

Document downloaded from:

<http://hdl.handle.net/10251/146157>

This paper must be cited as:

Moraes, J.; Font-Pérez, A.; Soriano Martinez, L.; Akasaki, J.; Tashima, M.; Monzó Balbuena, JM.; Borrachero Rosado, MV.... (20-0). New use of sugar cane straw ash in alkali-activated materials: a silica source for the preparation of alkaline activator. *Construction and Building Materials*. 171:611-621. <https://doi.org/10.1016/j.conbuildmat.2018.03.230>



The final publication is available at

<https://doi.org/10.1016/j.conbuildmat.2018.03.230>

Copyright Elsevier

Additional Information

1 **New use of sugar cane straw ash in alkali-activated materials: a silica source for**
2 **the preparation of the alkaline activator**

3 J. C. B. Moraes¹, A. Font², L. Soriano², J. L. Akasaki¹, M. M. Tashima¹, J. Monzó², M. V. Borrachero²
4 and J. Payá^{2*}

5
6 ¹ Grupo de Pesquisa MAC – Materiais Alternativos de Construção. Universidade Estadual Paulista
7 (UNESP), Faculdade de Engenharia de Ilha Solteira, Campus de Ilha Solteira. Alameda Bahia, 550,
8 15385-000, Ilha Solteira/SP, Brasil.

9 ² GIQUIMA Group - Grupo de Investigación en Química de los Materiales de Construcción. Instituto de
10 Ciencia y Tecnología del Hormigón (ICITECH), Universitat Politècnica de Valencia, Valencia, Spain.

11 * Corresponding author: jjpaya@cst.upv.es; tel: +34 96 387 75 69.

12
13 **ABSTRACT**

14 Alkali silicates, expensive and highly pollutant chemical reagents, are required to produce the alkaline
15 activator for high-performance alkali-activated materials. This study presents a new silica source for
16 producing the alkaline activator, sugar cane straw ash (SCSA). An activating suspension was prepared
17 with SCSA and NaOH by means of a thermal bottle. The ash reacting time inside the thermal bottle (τ)
18 was assessed from 0–48 h, and the SCSA amount in suspension, represented by the $\text{SiO}_2/\text{Na}_2\text{O}$ ratio (ε),
19 was analysed from 0–1.82. Compressive strengths were obtained from blast-furnace slag-based mortars
20 that were cured for three days at 65 °C, with the optimal mortars produced when $\tau = 24$ h and $\varepsilon = 1.46$.
21 Comparison of these new SCSA systems with two common silica sources, sodium silicate chemical
22 reagent and rice husk ash, revealed that SCSA yielded lower results than the former and similar results to
23 the latter silica source.

24
25 **Keywords:** Conservation; Thermal Treatment, Microstructure, Compressive Strength, Alkali-Activated
26 Cement

27
28
29
30

31

32 1. INTRODUCTION

33

34 Alkali-activated materials (AAMs) are alternative binders to Portland cement. Their advantages over
35 Portland cement are lower energy consumption and reduced CO₂ emissions during the manufacturing
36 process [1,2]. AAMs are obtained by mixing an amorphous aluminosilicate (solid precursor) with an
37 alkaline activator (also called by activating solution or alkaline solution), where the product from this
38 chemical reaction hardens and forms a material with interesting mechanical properties [1,2]. Examples of
39 common solid precursors are blast-furnace slag (BFS), metakaolin and fly ash. The most common
40 reagents in alkaline solutions are alkali hydroxides and silicates [1,2].

41

42 The use of waste materials in AAM production has become a recent trend [3–5]. This study describes the
43 utilisation of sugar cane straw ash (SCSA), an ash rich in amorphous silica (SiO₂), in AAM production.
44 The management of this waste is a particular interest due the recent increase of sugar cane production in
45 Brazil. In 2014, this country produced 736.11 million tonnes of sugar cane were produced, which
46 accounts for ~40% of the worldwide production [6]. The potential of SCSA in AAM comes from prior
47 studies that assessed its reactivity as pozzolan in calcium hydroxide blends [7-9]. Authors observed that
48 the ash presented high reactivity [7,8], where the SCSA activity was also compared to a densified silica
49 fume [9]. The justification for such SCSA reactivity is due the presence of the amorphous phase (mainly
50 silica) observed in X-ray diffraction. In the tests carried out, SCSA consumed large amount of calcium
51 hydroxide to form C-S-H (calcium silicate hydrate) gels. This reaction product was observed in the
52 following tests: X-ray diffraction [7], thermogravimetric analysis [7,9], Fourier transform infrared
53 spectroscopy and electrical conductivity measurement [9]. As consequence for the high consumption of
54 calcium hydroxide observed in these studies, SCSA was utilised as partial replacement of the Portland
55 cement. Authors concluded that the replacement percentage range of 20-30% presented similar
56 mechanical behaviour than the control [10-12]. The pozzolanic characteristic of the SCSA was the reason
57 for such good results: the consumption of portlandite was observed by thermogravimetric analysis [10]
58 and X-ray diffraction [11]. Related to the use of SCSA in AAM, the ash was previous studied as solid
59 precursor in a binary system with blast furnace slag [13]. The samples of blast furnace slag and SCSA
60 presented higher compressive strength than the control since 3 days of curing at 25 °C. Authors observed

61 that the amorphous silica from SCSA improved the blast furnace slag matrix, explaining the great results.
62 Now, this study presents another use of SCSA in AAM production, with SCSA being utilised as a source
63 of silica in the production an alkaline suspension (alkaline activator).

64

65 The SiO_4^{4-} anion is a key component in high-performance AAMs, because it favours the formation of a
66 denser and stronger structure [1,2]. Sodium silicate (SS) currently provides an important reactive part of
67 silica that is widely used in combination with sodium hydroxide to produce the activating solution [1,2].
68 However, the main disadvantages of SS are mainly due that it is the most expensive raw material to
69 produce AAM and the highest emitter of greenhouse gases among the basic materials [14-17]. The cost of
70 the sodium silicate can be around of 20% to produce a AAM concrete, depending on the dosage. About
71 the greenhouse gases, authors found that emissions from the production of the sodium silicate represents a
72 50-70% of the total in an AAM concrete design. This high value is due the melting of the raw materials
73 (silica and sodium carbonate) until 1400 °C to produce the sodium silicate [17]. One of the key research
74 targets in the near future will thus be the assessment of new sources of silica to produce the activating
75 solution due the sodium silicate disadvantages [18,19]. One of the key research targets in the near future
76 will thus be the assessment of new sources of silica to produce the activating solution [20-25]. Results
77 from these studies about new silica source to the alkaline activator will be discussed in the Results and
78 Discussions section, where they will be compared to the use of SCSA to the same purpose.

79

80 The main objective of this study is to assess the potential of employing SCSA as the silica source to
81 prepare the alkaline suspension (alkaline activator). SCSA was utilised in combination with sodium
82 hydroxide to produce an aqueous NaOH/SCSA suspension by means of a thermal bottle. The resulted
83 blend of water, NaOH and SCSA were not filtered, resulting in a suspension and not in a solution. The
84 study is divided into three parts: 1) deriving the optimum time for dissolving the ash to prepare the
85 activating reagent; 2) understanding the influence of SCSA on the activating reagent; and 3) comparing
86 SCSA to commercial SS chemical reagents and alternative silica sources. After this study, it is expected
87 to produce an alkaline reagent with best SCSA content and the finest dissolution time in a thermal bottle.

88

89 **2. EXPERIMENTAL**

90

91 2.1. Materials and equipment

92

93 BFS was supplied by Cementval S.A., Puerto de Sagunto, Spain, in the form of large grains, which was
94 then milled in a ball mill for 30 min to obtain a fine material that could be used as the solid precursor in
95 AAM production. The chemical composition of BFS is shown in Table 1. BFS is primarily composed of
96 CaO, SiO₂, Al₂O₃ and MgO. Its mean particle diameter (D_{mean}) was 25.6 μm. Sodium hydroxide (NaOH,
97 or NH in the context of solution composition and specimens nomenclature) is the chemical reagent
98 utilised to prepare the alkaline solutions and suspensions. This material was used in form of pellets, and
99 was supplied by Panreac S.A., with a purity of 98%. Natural sand from Caolines Lapiedra (Liria,
100 Valencia, Spain), with a fineness modulus of 4.30, was utilised to prepare the mortars.

101

102 SCSA was utilised as the silica source to produce the activating suspension. The ash preparation process
103 consisted of the following steps. The straw was first collected from sugar cane plantations near Ilha
104 Solteira city (São Paulo, Brazil), and then transformed to ash by means of an autocombustion process.
105 This procedure took 6 h, and a maximum temperature of 700 °C was observed during the combustion
106 process. The ash was then sieved (MESH #50; 300 μm) to remove any unburned particles. To complete
107 the process, the sieved ash was milled in a ball mill for 50 min. This milling process was carried out to
108 reduce liquid absorption capability of the ash and, consequently, enhancing rheological properties of the
109 fresh AAM. Table 1 shows the chemical composition of SCSA, with SiO₂ (58.6 wt%), Al₂O₃ (9.0 wt%)
110 and Fe₂O₃ (8.4 wt%) being the most abundant compounds. $D_{mean} = 18.1$ μm after the milling process. The
111 amount of insoluble residue was also determined for SCSA (34 wt%).

112

113 RHA was another silica source utilised to produce the activating suspension. This ash served as a
114 comparison material to SCSA. RHA was supplied by DACSA S.A., Tabernes Blagues, Spain, and it was
115 utilised as received ($D_{mean} = 62.3$ μm). Its chemical composition is summarized in Table 1, with the main
116 oxide being SiO₂ (85.6 wt%). The RHA was not milled prior to use, because a previous study concluded
117 that the particle diameter of this ash did not influence the mechanical AAM properties [20]. Unlike RHA,
118 the use of unground SCSA presented rheological problems during the AAM preparation due to
119 considerable water adsorption, thus requiring SCSA to be milled to achieve the appropriate workability of
120 the samples.

121

122 Commercial SS was also utilised as a silica source. This chemical reagent was acquired from Merck, and
123 its chemical composition was 8 wt% Na₂O, 28 wt% SiO₂ and 64 wt% H₂O.

124

125

Table 1. Chemical composition of BFS, SCSA and RHA, in wt%.

Raw materials	SiO₂	Al₂O₃	Fe₂O₃	CaO	MgO	K₂O	SO₃	Cl	Others	LOI
BFS	29.9	10.6	1.3	40.2	7.4	0.6	1.9	-	2.6	5.5
SCSA	58.6	9.0	8.4	4.6	1.6	5.4	1.9	0.7	3.3	6.5
RHA	85.6	0.3	0.2	1.8	0.5	3.4	0.3	0.3	0.6	7.0

126

127 2.2. Preparation of alkali-activated materials

128

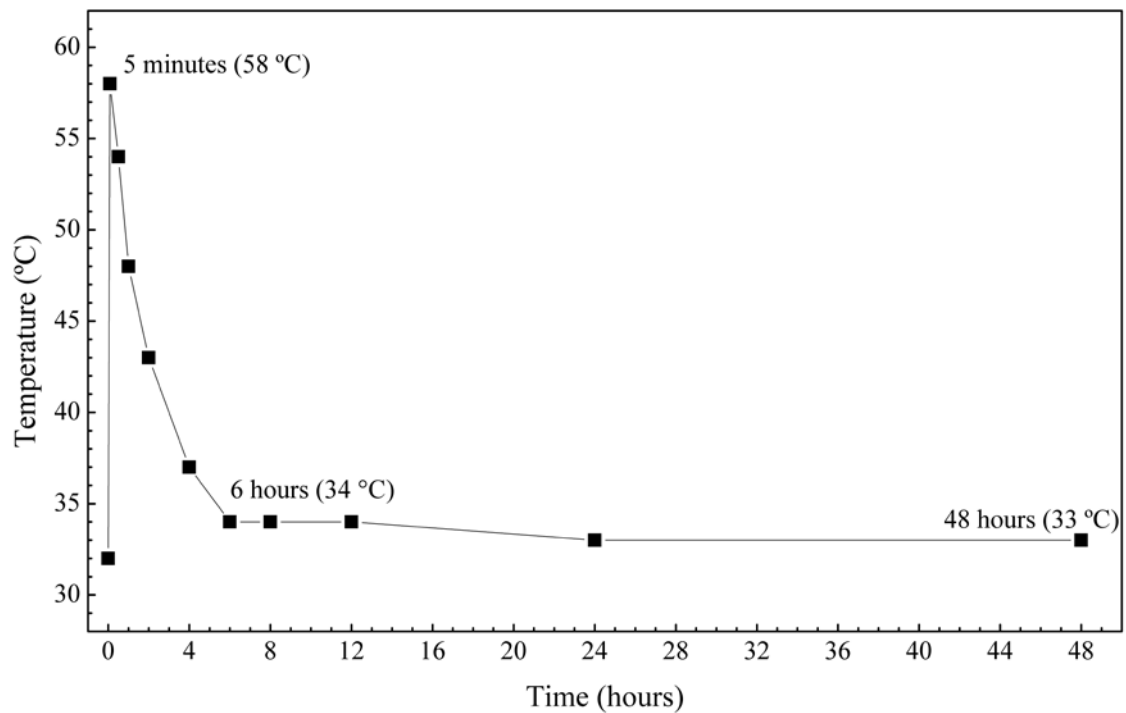
129 Three parameters were held constant during the AAM analysis, with the water/BFS ratio, sand/BFS ratio
130 (for mortars) and Na⁺ molality fixed at 0.45, 3.0 and 4 mol·kg⁻¹, respectively. The preparation of the
131 alkaline suspension and solution played an important role in this study, with two different suspensions
132 and two different solutions analysed: NH/SCSA, NH/RHA (both are suspensions), NH only (NH/-) and
133 NH/SS (both are solutions). The amount of SCSA, RHA and SS used to produce the alkaline solution is
134 represented in terms of the SiO₂/Na₂O molar ratio in the solution (ε). Here ε can only be varied by the
135 amount of silica source that is added to the activating solution, because the Na⁺ molality is fixed. Cases
136 with $\varepsilon = 0.73, 1.09, 1.46$ and 1.82 were assessed for the SCSA containing suspensions, whereas $\varepsilon = 1.46$
137 was the only case that was assessed for SS and RHA. $\varepsilon = 0$ was the only case assessed for the pure NaOH
138 solution.

139

140 The NH/SCSA and NH/RHA activating suspensions were produced by means of a thermal bottle. The use
141 of a thermal bottle was beneficial, because the NaOH dissolution in water releases heat, thus improving
142 the dissolution rate of the ash particles. The ash (SCSA or RHA) was first dry mixed with NaOH in the
143 thermal bottle, followed by the addition of water over the solids (the water/NaOH/SCSA mass ratio in g
144 was $202.5/32.4/x$, where $x = 29.5, 44.3, 59.0$ and 73.8 for $\varepsilon = 0.73, 1.09, 1.46$ and 1.82 respectively) and
145 the stirring of the solution for one minute. The thermal bottle was then sealed with a cap, and monitored
146 to determined time necessary to dissolve the ash (τ). Activating suspensions were obtained instead of

147 activating solutions for the instances where some of the ash would not dissolve. Here tests with $\tau = 0$ h (in
148 this case, the ash was added as the solid precursor together with BFS), 6 h, 24 h and 48 h were conducted
149 for SCSA, whereas $\tau = 24$ h was the test conducted for RHA. The temperature of the sodium hydroxide
150 solution ($4 \text{ mol}\cdot\text{kg}^{-1}$) inside the thermal bottle was also monitored during the production of the activating
151 suspension (Fig. 1). The water was $32 \text{ }^\circ\text{C}$ prior to being mixed with sodium hydroxide (0 h). The system
152 reached $58 \text{ }^\circ\text{C}$ after 5 min and started to decrease until 6 h, when the temperature of the suspension
153 reached a constant temperature of $\sim 35 \text{ }^\circ\text{C}$. The suspension was left in the thermal bottle until it returned to
154 room temperature after the thermal bottle treatment. The prepared suspensions were not filtered prior to
155 use. The solutions composed of NaOH and NaOH/SS were prepared by mixing the chemical reagents
156 with water in a common beaker. The beaker was then sealed with a plastic film, and the solution was left
157 to reach room temperature.

158



159

160 Fig. 1. Temperature evolution of the dissolution of sodium hydroxide (32.4 g) in water (202.5 g) inside
161 the thermal bottle.

162

163 Manufacturing of the mortars consisted of mixing BFS with the prepared suspensions or solutions until
164 proper homogenisation was attained (60 s). The prepared paste was then stirred with natural sand (150 s).
165 The resulting mortar was vibrated in a prismatic mould ($40 \times 40 \times 160 \text{ mm}^3$), and then stored under

166 controlled temperature (20 °C or 65 °C) and relative humidity (100%) conditions until the compressive
167 strength tests were conducted. The pastes produced for the microstructural analyses were made under the
168 same conditions as the mortars.

169

170 *2.3. Test procedures*

171

172 Compressive strength tests were carried out on mortars by means of a universal test machine, following
173 the UNE-EN 196-1 standard [26]. Pastes were assessed with XRD, FTIR, TGA and FESEM techniques.
174 XRD patterns were obtained by a Bruker AXS D8 Advance with a voltage of 40 kV, a current intensity of
175 20 mA and a Bragg's angle (2θ) in the 5–70° range, with a step of 0.02° at 2 s/step. FTIR spectra were
176 acquired with a Bruker Tensor 27 and analysed in the 400–4000 cm^{-1} range. TGA were performed with a
177 TGA Mettler-Toledo TGA 850, where the specimen was heated in a 70 μL alumina crucible to the 35–
178 1000 °C temperature range, at a heating rate of 20 °C·min⁻¹ and in the presence of a dry air atmosphere
179 (75 mL·min⁻¹ gas flow). The amount of sample utilised in TGA test was 35.0 ± 1.0 mg. Mass loss and
180 derivative curves (DTG curves) were extracted from the TG curve. FESEM images were taken using a
181 ZEISS Supra 55, with the fractured surface sample covered with carbon. The extra high tension was 20
182 kV and the working distance was 6–8 mm for the energy dispersive X-ray spectroscopy (EDS).

183

184 Mortars and pastes were assessed after three days of curing at 65 °C and 100% relative humidity. Mortars
185 and pastes were also assessed after 28 days of curing at 20 °C and 100% relative humidity for a series of
186 comparative compressive strength tests (see Section 3). Table 2 summarizes the tests performed and the
187 selected mixtures assessed for each test.

188

189 *2.4. Alkali-activated materials studies*

190

191 This study was divided into three parts to assess the potential of SCSA as a silica source in the
192 preparation of the activating suspension: 1) to obtain the optimum treatment time on leaving the ash in the
193 thermal bottle (τ); 2) to determine the optimal amount of SCSA in the activating suspension (ε); and 3) to
194 compare the NH/SCSA suspension to different silica sources (NH/SS (solution) and NH/RHA
195 (suspension) systems). Both τ and ε were determined from the compressive strengths of the mortars after

196 three days of curing at 65 °C. In the first part, four time intervals ($\tau = 0$ h, 6 h, 24 h and 48 h) were
 197 assessed, with $\varepsilon = 1.46$, to obtain the optimal τ . In the second part, the influence of the SCSA amount was
 198 assessed by varying the ε , with $\varepsilon = 0$ (solution with only NaOH), 0.73, 1.09, 1.46 and 1.82 (common ε
 199 values in the alkaline activation of BFS [27]), and maintaining a constant time ($\tau = 24$ h). In the third
 200 component, the NH/SCSA suspensions, with $\varepsilon = 1.46$ and $\tau = 24$ h, were compared to NH/SS and
 201 NH/RHA systems with the same ε and τ . Four silica mixtures were selected and manufactured into pastes
 202 to study their microstructures: two produced by the activating solution with NH/SCSA ($\varepsilon = 1.46$, $\tau = 0$ h
 203 and 24 h); one produced by a solution of NaOH ($\varepsilon = 0$); and one produced by a solution of NH/SS ($\varepsilon =$
 204 1.46).
 205
 206 The nomenclature adopted for these dosages is X- ε - τ , where X is the alkaline suspension or solution
 207 system (NH, SCSA, RHA or SS), ε is the SiO₂/Na₂O molar ratio and τ is only defined for the suspension
 208 with SCSA and RHA. Table 2 summarizes the three parts of the study, the dosages assessed and their
 209 respective nomenclatures.

210

211

Table 2. Mixtures and tests carried out.

Study Section	Activating Suspension or Solution	SiO ₂ /Na ₂ O molar ratio (ε)	Thermal bottle time (τ)	Nomenclature	Testing ages	
					Compressive strength tests	Microstructural tests (XRD, FTIR, TG and FESEM)
1	NH/SCSA	1.46	0	SCSA-1.46-0	3 days (65 °C)	3 days
			6	SCSA-1.46-6		-
			24	SCSA-1.46-24		3 days
			48	SCSA-1.46-48		-
2	NH/SCSA	0	-	NH-0	3 days (65 °C)	3 days
			0.73	SCSA-0.73-24		-
			1.09	SCSA-1.09-24		-
			1.46	SCSA-1.46-24		3 days
			1.82	SCSA-1.82-24		-
3	NH/SS	1.46	-	SS-1.46	3 days (65 °C) and 28 days (20 °C)	3 days
			24	SCSA-1.46-24		3 days
			24	RHA-1.46-24		-

212

213 3. RESULTS AND DISCUSSION

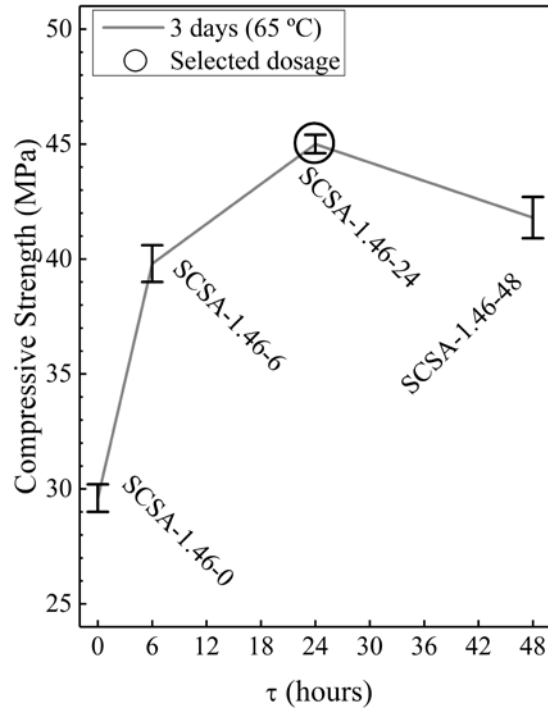
214

215 3.1 Effect of τ on SCSA/NaOH suspensions

216

217 The compressive strengths of SCSA-1.46-0, SCSA-1.46-6, SCSA-1.46-24 and SCSA-1.46-48 after three
218 days of curing at 65 °C are shown in Fig. 2. It is observed that τ had a crucial effect on the compressive
219 strength development of the mortar, with strengthening observed for $\tau \leq 24$ h. The SCSA-1.46-24 mortar
220 showed the highest compressive strength (45.0 MPa). These results appear to suggest that the use of
221 thermal bottle accelerated the silica dissolution from SCSA in the activating suspension. The increased
222 presence of SiO_4^{4-} anions in the alkaline suspension probably favoured the formation of more cementing
223 gel when it reacted with BFS. The high suspension temperature (maximum of 58 °C) observed during the
224 first 6 h (Fig. 1) favoured the SCSA dissolution, with a marked increase in compressive strength
225 development between SCSA-1.46-0 and SCSA-1.46-6. However, some SCSA dissolution must be carried
226 out due the high alkaline conditions after the suspension reached a constant temperature after 6 h, leading
227 to the increased compressive strength of SCSA-1.46-24. Alternatively, it may not be advantageous to
228 utilise the thermal bottle beyond $\tau = 24$ h, as the compressive strength of SCSA-1.46-48 (41.8 MPa) was
229 only slightly lower than that of SCSA-1.46-24. It was observed that the alkaline suspension produced
230 from SCSA for $\tau = 48$ h suffered gelification as reported for RHA [20]. This gel likely did not take part in
231 the formation of the AAM products, thus contributing to the decrease in compressive strength. Therefore,
232 it can be concluded that $\tau = 24$ h is the optimum thermal bottle time for SCSA.

233



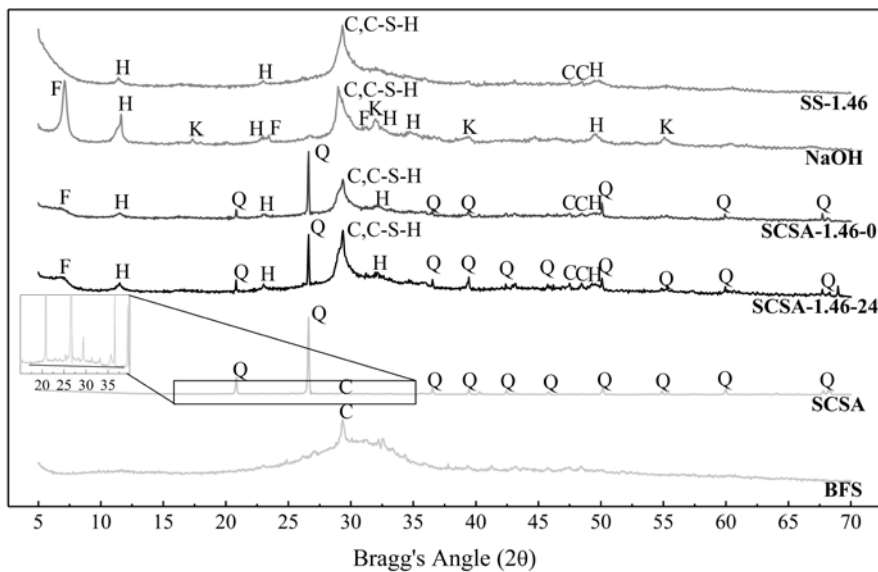
234

235 Fig. 2. Compressive strength of four mortars (cured at 65 °C for three days), with the activator prepared
 236 by thermal bottle treatment with $\varepsilon = 1.46$ and $\tau = 0$ h, 6 h, 24 h and 48 h.

237

238 The XRD patterns for the raw materials (BFS and SCSA) and the SCSA-1.46-24 and SCSA-1.46-0 pastes
 239 (cured after three days at 65 °C) are shown in Fig. 3. The baseline deviation observed in the
 240 diffractograms of the raw materials and pastes was due the presence of the amorphous phase in their
 241 compositions. The baseline deviation was observed in the Bragg's Angle range of 22–37° and 17–35° in
 242 the BFS and SCSA diffractograms, respectively. The baseline deviation of the ash was only detected after
 243 the enlargement of the diffractogram due the large quantity of quartz. There were no differences among
 244 the baseline deviations of the prepared pastes, suggesting that they all formed a similar gel of the form
 245 (N,C)-A-S-H [28]. Crystalline compounds were also observed on the diffractograms. Calcite (CaCO_3 ,
 246 PDF Card #050586; BFS and SCSA) and quartz (SiO_2 , PDF Card #331161; SCSA) were detected on the
 247 XRD patterns of the raw materials. Faujasite ($\text{Na}_2\text{Al}_2\text{Si}_4\text{O}_{12} \cdot 8\text{H}_2\text{O}$, PDF Card #391380), hydrotalcite
 248 ($\text{Mg}_6\text{Al}_2\text{CO}_3(\text{OH})_{16} \cdot \text{H}_2\text{O}$, PDF Card #140191) and a C-S-H phase (probably a (C,N)-A-S-H phase due to
 249 the presence of sodium and aluminium in the reaction) were detected on the XRD patterns of the pastes.
 250 The broad XRD peak associated with this gel overlapped the main calcite XRD peak. However, other
 251 peaks related to the presence of calcite were also observed, which indicates that some slight carbonation

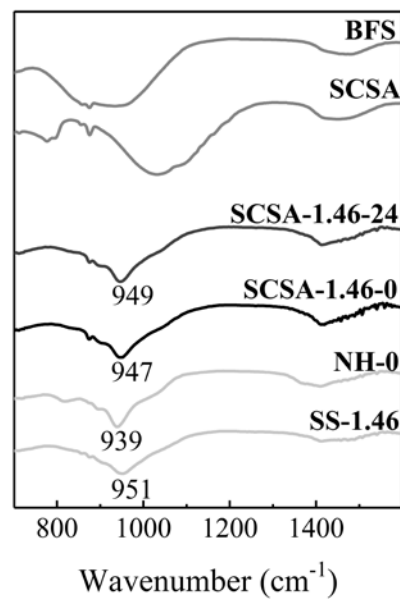
252 of the samples took place either during the curing process or in the sample handling. SCSA-1.46-24 and
 253 SCSA-1.46-0 were analysed in greater detail to determine the influence of τ . Whilst both pastes presented
 254 similar diffractogram patterns, some key differences were observed. Larger XRD peaks related to
 255 ~~faujasite and hydrocalcite were observed as τ increased.~~ The main band for the C-S-H gel in the 28.3–
 256 30.3° range also varied in size, with the paste prepared in the thermal bottle possessing stronger intensities
 257 than the sample produced by SCSA and without the bottle treatment. The areas under the main XRD
 258 peaks for quartz (A_Q) and C-S-H (A_{gel}) were measured with the OriginPro 8 graphing software package in
 259 the range of the analysed peak and without taking the baseline deviation into account. The area ratio, $\phi =$
 260 A_{gel}/A_Q , was calculated for both pastes. $\phi = 5.58$ for SCSA-1.46-24, whereas $\phi = 2.87$ for SCSA-1.46-0.
 261 Assuming the quartz fraction in SCSA was not dissolved during the preparation of the suspension and did
 262 not react with BFS, these ϕ values suggest the formation of more reaction products when $\tau = 24$ h.
 263



264
 265 Fig. 3. XRD patterns of the raw materials (SCSA and BFS) and pastes (SCSA-1.46-24, SCSA-1.46-0,
 266 NH-0 and SS-1.46) after three days of curing at 65 °C. Q: quartz; C: calcite; H: hydrocalcite; F: faujasite;
 267 C-S-H: calcium silicate hydrate; K: katoite.

268
 269 The FTIR spectra of the raw materials (BFS and SCSA) and pastes (SCSA-1.46-24 and SCSA-1.46-0)
 270 after three days of curing at 65 °C are presented in Fig. 4. The main bands detected in the spectra for the
 271 raw materials and pastes were in the wavenumber ranges of 800–1250 cm^{-1} (stretching mode of Si-O) and
 272 1400–1550 cm^{-1} (stretching mode of O-C-O bonds). The band in the 1400–1550 cm^{-1} range was due to

273 the presence of calcite in the raw materials and minimal carbonation of the samples, as observed in the
 274 XRD analysis. Here the most important wavenumber interval is 800–1250 cm^{-1} , because the main
 275 differences among the spectra are observed within this wavenumber range. BFS and SCSA possessed
 276 broad bands in the 800–1050 cm^{-1} and 900–1250 cm^{-1} wavenumber ranges, respectively, while the pastes
 277 showed narrower bands in the 900–1050 cm^{-1} interval, with a peak centred near 950 cm^{-1} . This behaviour
 278 is justified by the bonds breaking of the raw material particles and the subsequent formation of AAM
 279 products [29]. No significant differences were observed between the two pastes produced with SCSA,
 280 indicating that their gels are similar.
 281

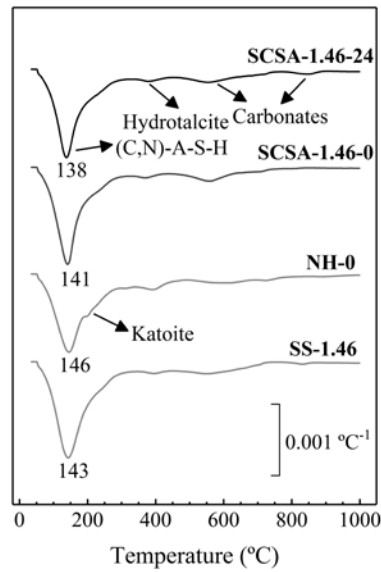


282
 283 Fig. 4. FTIR spectra of the raw materials (SCSA and BFS) and pastes (SCSA-1.46-24, SCSA-1.46-0,
 284 NH-0 and SS-1.46) after three days of curing at 65 °C.

285
 286 The TGA results are represented in Fig. 5 (DTG curves) and Table 3 (mass losses) for the SCSA-1.46-24
 287 and SCSA-1.46-0 pastes after three days of curing at 65 °C. Four peaks were observed on the DTG curves
 288 of these pastes at the following temperatures: 140 °C, 400 °C, 550 °C and 800 °C. The first peak, centred
 289 at 140 °C, was related to the decomposition of the (C,N)-A-S-H or N-A-S-H products. The peak at 400 °C
 290 was identified as the decomposition of hydrotalcite, and the peaks at 550 °C and 800 °C were related to
 291 the decomposition of carbonates [30]. Normally, carbonates decompose in the 700–850 °C temperature
 292 range. However, the formation of a poorly crystallised phase and the presence of sodium ions can cause a
 293 decrease in the decomposition temperature of the carbonates to 550 °C [30]. Table 3 shows the mass

294 losses for the two different temperature intervals to the importance of temperature on the formation of the
 295 reaction products: 35–300 °C (P_{35–300}) and 300–1000 °C (P_{300–1000}). Similar mass losses were also
 296 observed in the 35–300 °C temperature range (12.67% and 11.68% for SCSA-1.46-0 and SCSA-1.46-24,
 297 respectively), suggesting that the amount of combined water of the reaction products was in the same
 298 order of magnitude. This fact means that the differences in the compressive strengths are not directly
 299 related to the percentage of combined water, and may thus be analysed based on the distribution of
 300 products in the cementing matrix.

301



302

303 Fig. 5. DTG curves of the pastes (SCSA-1.46-24, SCSA-1.46-0, NH-0 and SS-1.46) after three days of
 304 curing at 65 °C (numbers close to main peaks are in °C).

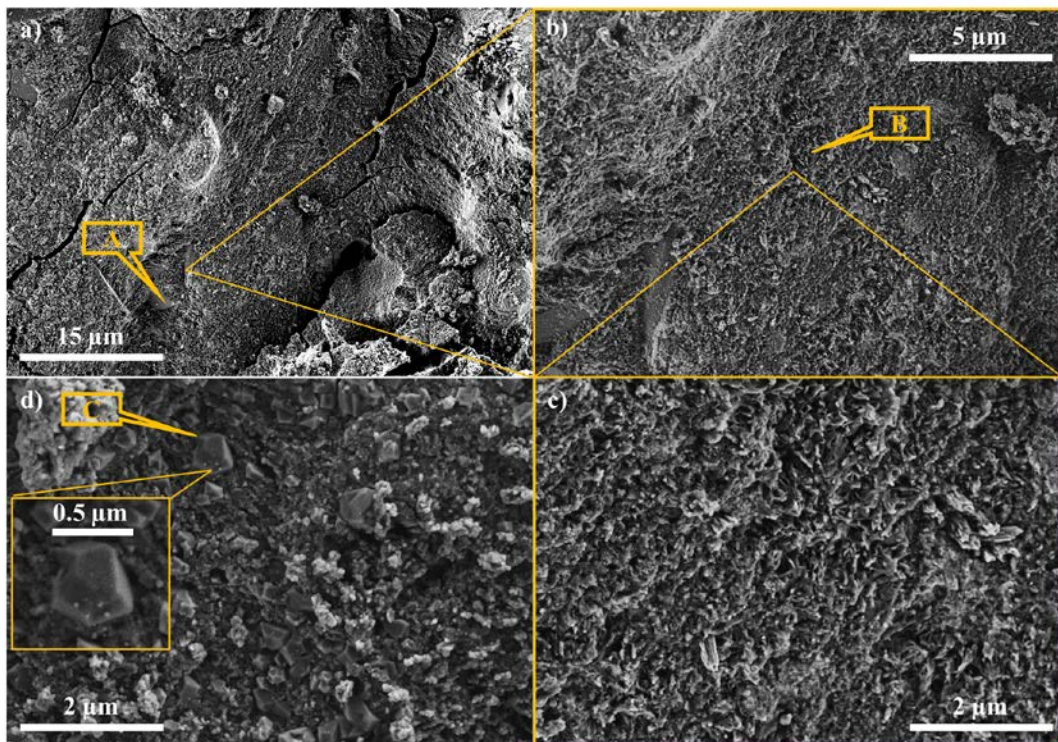
305

306 Table 3. Mass losses (%) of the SCSA-1.46-24, SCSA-1.46-0, NaOH and SS-1.46 pastes after three days
 307 of curing at 65 °C over the 35–300 °C (P_{35–300}) and 300–1000 °C (P_{300–1000}) test temperature intervals.

Specimen	Mass Losses (%)		
	P ₃₅₋₃₀₀	P ₃₀₀₋₁₀₀₀	TOTAL
SCSA-1.46-24	11.68	5.57	17.25
SCSA-1.46-0	12.67	5.06	17.73
NaOH	12.40	5.76	18.16
SS-1.46	14.50	5.19	19.69

308

309 The FESEM micrographs for the SCSA-1.46-24 paste after three days of curing at 65 °C are shown in
310 Fig. 6. A dense and amorphous structure was observed on the fractured surface paste (as suggested from
311 the XRD test; Fig. 6a). A compacted gel and unreacted particles of BFS (Spot A) were also identified. No
312 unreacted SCSA particles were found. The formed gels (Spot B) can be seen at a higher magnification in
313 Figs. 6b and 6c. They appear as very small and irregular particles (<0.2 μm in diameter) that are piled
314 against each other in a homogeneous pattern, yielding a low porosity matrix. EDS analysis of this gel
315 (seven points) yielded the following molar ratios: Ca/Si = 0.95 ± 0.07 , Al/Si = 0.32 ± 0.03 , Na/Al = 1.34
316 ± 0.05 and Mg/Si = 0.13 ± 0.05 . Table 4 shows the EDS results represented by molar contents for each
317 element utilised to calculate these ratios from spots B. This gel is considered to be a (C,N)-A-S-H
318 product. Some potassium (K) was detected in EDS results, where it comes from the chemical composition
319 from the SCSA. The magnesium observed in the EDS analysis can be related to both possibilities: EDS
320 detected an unreacted slag under the formed products or the presence of hydrotalcite among the gels [31].
321 Fig. 6d highlights some microcrystals (Spot C) that are distributed in the gel matrix (probably faujasite
322 crystals).
323



324
325
326
327

Fig. 6. FESEM micrographs of the SCSA-1.46-24 paste.

328
 329
 330
 331

Table 4. Molar content (in %) of the elements at points related to Spot B from SCSA-1.46-24 paste.

Spot	Element (molar, %)						TOTAL (%)
	Si	Al	Na	Ca	K	Mg	
Spot B – 1	34.89	10.24	14.40	35.00	1.35	4.12	100
Spot B – 2	33.88	12.21	14.42	31.00	2.12	6.37	100
Spot B – 3	33.34	10.44	14.14	35.36	1.28	5.44	100
Spot B – 4	35.32	12.49	14.27	31.22	0.97	5.73	100
Spot B – 5	36.89	10.45	16.45	33.13	1.46	1.61	100
Spot B – 6	37.18	10.54	14.18	33.49	1.49	3.12	100
Spot B – 7	34.89	10.24	14.40	35.00	1.35	4.12	100

332
 333
 334
 335
 336
 337
 338
 339
 340
 341
 342
 343

The FESEM images for the SCSA-1.46-0 paste after three days of curing at 65 °C are shown in Fig. 7. A dense structure, similar to SCSA-1.46-24, is observed at Spot D on Fig. 7a. Fig. 7b shows a quartz particle from SCSA in Spot E surrounded by gel. The magnifications of Fig. 7a are shown in Figs. 7c and 7d. The formed gel is also composed of small hydration products, similar to those found on SCSA-1.46-24. The molar ratios of the gel on SCSA-1.46-0 were: $Ca/Si = 0.92 \pm 0.06$, $Al/Si = 0.33 \pm 0.01$, $Na/Al = 1.86 \pm 0.12$ and $Mg/Si = 0.18 \pm 0.05$. The Ca/Si , Al/Si and Mg/Si ratios on SCSA-1.46-0 were similar to those observed on SCSA-1.46-24. Table 5 presents the molar content for each element that was obtained to calculate the molar ratio values from Spot D. The largest difference was between the two pastes was the Na/Al ratio of the formed gel, despite both pastes possessing the same composition.

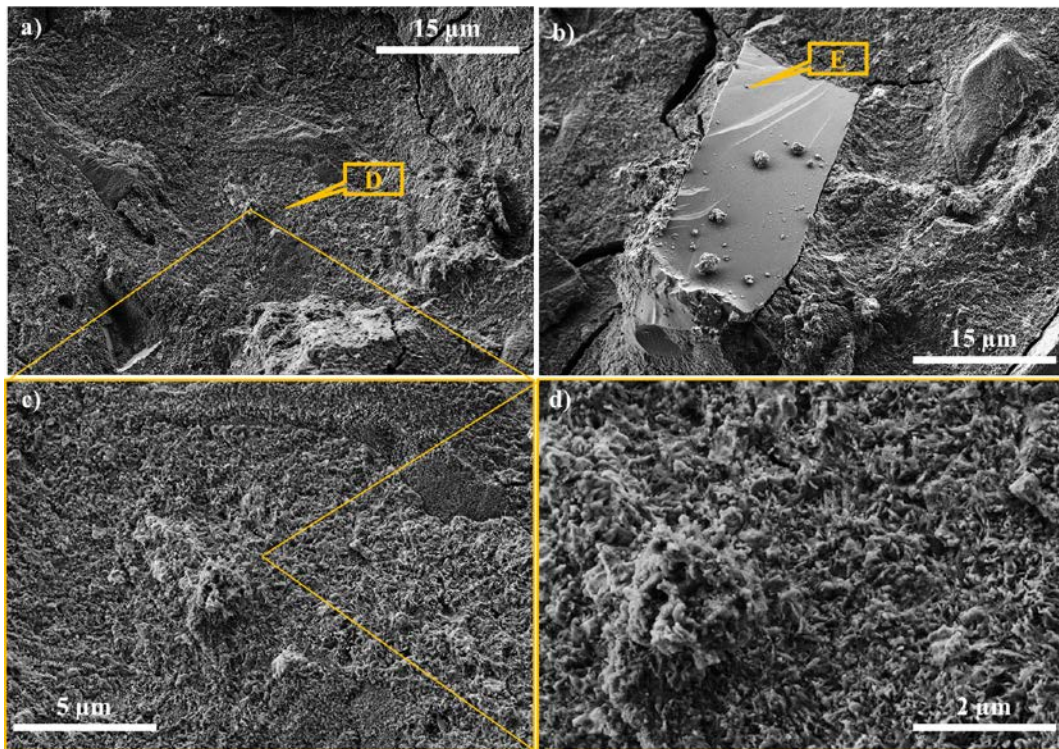
Table 5. Molar content (in %) of the elements at points related to spots D from SCSA-1.46-0 paste.

Spot	Element (molar, %)						TOTAL (%)
	Si	Al	Na	Ca	K	Mg	
Spot D – 1	32.10	10.95	19.30	28.31	1.77	7.58	100
Spot D – 2	32.82	10.56	21.07	29.20	2.07	4.29	100
Spot D – 3	31.75	10.38	19.06	31.33	1.78	5.70	100

344

345 It has been reported that the strength development of AAMs depends on the ε value of the activating
346 solution, with the low compressive strength of BFS systems attributed to cases where $\varepsilon < 1.0$ [32]. The
347 theoretical ε value was 1.46 in both SCSA pastes. However, there was less solubilised silica in SCSA-
348 1.46-0, which means that the real ε of SCSA-1.46-0 is lower than that for SCSA-1.46-24. More Na^+ is
349 thus incorporated into the formed gel in this situation, which significantly lowers the strength of the
350 sample (34.2% lower strength; see Fig. 2). Therefore, the chemical composition of the formed gel formed
351 makes a critical contribution to the strength of the matrix.

352



353

Fig. 7. FESEM micrographs of the SCSA-1.46-0 paste.

354

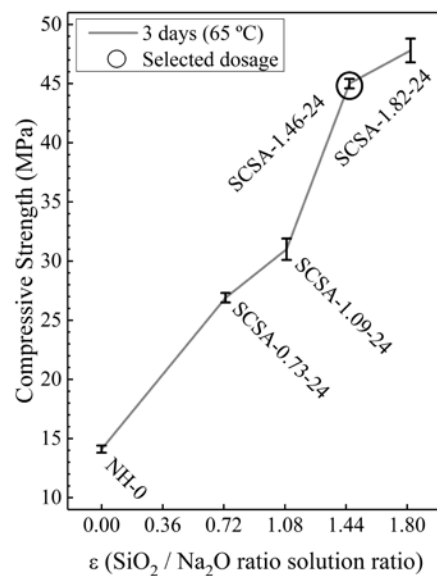
355

356 3.2. Effect of ε on the SCSA/NaOH suspensions

357

358 The compressive strengths of NH-0, SCSA-0.73-24, SCSA-1.09-24, SCSA-1.46-24 and SCSA-1.82-24
359 after three days of curing at 65 °C are shown in Fig. 8. The SCSA amount, which is represented by ε ,
360 clearly influences the compressive strength of the mortars at a constant τ (24 h). The increase in ε also
361 generates an increase in the compressive strength of mortars. A noticeable result is that all the ash

362 samples (compressive strength results in the 26.9–47.8 MPa range) yield significantly better results than
 363 the mortar with only NaOH (14.1 MPa). As expected, the dissolved SiO_4^{4-} from SCSA promotes the
 364 formation of a stronger structure. The dosage selected for the final component of the analysis was SCSA-
 365 1.46-24. Whilst SCSA-1.82-24 (47.8 MPa) possessed a slightly higher compressive strength than SCSA-
 366 1.46-24 (45.0 MPa), the latter mortar presented better rheological properties than the former one.
 367
 368 Correlation of the compressive strength results in Figs. 2 and 8 highlights the important role of SCSA in
 369 the reaction process without the thermal bottle. SCSA-1.46-0 possessed a compressive strength that was
 370 113% greater than that of NH-0 (30.0 and 14.1 MPa, respectively; Fig. 8). Furthermore, SCSA-1.46-0
 371 possessed a higher compressive strength than SCSA-0.73-24 (26.9 MPa) and a similar value to SCSA-
 372 1.09-24 (31.0 MPa). These results demonstrate that a sample produced without the thermal bottle can
 373 possess a similar (or even higher) compressive strength when compared to a sample produced with a
 374 thermal bottle. However, when mixtures with the same ε value and different τ values are compared (e.g.,
 375 SCSA-1.46-0 and SCSA-1.46-24), the samples produced with the thermal bottle possessed higher
 376 compressive strengths than those produced without it, as observed in Fig. 2.
 377



378
 379 Fig. 8. Compressive strength of the mortars after three days of curing at 65 °C, with the activator prepared
 380 by thermal bottle treatment and $\varepsilon = 0$ (only NaOH), 0.73, 1.09, 1.46 and 1.82.
 381

382 The XRD results highlight the key differences between the NaOH and SCSA-1.46-24 pastes (Fig. 3). The
383 NaOH sample possessed more intense peaks of the crystalline phases (faujasite and hydrotalcite) than the
384 SCSA sample, with the formation of katoite also observed in the NaOH sample. Previous studies have
385 also noted the high formation of crystalline compounds, with more ordered phases produced by a pure
386 NaOH solution than a solution that is a combination of sodium hydroxide and a silica source (e.g., SS)
387 [33,34]. The advanced formation of these ordered structures supports the lower compressive strength
388 results of the NaOH samples.

389
390 FTIR analysis revealed that the main Si-O peak of the SCSA paste was located at a higher wavenumber
391 than the NaOH paste (Fig. 4). The increase in the silica content of the SCSA system in the mixture likely
392 produced a stronger gel, thus causing the mortars produced with SCSA to possess higher compressive
393 strengths.

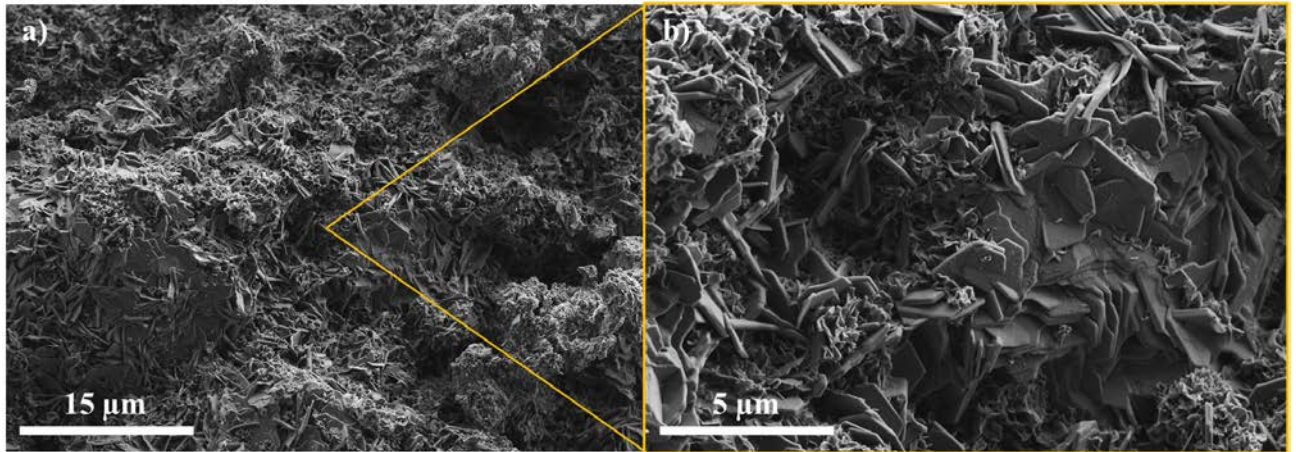
394
395 The TGA studies showed that the NaOH sample underwent a mass loss similar to the other pastes, even
396 though it possesses a lower compressive strength (Fig. 5 and Table 3). XRD analysis of the NaOH sample
397 indicated the strong formation of crystalline compounds, such as faujasite and katoite
398 ($\text{Ca}_3\text{Al}_2(\text{SiO}_4)(\text{OH})_8$, PDF Card #380368), that decompose at $\sim 140^\circ\text{C}$ and $\sim 200^\circ\text{C}$, respectively. The
399 decomposition of alkali-activated materials gels was also observed at these temperatures. A bulk of the
400 mass loss of the NH-0 sample in the $35\text{--}300^\circ\text{C}$ temperature range is likely related to the crystalline phase
401 decompositions, with dehydration due to the cementing gels having a secondary effect. This observation
402 explains why the mass loss of the NH-0 specimen was comparable to the other pastes, even though the
403 compressive strength was very low.

404
405 Fig. 9 shows the FESEM micrographs of the NH-0 sample after three days of curing at 65°C . The
406 structure of the NH-0 paste was more crystalline and porous than the SCSA pastes. This kind of structure
407 was already expected due to our prior analyses. The molar ratios of the gel on NH-0 were: $\text{Ca/Si} = 1.18 \pm$
408 0.02 , $\text{Al/Si} = 0.38 \pm 0.02$, $\text{Na/Al} = 1.89 \pm 0.15$ and $\text{Mg/Si} = 0.07 \pm 0.01$. Table 6 presents the molar
409 content for each element obtained from EDS results. The Ca/Si ratio for NH-0 was higher than those from
410 the SCSA pastes (1.18 vs. 0.92–0.95). It is clearly noticed that the NH-0 paste possessed less silicon than

411 the SCSA samples, which probably led to the formation of a more crystalline, porous and softer structure
 412 with a lower compressive strength.

413

414



415

416

Fig. 9. FESEM micrographs of the NH-0 paste.

417

418 Table 6. Molar content (in %) of the elements at points related to gel formation from NH-0 paste.

Spot	Element (molar, %)						TOTAL (%)
	Si	Al	Na	Ca	K	Mg	
Gel – 1	30.17	10.58	21.87	35.04	0.00	2.34	100
Gel – 2	29.60	11.85	21.12	35.11	0.00	2.33	100
Gel – 3	30.00	11.36	20.85	35.87	0.00	1.92	100

419

420

421 3.3. Comparison of SCSA to other silica sources

422

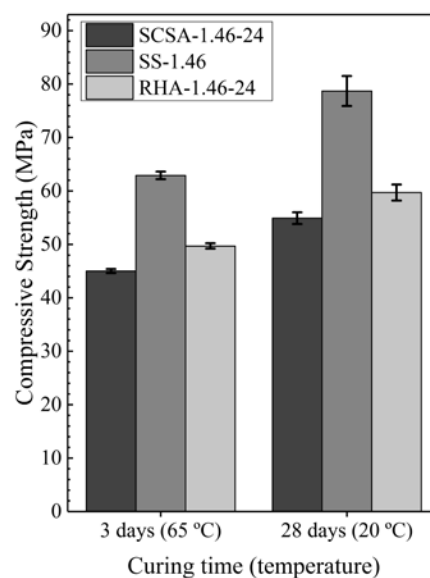
423 The compressive strength of SCSA-1.46-24 was compared to SS-1.46 and RHA-1.46-24 after three days
 424 of curing at 65 °C, and also after 28 days of curing at 20 °C (Fig. 10). SS-1.46 possessed the highest
 425 compressive strength, with 62.9 MPa and 78.8 MPa observed for the 3-day (at 65 °C) and 28-day (at 20
 426 °C) curing times, respectively. The two ash systems, SCSA and RHA, yielded similar results. SCSA
 427 possessed compressive strengths of 45 MPa and 54.9 MPa for 3 days and 28 days, respectively, whereas
 428 RHA reached 49.7 MPa and 59.7 MPa for the same curing ages. Curing at 20 °C yielded higher

429 compressive strengths in all cases. Although ε was equal for the three mixtures, the presence of non-
430 dissolved silica from the ashes (SCSA and RHA) likely caused the lower compressive strengths. It was
431 difficult to determine the amount of solubilised silica after the treatment in the thermal bottle, because the
432 gelification of the suspension impeded filtration of the solution. However, it was obvious that the quartz
433 present in SCSA was not dissolved, which thus generated less solubilised silica. This is the likely reason
434 for the slightly higher compressive strengths of the RHA samples.

435

436 SCSA has the potential to be utilised as a silica source to produce the activating suspension, even though
437 SCSA possessed only 75–80% of the compressive strength obtained for SS-1.46. Spent diatomaceous
438 earth was analysed in a previous study that assessed the production of alternative solutions in a fly
439 ash/metakaolin system [22]. However, the compressive strength of the pastes obtained from this solution
440 was only ~50% of the strength of the SS control. The spent diatomaceous earth sample also yielded
441 similar results to RHA. Waste glass has also been utilised as a silica source to produce the activating
442 solution [23]. The pastes produced by the activating solution with glass waste from a BFS-based system
443 possessed ~75–80% of the compressive strength of the control, which is similar to the SCSA results
444 presented here. More recently, sugar cane bagasse ash was evaluated as a silica source in the production
445 of the activating solution [25]. This new solution yielded similar results in compressive strength tests to
446 those from waste glass and RHA in metakaolin-based AAM.

447



448

449 Fig. 10. Comparison of the compressive strengths of three mortars (cured at 65 °C for 3 days and 20 °C
450 for 28 days) obtained from SCSA, SS and RHA.

451

452 The XRD patterns of SCSA-1.46-24 and SS-1.46 possessed several differences (Fig. 3). The main one
453 was the obvious absence of quartz in SS-1.46. The SCSA-1.46-24 diffractogram also highlighted the
454 presence of faujasite, which was not observed in the SS-1.46 sample. Low intensity peaks that
455 corresponded to hydrotalcite and calcite were also identified in SS-1.46. These XRD observations and the
456 higher soluble silica content explain why SS-1.46 possessed the highest compressive strength.

457

458 The FTIR studies (Fig. 4) did not differentiate between SCSA-1.46-24 and SS-1.46, as only a small
459 difference in the energy of the main Si-O band exists. This result suggests that the chemical nature of the
460 formed hydrates is similar.

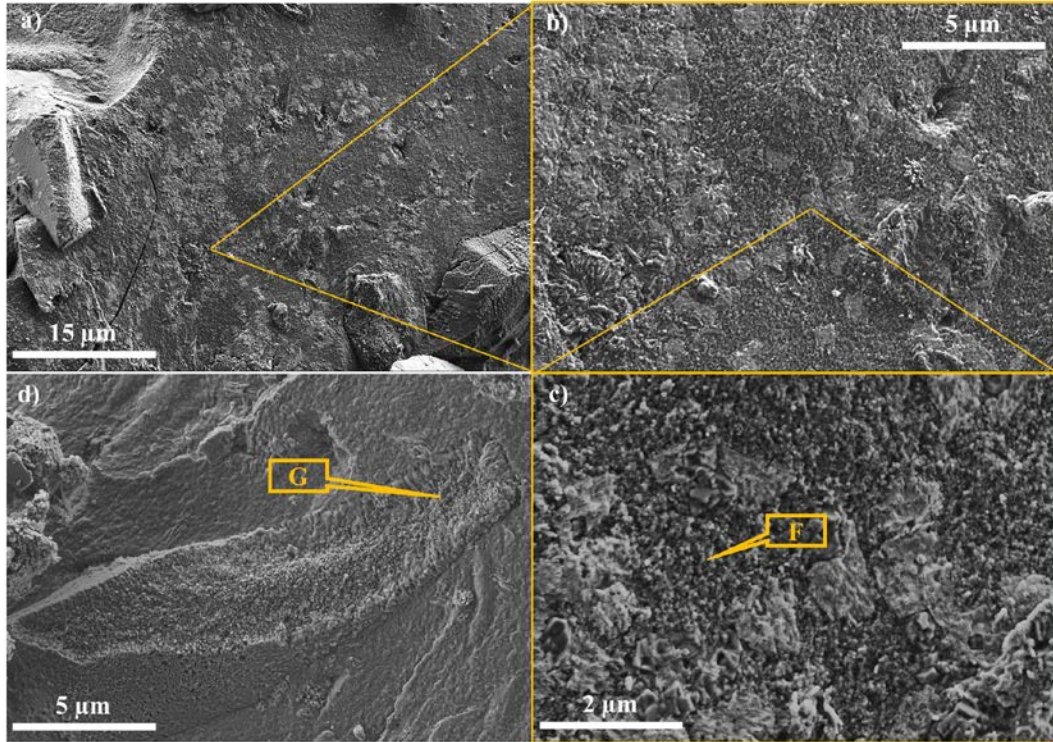
461

462 The mass loss of SS-1.46 was higher than that of SCSA-1.46-24 (Fig. 5 and Table 3). This is likely due to
463 the increased formation of cementing gels in SS-1.46, which also justifies the higher compressive
464 strength. The shapes of the DTG curves of both SCSA pastes are also more similar to the specimen
465 produced with SS than NaOH, with a large peak related to the mass loss due to the dehydration of (C,N)-
466 A-S-H products and no peaks related to decomposition of C-A-S-H compounds. This behaviour suggests
467 that the SCSA and SS pastes produce similar products.

468

469 The FESEM micrographs of the SS-1.46 paste after three days of curing at 65 °C are depicted in Fig. 11.
470 This paste possesses an amorphous and dense structure, highlighted in the magnification at Spot F (Fig.
471 11c). A comparison of Fig. 11c to Fig. 6c (SCSA-1.46-24) reveals that the compactness of the former was
472 slightly lower than that found for the SCSA paste. The formed gel in the SS paste was also similar to the
473 ones observed in the SCSA pastes. The molar ratios of the gel on SS-1.46 were: $\text{Ca/Si} = 0.90 \pm 0.04$,
474 $\text{Al/Si} = 0.23 \pm 0.02$, $\text{Na/Al} = 2.40 \pm 0.16$ and $\text{Mg/Si} = 0.09 \pm 0.03$. Table 7 brings the elements molar
475 content to calculate these ratios from Spot F. The Ca/Si ratio was slightly lower than those for the SCSA
476 pastes, probably due to the chemical combination of more silica from the SS solution. A BFS particle
477 covered with reaction products was observed at Spot G on Fig. 11d. The chemical composition for this
478 spot was: $\text{Ca/Si} = 1.09 \pm 0.13$, $\text{Al/Si} = 0.45 \pm 0.11$, $\text{Na/Al} = 1.19 \pm 0.26$ and $\text{Mg/Si} = 0.64 \pm 0.26$. Table 8

479 shows the molar content of elements from EDS results for Spot G. A comparison of Spot G to Spot F
 480 showed that the former presented higher ratios of Ca/Si, Al/Si and Mg/Si than the latter. The more
 481 relative presence of Ca, Al and Mg with respect to Si suggests that the BFS particle was partially reacted.
 482



483

484

485

486

Fig. 11. FESEM micrographs of the SS-1.46 paste.

Table 7. Molar content (in %) of the elements at points related to spots F from SS-1.46 paste.

Spot	Element (molar, %)						TOTAL (%)
	Si	Al	Na	Ca	K	Mg	
Spot F – 1	35.14	8.12	18.93	33.03	0.85	3.93	100
Spot F – 2	34.92	8.72	19.37	32.82	0.00	4.17	100
Spot F – 3	35.84	8.26	20.38	31.27	0.73	3.51	100
Spot F – 4	36.48	8.49	18.93	32.01	0.64	3.45	100
Spot F – 5	36.85	7.69	19.75	32.82	1.01	1.88	100
Spot F – 6	37.48	7.72	19.89	32.16	0.84	1.91	100

487

488

Table 8. Molar content (in %) of the elements at points related to spots G from SS-1.46 paste.

Spot	Element (molar, %)						TOTAL (%)
	Si	Al	Na	Ca	K	Mg	
Spot G – 1	32.95	13.64	2.87	37.20	0.50	12.84	100
Spot G – 2	30.04	14.04	5.75	34.81	0.00	15.37	100
Spot G – 3	23.97	11.61	13.97	30.93	0.00	19.52	100
Spot G – 4	26.48	14.76	10.77	25.84	0.00	22.15	100
Spot G – 5	33.96	9.06	17.92	31.67	0.72	6.67	100
Spot G – 6	29.87	14.47	6.05	31.41	0.57	17.63	100
Spot G – 7	26.84	14.98	9.59	26.38	0.00	22.22	100
Spot G – 8	23.43	13.85	10.57	29.61	0.00	22.55	100
Spot G – 9	26.81	11.97	12.34	30.84	0.00	18.04	100
Spot G – 10	32.16	9.88	17.57	30.62	0.66	9.11	100

490

491 **4. CONCLUSIONS**

492

493 SCSA was successfully utilised as a silica source to produce the alkaline activating reagent for
494 manufacturing BFS-based AAM materials by means of partial dissolution in a thermal bottle. The release
495 of heat due to the dissolution of NaOH in water facilitates the dissolution of silica from SCSA.
496 Furthermore, the obtained suspensions yielded good behaviour as an activating reagent with no filtering
497 required, thus facilitating their implementation in AAMs. Compressive strength tests of the mortars
498 showed that the optimum time for dissolving the NaOH/SCSA suspension in the thermal bottle (τ) was 24
499 h. The optimum amount of SCSA (ε) was 1.46. Although the SCSA samples possessed lower mechanical
500 properties compared to the specimens produced with the commonly used NH/SS solution, the ash-based
501 systems showed significantly better results than the samples prepared with only NaOH. Furthermore, the
502 alkaline treatment of SCSA in the thermal bottle with NaOH produced a very reactive alkaline activator
503 for BFS-based AAMs. It can be concluded from the microstructural studies that the SCSA pastes
504 presented characteristics that were similar to the matrices obtained with an SS-based reagent. Dense and
505 amorphous gels with comparable Ca/Si, Al/Si and Na/Al molar ratios were also identified.

506

507 **ACKNOWLEDGMENTS**

508 The authors would like to thank CNPq processo n° 401724/2013-1 and CNPq processo n° 140779/2015-0.

509 The authors would also like to thank the Electron Microscopy Service of the Universitat Politècnica de
510 València.

511

512 **REFERENCES**

513 [1] F. Pacheco-Torgal, J.A. Labrincha, C. Leonelli, A. Palomo, P. Chindapasirt, Handbook of Alkali-
514 activated Cements, Mortars and Concretes, first ed., Woodhead Publishing, Cambridge, 2015.

515 [2] J.L. Provis, J.S.J. van Deventer, Alkali Activated Materials: State-of-the-Art Report, RILEM TC 224-
516 AAM, Springer, Dordrecht, 2014.

517 [3] I. Kourti, D.A. Rani, D. Deegan, A.R. Boccaccini, C.R. Cheeseman, Production of geopolymers using
518 glass produced from DC plasma treatment of air pollution control (APC) residues, *J. Hazard. Mater.* 176
519 (2010) 704–709.

520 [4] S.A. Bernal, E.D. Rodríguez, A.P. Kirchheim, J.L. Provis, Management and valorisation of wastes
521 through use in producing alkali-activated cement materials, *J. Chem. Technol. Biotechnol.* 91 (2016)
522 2635–2388.

523 [5] N. Toniolo, A.R. Boccaccini, Fly ash-based geopolymers containing added silicate waste. A review,
524 *Ceram. Int.* 43 (2017) 14545–14551.

525 [6] Food and Agriculture Organization of the United Nations (FAOSTAT).
526 <http://www.fao.org/faostat/en/#home> (accessed 21 September 2017).

527 [7] M. Frías, E. Villar-Cociña, E. Valencia-Morales, Characterisation of sugar cane straw waste as
528 pozzolanic material for construction: Calcining temperature and kinetic parameters, *Waste Manage.* 27
529 (2007) 533–538.

530 [8] F. Martirena, J. Monzó, Vegetable ashes as Supplementary Cementitious Materials, *Cem. Concr. Res.*
531 (2017) In Press.

532 [9] J.C.B. Moraes, J.L.P. Melges, J.L. Akasaki, M.M. Tashima, L. Soriano, J. Monzó, M.V. Borrachero,
533 J. Payá, Pozzolanic Reactivity Studies on a Biomass-Derived Waste from Sugar Cane Production: Sugar
534 Cane Straw Ash (SCSA), *ACS Sust. Chem. Eng.* 4 (2016), 4273-4279.

535 [10] J.C.B. Moraes, J.L. Akasaki, J.L.P. Melges, J. Monzó, M.V. Borrachero, , L. Soriano, J. Payá, M.M.
536 Tashima, Assessment of sugar cane straw ash (SCSA) as pozzolanic material in blended Portland cement:

537 Microstructural characterization of pastes and mechanical strength of mortars, *Constr. Build. Mater.* 94
538 (2015) 670-677.

539 [11] G.A. Calligaris, M.K.K.D Franco, L.P. Aldrige, M.S. Rodrigues, A.L. Beraldo, F. Yokaichiya, X.
540 Turrillas, L.P. Cardoso, Assessing the pozzolanic activity of cements with added sugar cane straw ash by
541 synchrotron X-ray diffraction and Rietveld analysis, *Constr. Build. Mater.* 98 (2015) 44-50.

542 [12] A. Guzmán, C. Gutiérrez, V. Amigó, R.M. Gutiérrez, S. Delvasto, Valoración puzolánica de la hoja
543 de la caña de azúcar, *Mater. Construcc.* 61 (2011) 213-225.

544 [13] J.C.B. Moraes, M.M. Tashima, J.L. Akasaki, J.L.P. Melges, J. Monzó, M.V. Borrachero, L. Soriano,
545 J. Payá, Increasing the sustainability of alkali-activated binders: The use of sugar cane straw ash (SCSA),
546 *Constr. Build. Mater.* 124 (2016) 148–154.

547 [14] A. Nazari, J. Sanjayan. *Handbook of Low Carbon Concrete*, Butterworth-Heinemann, Woburn,
548 2016.

549 [15] B.C. McLellan, R.P. Williams, J. Lay, A. van Riessen, G.D. Corder, Costs and carbon emissions for
550 Geopolymer pastes in comparison to Ordinary Portland Cement, *J. Clean. Prod.* 19 (2011) 1080-1090.

551 [16] G. Habert, J.B. d'Espinose de Lacaillerie, N. Roussel, An environmental evaluation of geopolymer
552 based concrete production: reviewing current research trends, *J. Clean. Prod.* 19 (2011) 1229-1238.

553 [17] L.K. Turner, F.G. Collins, Carbon dioxide equivalent (CO₂-e) emissions: A comparison between
554 geopolymer and OPC cement concrete, *Constr. Build. Mater.* 43 (2013) 125-130.

555 [18] J.L. Provis, A. Palomo, C. Shi, Advances in understanding alkali-activated materials, *Cem. Concr.*
556 *Res.* 78 (2015) 110–115.

557 [19] J.L. Provis, Alkali-activated materials, *Cem. Concr. Res.* (2017) In press.

558 [20] N. Bouzón, J. Paya, M.V. Borrachero, L. Soriano, M.M Tashima, J. Monzó, Refluxed rice husk
559 ash/NaOH suspension for preparing alkali activated binders, *Mater. Lett.* 115 (2014) 72–74.

560 [21] J.M. Mejía, R.M. Gutiérrez, F. Puertas, Rice husk ash as a source of silica in alkali-activated fly ash
561 and granulated blast furnace slag systems, *Mater. Construcc.* 63 (2013) 361–375.

562 [22] J.M. Mejía, R.M. Gutiérrez, C. Montes, Rice husk ash and spent diatomaceous earth as a source of
563 silica to fabricate a geopolymeric binary binder, *J. Clean. Prod.* 118 (2016) 133–139.

564 [23] F. Puertas, M. Torres-Carrasco, Use of glass waste as an activator in the preparation of alkali-
565 activated slag. Mechanical strength and paste characterisation, *Cem. Concr. Res.* 57 (2014) 95–104.

566 [24] M. Torres-Carrasco, F. Puertas, Waste glass in the geopolymer preparation. Mechanical and
567 microstructural characterisation, *J. Clean. Prod.* 90 (2015) 397–408.

568 [25] H. K. Tchakouté, C. H. Rüschler, M. Hinsch, J. N. Y. Djobo, E. Kamseu, C. Leonelli, Utilization of
569 sodium waterglass from sugar cane bagasse ash as a new alternative hardener for producing metakaolin-
570 based geopolymer cement, *Chemie der Erde* 77 (2017) 257–266.

571 [26] UNE-EN 196-1 2005, 2005. Methods of Testing Cement – Part 1, Determination of Strength.

572 [27] C. Shi, P. V. Krivenko, D. Roy, *Alkali-Activated Cements and Concretes*, Taylor & Francis,
573 Abingdon, 2006.

574 [28] I. Ismail, S.A. Bernal, J.L. Provis, R.S. Nicolas, S. Hamdan, J.S.J. van Deventer, Modification of
575 phase evolution in alkali-activated blast furnace slag by the incorporation of fly ash, *Cem. Concr.*
576 *Compos.* 45 (2014) 125–135.

577 [29] A. Fernández-Jiménez, F. Puertas, Structure of Calcium Silicate Hydrates Formed in Alkaline-
578 Activated Slag: Influence of the Type of Alkaline Activator, *J. Am. Ceram. Soc.* 86 (2003) 1389–1394.

579 [30] S.A. Bernal, R.M. Gutierrez, J.L. Provis, V. Rose, Effect of silicate modulus and metakaolin
580 incorporation on the carbonation of alkali silicate-activated slags, *Cem. Concr. Res.* 40 (2010) 898–907.

581 [31] A.F. Abdalqader, F. Jin, A. Al-Tabbaa, Development of greener alkali-activated cement: utilisation
582 of sodium carbonate for activating slag and fly ash mixtures, *J. Clean. Prod.* 113 (2016) 66–75.

583 [32] X. Gao, Q.L. Yu, H.J.H. Brouwers, Reaction kinetics, gel character and strength of ambient
584 temperature cured alkali activated slag–fly ash blends, *Constr. Build. Mater.* 80 (2015) 105–115.

585 [33] J.I. Escalante-García, A.F. Fuentes, A. Gorokhovskiy, P.E. Fraire-Luna, G. Mendonza-Suarez,
586 Hydration Products and Reactivity of Blast-Furnace Slag Activated by Various Alkalis, *J. Am. Ceram.*
587 *Soc.* 86 (2003) 2148–2153.

588 [34] A. Buchwald, H. Hilbig, C. Kaps, Alkali-activated metakaolin-slag blends—performance and
589 structure in dependence of their composition, *J. Mater. Sci.* 42 (2007) 3024–3032.