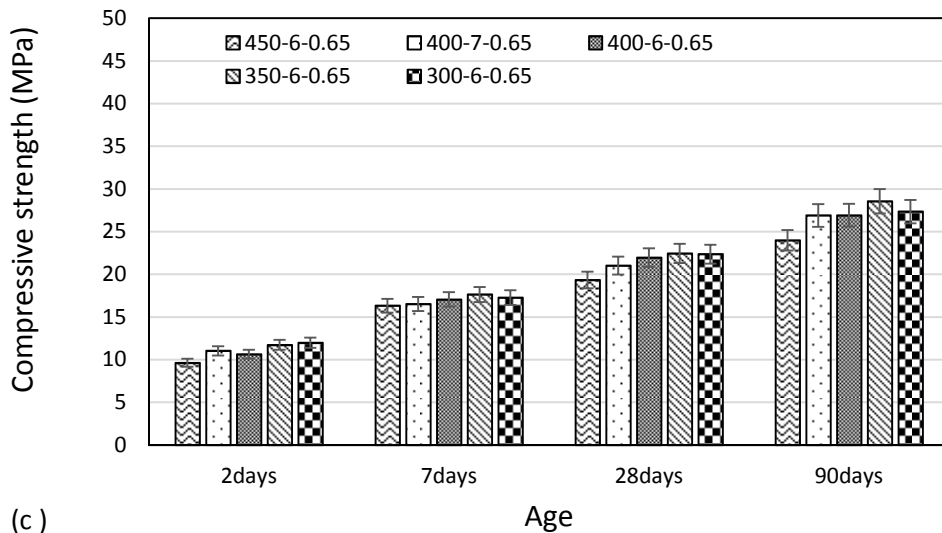
Fig. 12 Compressive strength at 2 days relative to dry density at the same age



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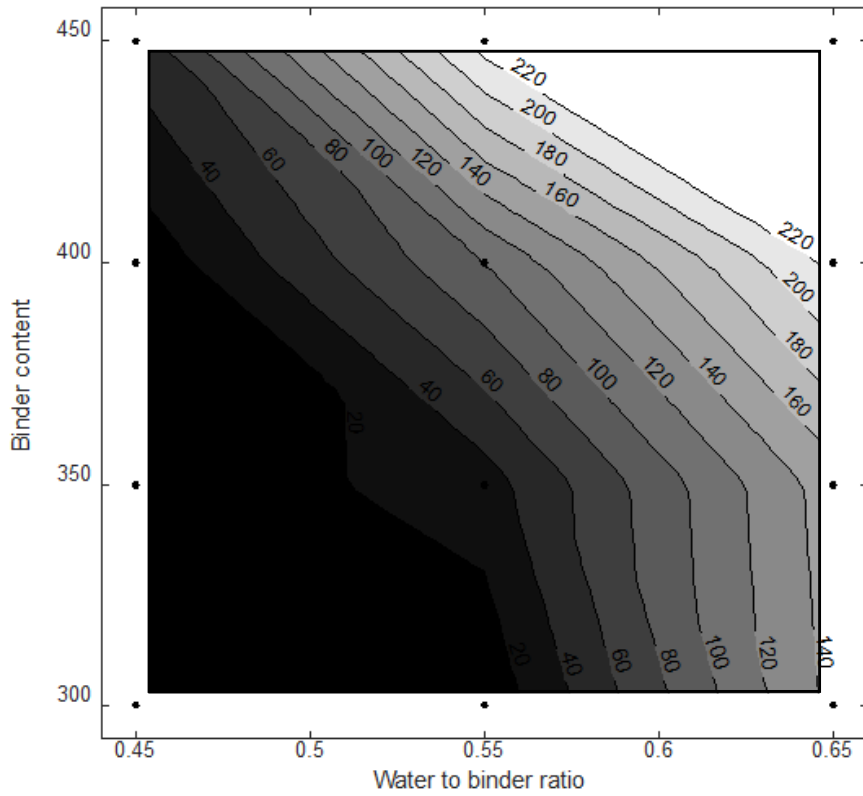
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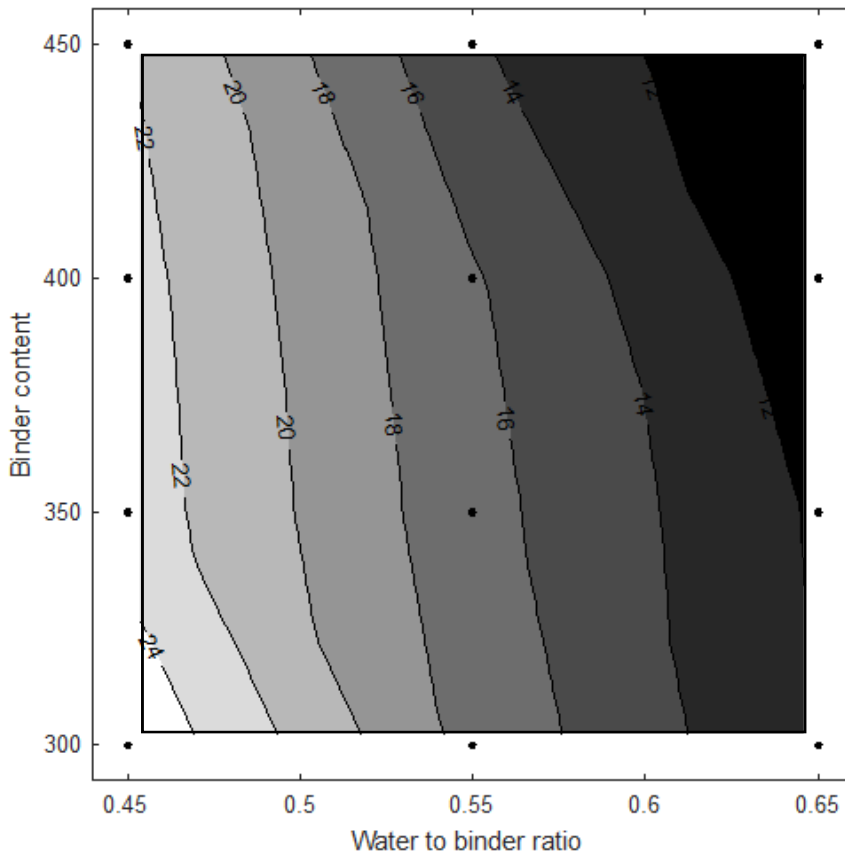
466 Fig. 13 Compressive strength of different mixes at different ages: (a) Water/binder ratio equal to 0.45;

467 (b) Water/binder ratio equal to 0.55; (c) Water/binder ratio equal to 0.65



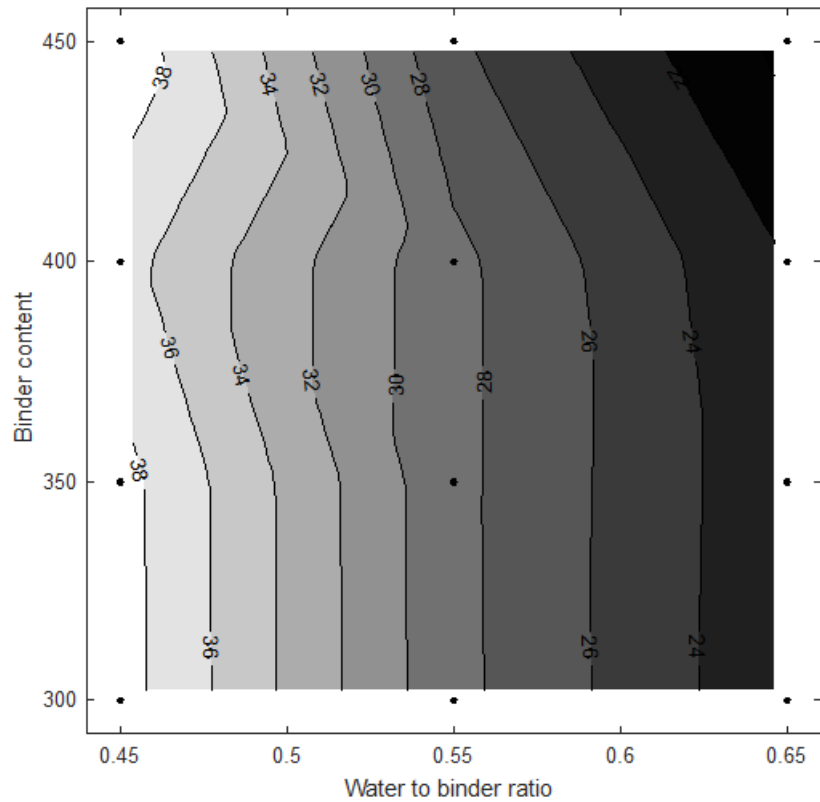
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(a) Slump



469

(b) 2-day compressive strength (MPa)



470

471

(c) 28-day compressive strength (MPa)

472 Fig. 14 Contour graphs for workability, early age (2 days) and 28-day compressive strengths of mixes
 473 made with the lowest amount of chemical activator (4% NaOH & 6% WG) and different binder
 474 contents and water to binder ratios

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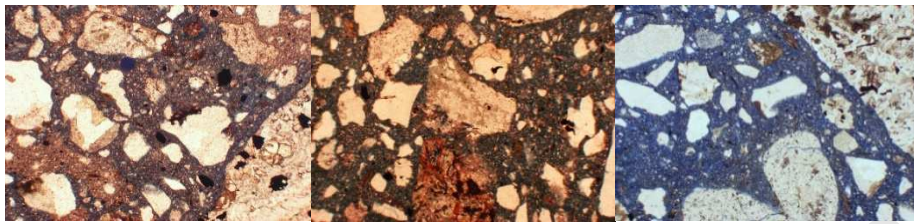
486

W/B=0.45

W/B=0.55

W/B=0.65

B=300



487

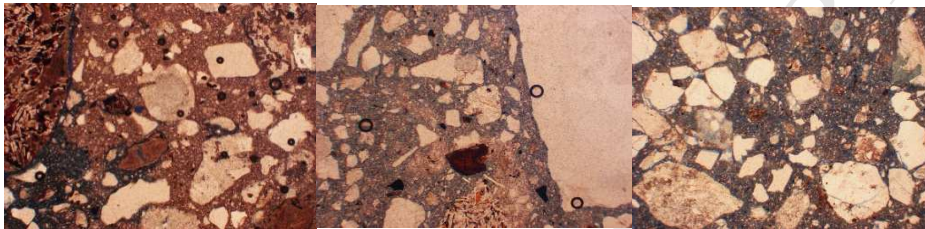
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(a)

(b)

(c)

B=350



489

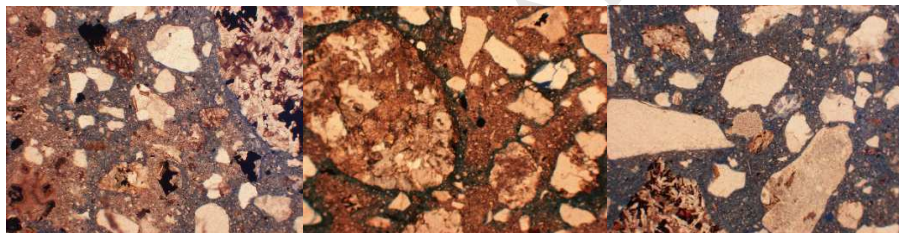
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(d)

(e)

(f)

B=400



491

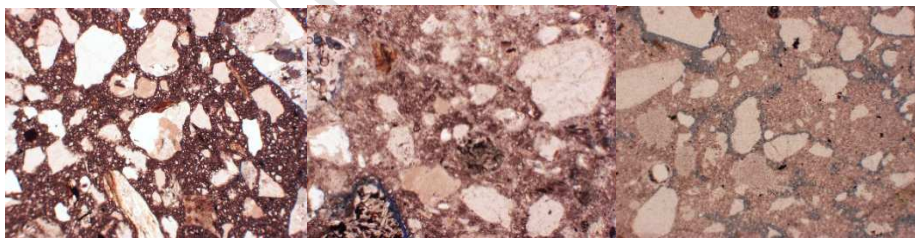
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(g)

(h)

(i)

B=450



493

494

(j)

(k)

(l)

495

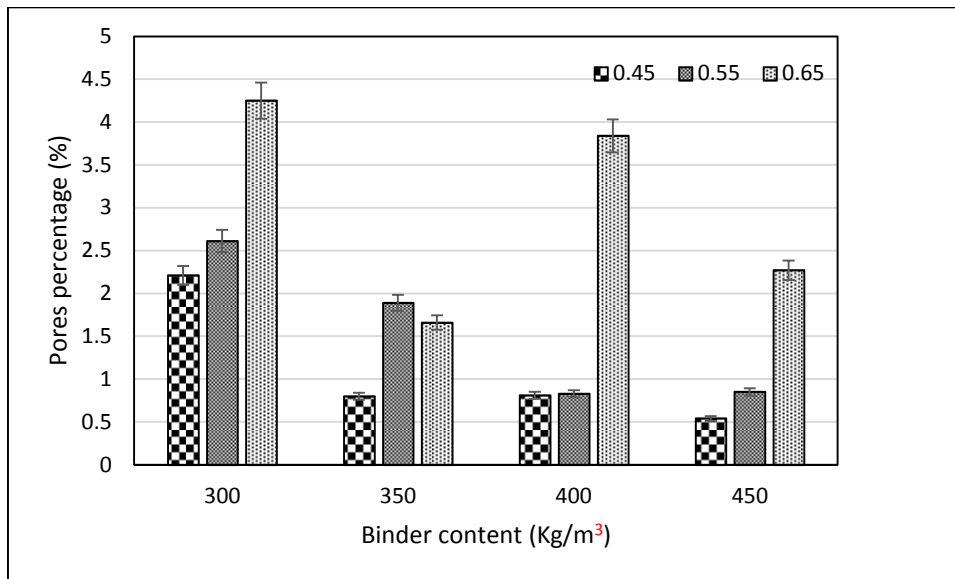
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Fig. 15 Thin section micrograph of samples of alkali activated slag concretes with different binder contents (B , kg/m^3) and water to binder ratios. Fields of view for all images are 1.1×0.82 cm.



500

501 Fig. 16 Pore percentages for mixes with different binder contents, and water/binder ratios as marked

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- Rational mix design for AAM concretes can be achieved based on optimising packing fraction of the ingredients.
- AASC designed based on optimising packing fraction of the ingredients has higher slump for a given water content.
- AASC with least dosage of activators can be designed for different workability and concrete grades up to C32/40.
- The binder contents selected in AASC designed based on WPF result in denser mixes as a good indicator of durability.