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# Valorization of dredged sediments in self-consolidating concrete: Fresh, hardened, and microstructural properties

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1 Word count: 7413

2 **Valorization of Dredged Sediments in Self-Consolidating Concrete: Fresh,**  
3 **Hardened, and Microstructural Properties**

4 **Abstract**

5 Several studies have proven the use of dredged sediments as supplementary cementitious  
6 materials (SCMs), but limited information is available on the effect of such treated sediments on  
7 self-consolidating concrete performance. The main objective of this study was to evaluate the  
8 performance of self-consolidating concrete (SCC) fabricated with treated sediments. The sediments  
9 were thermally treated at 800 °C for 1 h. The packing density of the granular skeleton was  
10 optimized to reduce the paste content and produce SCC with relatively low binder content. Three  
11 different SCC mixtures were prepared with 0%, 10%, and 20% cement replaced with treated  
12 sediments by mass. Key fresh, physical, hardened, and microstructural properties of the investigated  
13 SCC mixtures subject to different curing regimes were evaluated. The test results showed that the  
14 optimized SCC mixtures exhibited adequate self-consolidation characteristics. The particle size and  
15 high chemical activity of the sediments led to pore refinement of micro-pores, increased density,  
16 improved microstructure, and reduced micro-cracks of the investigated SCC mixtures. Furthermore,  
17 the use of up to 20% of treated sediments resulted in a compressive strength of  $66 \pm 1$  MPa at 91  
18 days, which is comparable to that of the reference mixture made without any sediments. Leaching  
19 test results confirmed the ecological potential of producing SCCs based on sediments, which could  
20 be an interesting alternative of using local materials to reduce the high demand of cement, thus  
21 further reducing the CO<sub>2</sub> footprint of concrete structures.

22 **Keywords:** Dredged sediments; Mechanical properties; Packing density; Self-consolidating  
23 concrete; Supplementary cementitious material; Ultrasonic pulse velocity.

## 24 **1. Introduction**

25 Self-consolidating concrete (SCC) can be used to facilitate concrete mixing and placement  
26 and ensure proper filling of complex frameworks without any mechanical consolidation. This can  
27 help reduce cost, labor, and construction time and provide more freedom in the design and  
28 enhancement of surface finish (Khayat et al., 1999). The design of stable SCCs can enhance the  
29 interfacial transitional zone (ITZ) properties with embedded reinforcement and with aggregates that  
30 can lead to greater SCC performance (Khayat et al., 1999). One of the drawbacks of SCC is its  
31 higher content of fine powder attributed to the relatively high-binder content than that in  
32 conventional concrete. Extensive research has been conducted on the feasibility of valorizing  
33 abundantly available by-products as supplementary cementitious materials (SCMs) to partially  
34 replace cement and reduce the environmental footprint of SCC. Commonly used SCMs are fly ash,  
35 ground-granulated blast-furnace slag, silica fume, limestone filler, marble powder, calcined clay,  
36 palm oil fuel ash (Mohammadhosseini et al., 2015), and waste ceramic nanoparticles (Lim et al.,  
37 2018). These materials are primarily composed of silicon dioxide ( $\text{SiO}_2$ ), aluminum oxide ( $\text{Al}_2\text{O}_3$ ),  
38 and calcium oxide ( $\text{CaO}$ ). The presence of additional oxides can be beneficial to produce additional  
39 pozzolanic calcium-silicate-hydrate (C-S-H) gel.

40 Significant volumes (more than one billion cubic meter) of dredged sediments are produced  
41 worldwide every year (Benzerzour et al., 2017). These materials are stored or disposed at sea when  
42 they are not contaminated. However, these sediments are very heterogenous: the provenance and  
43 dredging time during the year have a crucial influence on their composition. This makes each  
44 sediment a unique material which requires special treatment. The valorizing of dredged sediments  
45 in concrete, engineered materials, and civil engineering applications presents considerable benefits.  
46 Such valorization includes the fabrication of bricks (Samara et al., 2009), ceramic products (Xu et  
47 al., 2014), lightweight aggregates (Liu et al., 2018), and road construction materials (Kasmi et al.,  
48 2017). Recent studies have demonstrated that thermally treated sediments can render the sediments

49 more reactive and that treated sediments can be useful for clinker production (Aouad et al., 2012;  
50 Faure et al., 2017) or as SCM.

51 Dang et al. (2013) designed a new blended cement containing 8%, 16%, and 33% of treated  
52 sediments from the trap Lyvet on the Rance River in France. The sediments were heated at 650 °C  
53 and at 850 °C for 5 h. Blended cements containing 8% treated sediments secured equivalent long-  
54 term compressive strength compared with the reference mixture made without any sediment. The  
55 authors reported that the composite binder required a longer curing period for the development of  
56 mechanical properties. Ez-zaki and Diouri (2019) investigated the microstructural and physic-  
57 mechanical properties of mortars made with 8% and 33% thermally treated sediments and shell  
58 powder at 650 and 850 °C for 5 h. Despite the pozzolanic reactivity of treated sediments, the  
59 compressive strength of the mixtures containing the treated sediment was lower than that of the  
60 control mortar. Van Bunderen et al. (2019, 2017) studied the early-age hydration and autogenous  
61 deformation of cement paste containing 20%, 30%, and 40% of flash calcined dredging sediments  
62 treated at 865 °C. The treated sediments and fly ash developed a similar early-age hydration  
63 behavior, and the addition of sediments reduced the autogenous shrinkage of mortar. Snellings et al.  
64 (2017, 2016) studied the pozzolanic reactivity of flash calcined sediments (from the port of  
65 Antwerp, Belgium) treated at temperatures of 820, 865, and 905 °C. The pozzolanic reactivity of  
66 sediments was found to be inferior to that of metakaolin but was superior of that of siliceous fly ash  
67 in a calorimetry based pozzolanic reactivity test. Zhao et al. (2018) investigated the effect of using  
68 10%, 20%, and 30% cement replacement of uncontaminated sediments on properties of mortar and  
69 concrete mixtures. The reported results showed that dried and finely ground sediments can be used  
70 as partial substitutes of cement by up to 20% without hindering the mechanical properties of the  
71 mixtures. Benzerzour et al. (2018, 2017) developed a new blended cement containing 8% and 15%  
72 sediments thermally treated at temperatures of 650, 750, and 850 °C for different durations ranging  
73 between 1 and 3 h. Test results showed that mortar mixtures incorporating 15% sediments treated at  
74 850 °C for 1 h developed better mechanical strength than the control mortar.

75           Although various non-traditional by-products have been recycled to SCC, such as waste  
76 carpet fibers and palm oil fuel ash (Mohammadhosseini et al., 2018, 2017), limited research has  
77 been conducted on the engineering properties of SCC incorporating the treated sediments.  
78 Sediments used as fillers in SCC achieved good performance (Rozas et al., 2015; Rozière et al.,  
79 2015). Bouhamou et al. (2016) evaluated the influence of using 10%, 15%, and 20% replacements  
80 of cement by calcined dam mud sediments treated at 750 °C for 5 h on the shrinkage of SCC. They  
81 reported that the increase in calcined mud content resulted in more viscous and less workable SCC  
82 for a given water content. A higher mechanical strength of SCC containing 10% calcined mud was  
83 reported. However, notably, the control SCC mixture exhibited the highest strength development. It  
84 was also reported that the addition of calcined mud resulted in lower autogenous shrinkage. This  
85 was attributed to the formation of expansive calcium aluminate hydrates, which compensated for  
86 the shrinkage. Nevertheless, the effect of sediments on fresh, hardened, and microstructural  
87 properties of SCC are still not well understood.

## 88 **2. Research significance**

89           The present study is part of a wide investigation aimed at evaluating the feasibility of using  
90 dredged marine sediments from Dunkirk Port as SCM in SCC mixtures. Valorizing the sediments  
91 using high-fluid cement paste mixtures at different substitution rates did not significantly affect the  
92 fresh properties of paste when the water demand of sediments was considered (Safhi et al., 2018).  
93 The study showed that the treated sediments can be used for partial replacement of cement by up  
94 30%. Another study conducted on equivalent-SCC mortar mixtures confirmed the previously  
95 reported results (Safhi et al., 2019). However, although the treated sediments are considered as  
96 pozzolanic materials, the use of higher substitution rate reduced the strength and modulus of  
97 elasticity of SCC mortars. Optimum replacement rates of 7% and 35% in the case of binder content  
98 ranging between 380 and 600 kg/m<sup>3</sup> and w/cm of 0.32 and 0.34, respectively, were reported.

99           The main objective of this study is to obtain deeper insights into the effect of using treated  
100 sediment on the fresh, physical, hardened, and microstructural properties of SCC proportioned with

101 a fixed binder content of 450 kg/m<sup>3</sup>. A leaching test was conducted to evaluate the potential of  
102 producing SCC containing recycled sediments.

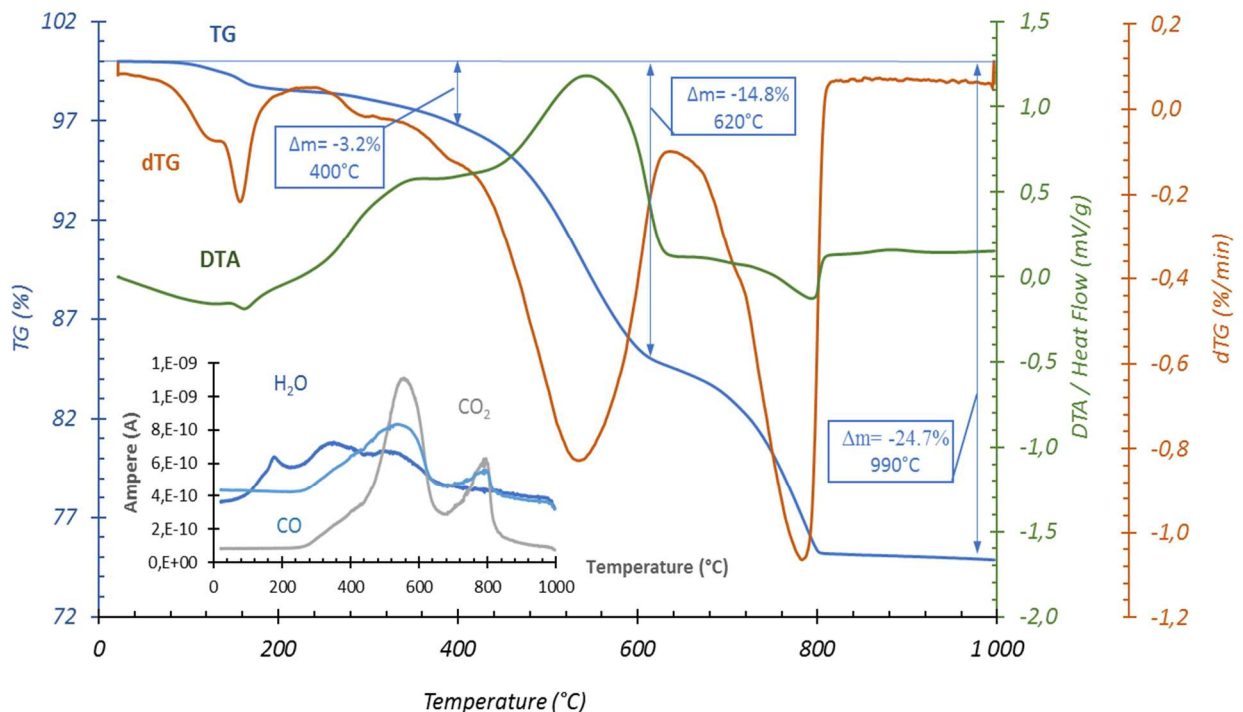
### 103 **3. Materials and experimental program**

#### 104 **3.1. Material characteristics**

105 The investigated SCC mixtures were fabricated using a ternary binder containing  
106 approximately 70% General Use (GU) cement, 25% class F fly ash, and 5% silica fume. Two  
107 crushed coarse aggregates (CA), including CA<sub>1</sub> and CA<sub>2</sub> with nominal maximum sizes of 20 and 10  
108 mm, respectively, were used. The CA<sub>1</sub> and CA<sub>2</sub> aggregates, from St Dominique career, had a  
109 specific density of 2.76 and 2.72, water absorption values of 0.48% and 0.41%, and bulk densities  
110 of 1465 and 1444 kg/m<sup>3</sup>, respectively. A siliceous river sand with a specific density of 2.66, a water  
111 absorption of 1.09%, and a bulk density of 1785 kg/m<sup>3</sup> was used.

112 The sediments used in this study were extracted from the Port of Dunkirk in France. They  
113 were dried in an oven at 60 °C and subsequently sieved to recuperate fine particles smaller than 2  
114 mm. Thermogravimetric analysis (TGA) was performed using NETZSCH STA 449 and nitrogen  
115 gas in a controlled environment with argon flow (75 ml/min) at variable temperatures ranging  
116 between 105 and 1100 °C at an increasing rate of 2 °C/min. Differential thermal analysis was also  
117 performed using NETZSCH DSC 404F3 at ambient air and variable temperatures (0 to 1000 °C, at  
118 a range of 5 K/min/1000). As can be observed in Fig. 1, the first weight loss around 300 °C was due  
119 to the evaporation of water and dehydration of gypsum (Siavalas et al., 2013). The weight loss of  
120 3.2% observed between 400 and 500 °C was due to the combustion of carbon and organic matter, as  
121 shown by the increase in the released carbon. This loss was associated with an exothermal peak. A  
122 higher weight loss between 550 and 750 °C was observed, which was associated with an  
123 endothermic peak due to the decomposition of calcite (CaCO<sub>3</sub>) (Madrid et al., 2017). Above 800  
124 °C, the weight loss and calorimetry flux tended to stabilize, which confirmed that almost all organic  
125 matter was eliminated. Calcination at 800 °C for 1 h was chosen as the thermal treatment.

126 After calcination, the powder was ground and then sieved to meet a  $d_{50}$  of less than 45  $\mu\text{m}$ , and the  
 127 obtained product was referred to as treated marine sediment (TMS). The visual aspect of those  
 128 sediments at the various stages of treatment is shown in Fig. 2. The physical and chemical  
 129 characteristics of the ternary binder, raw sediments, and TMS are summarized in Table 1.  
 130 Calcination had a beneficial effect in that it increased the density of the sediments through the  
 131 elimination of organic matter. The sediments were also chemically activated with an increase in  
 132 silica (+37%), despite the decrease in the concentration of aluminum, calcium, ferric, and  
 133 magnesium oxides. Silicon dioxide can react with calcium hydroxide (CH) to form additional  
 134 pozzolanic (C-S-H) gel. The content of  $\text{Fe}_2\text{O}_3$  and  $\text{K}_2\text{O}$  indicates the presence of illite. The changes  
 135 in the colors of sediments after calcination were due to the decrease of  $\text{Fe}_2\text{O}_3$  content from 9.7% to  
 136 5.3%. Notably, a decrease in sulfur trioxide ( $\text{SO}_3$ ) content was observed, which decreased the risk  
 137 of delayed ettringite formation. The particle-size distributions of the CA, sand, cement, and TMS  
 138 are shown in Fig. 3. As can be observed, the particles of TMS have a spherical morphology.



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Fig. 1 : TGA-DSC curves of raw sediments





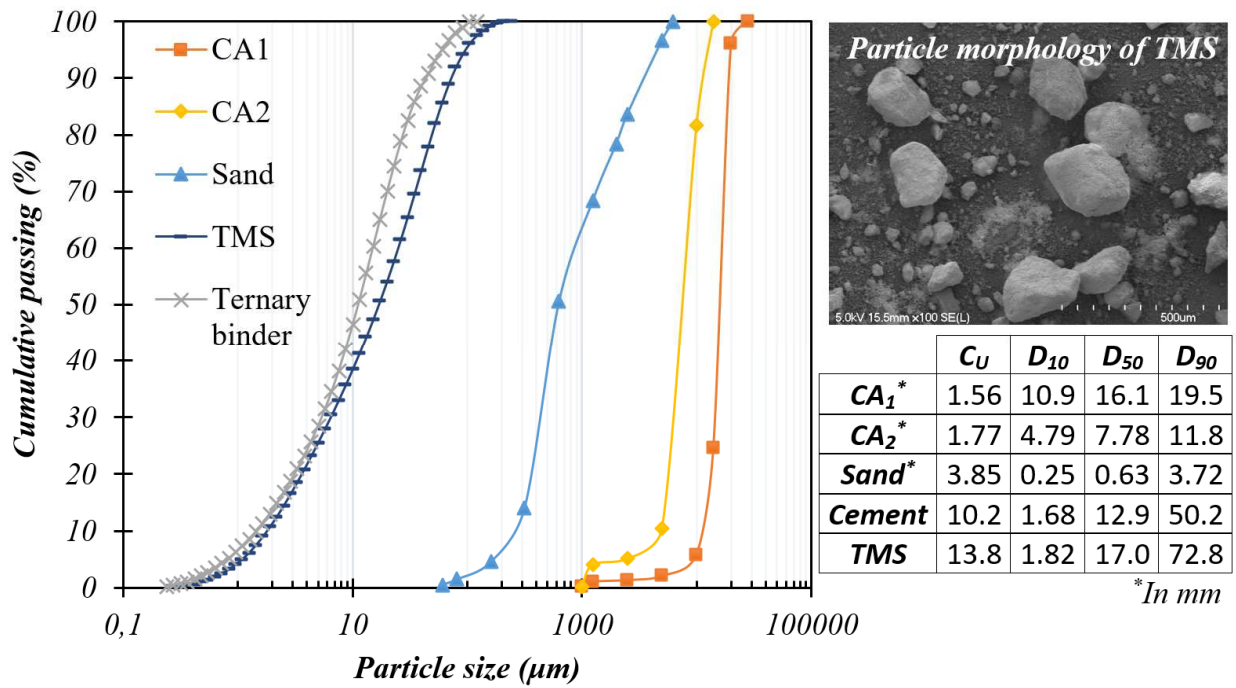
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142 Fig. 2 : Visual aspect from the left to right: raw dried sediment; calcined, ground sediments

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Table 1 : Physio-chemical characteristics of powders

<b>Characteristics</b>		<b>Ternary binder</b>	<b>Raw Sediments</b>	<b>TMS</b>
<b>Physical properties</b>	<i>Density (kg/m<sup>3</sup>)</i>	2888	2467	2704
	<i>BET Surface Area (m<sup>2</sup>/kg)</i>	2535	-	3143
<b>Major oxide (wt %)</b>	<i>Al<sub>2</sub>O<sub>3</sub></i>	8.54	4.12	3.85
	<i>CaO</i>	43.1	11.4	9.73
	<i>Fe<sub>2</sub>O<sub>3</sub></i>	6.46	9.75	5.32
	<i>MgO</i>	1.62	0.96	0.76
	<i>P<sub>2</sub>O<sub>5</sub></i>	0.14	0.14	0.14
	<i>SO<sub>3</sub></i>	3.39	1.49	0.30
	<i>SiO<sub>2</sub></i>	29.4	52.3	71.9
	<i>TiO<sub>2</sub></i>	0.41	0.42	0.27
	<i>K<sub>2</sub>O</i>	1.09	0.95	1.07
	<i>Na<sub>2</sub>O</i>	0.26	0.66	0.73
	<i>Alkali equivalent Na<sub>2</sub>O + 0.658 × K<sub>2</sub>O</i>	0.98	1.29	1.43
	<i>Other undetected oxides</i>	1.68	0.64	0.67
	<i>LOI (1050°C)</i>	3.75	16.9	5.18



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Fig. 3 : Particle-size distributions of powders and aggregates

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### 3.2. Packing density of aggregate

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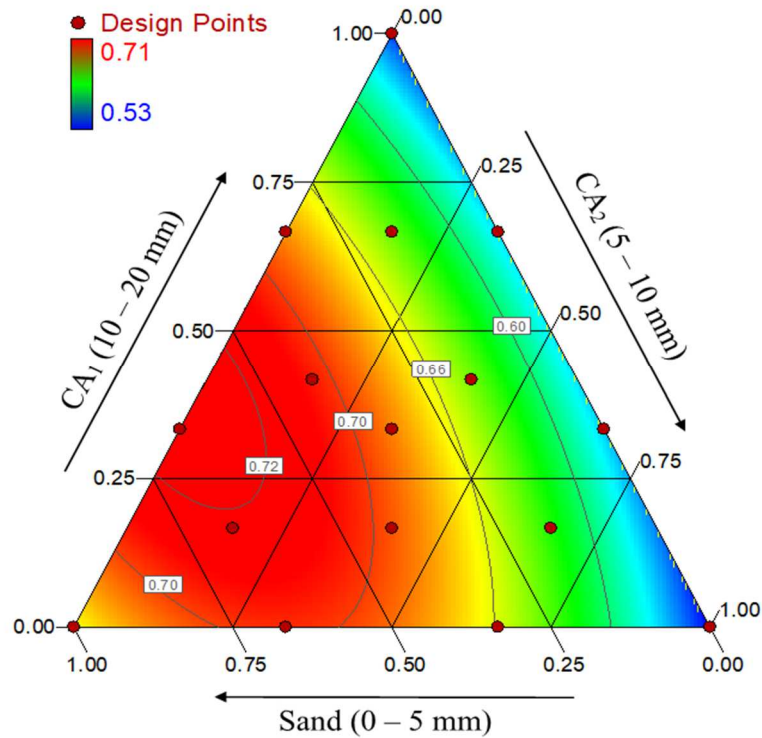
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The packing density (PD) of the aggregate was determined to minimize the paste content and produce SCC with a relatively low binder content. Enhancing the PD can also reduce the high-range water reducing admixture (HRWRA) demand and the viscosity of the concrete (Khayat et al., 2002) and can increase the static stability, compressive strength (Aïssoun et al., 2016), and modulus of elasticity. A high PD can reduce porosity, chloride diffusion, and shrinkage (Abd Elrahman and Hillemeier, 2014). The PD was experimentally determined using a modified version of the test procedure described in ASTM C29 (2017). It involved determining the PD of materials poured from a  $V_{\text{funnel}}$  into a recipient. The used  $V_{\text{funnel}}$  had an upper opening of  $75 \times 515 \text{ mm}^2$  and an outlet opening of  $75 \times 65 \text{ mm}^2$ , as described by the EFNARC (Self-Compacting Concrete European Project Group, 2005). A 7-l cylindrical container with a diameter of 200 mm was used as the recipient. Goltermann et al. (1997) suggested that the diameter of the cylinder should be larger than 10 times the nominal maximum size of the aggregate. Nanthagopalan and Santhanam (2012) used the same test procedure with different test setups.

160 To establish a compact granular skeleton, a mixture design approach based on varying the  
 161 fine and coarse aggregate, CA<sub>1</sub>, CA<sub>2</sub>, and sand at volumetric proportions from 0 to 100% was used.  
 162 Different volumetric proportions of CA<sub>1</sub>, CA<sub>2</sub>, and sand were manually mixed and used to  
 163 determine the PD according to the ENFRAC method. The premixed aggregate combinations were  
 164 placed in the V<sub>funnel</sub> and poured into the container from a height of 0.20 m without any vibration.  
 165 The exceeded quantity was stricken off, and the mass of the filled container was measured. As can  
 166 be observed in Fig. 4, the experimental data, in which each red point represented 16 different  
 167 combinations of aggregates, were used to establish the corresponding contour diagrams (red points  
 168 in Fig. 4). Using the proportions, the reel masses of each component and the voids content were  
 169 determined, and then, the PD values were calculated as follows:

$$170 \quad \text{Packing Density} = 1 - \text{Void content} = 1 - \frac{(V_{\text{container}} - V_{CA1} - V_{CA2} - V_S)}{V_{\text{container}}} \quad \text{Eq. 1}$$

171 where V<sub>container</sub>, V<sub>CA1</sub>, V<sub>CA2</sub>, and V<sub>S</sub> are the volumes of the used container, CA<sub>1</sub>, CA<sub>2</sub>, and sand. The  
 172 quadratic multi-regression model showed the best fit for the experimental measurement (R<sup>2</sup> = 0.980,  
 173 a p-value < 0.001, and a low standard deviation of 0.01). Depending on the selected aggregate  
 174 combination, the PD varied from 0.530 to 0.720. The sand was found to be the most dominant  
 175 parameter. The optimal PD of 0.72 was reached by different combinations of aggregates as shown  
 176 in Fig. 4. Following the recommendations of EFNARC to produce SCCs (48 ≤ V<sub>FA</sub>/V<sub>A</sub> ≤ 55%), a  
 177 higher PD of 0.72 could not be directly assumed; then, a PD of 0.70 was chosen. This PD can be  
 178 achieved using 893 kg/m<sup>3</sup> of sand, 147 kg/m<sup>3</sup> of CA<sub>1</sub>, and 578 kg/m<sup>3</sup> of CA<sub>2</sub>, which correspond to  
 179 55%, 10%, and 40% by volume, respectively.



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*Fig. 4: Ternary PD diagram, the red points represent experimental measurements*

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### 3.3. Mixtures proportioning

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Three SCC mixtures were prepared with 0 (SCC-R), 10% (SCC-1), and 20% (SCC-2) cement replaced with TMS by mass (Table 2). The mixtures were prepared with a constant water-to-cementitious materials ratio (w/cm) of 0.40. The water demand of sediments was calculated according to the method reported by Sedran et al. (2007). A commercially compatible HRWRA was used at an optimum dosage of 1200 ml/100 kg of binder to secure the targeted slump flow ( $S_{flow}$ ) of  $680 \pm 20$  mm.

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All the investigated mixtures were prepared in a rotating drum mixer in batches of 60 l. The mixing sequence was as follows: homogenizing sand for 30 s, followed by adding coarse aggregate and mixing for 60 s. One third of the water was then added, and the material was mixed for 30 s. The cementitious materials were added and mixed for 30 s before introducing HRWRA diluted with one third of the water. The concrete was then mixed for an additional 120 s followed by the addition of the last portion of water. The concrete was mixed for 60 s and left to rest for 2 min, after which the mixing was resumed for another 3 min.

Table 2 : Mixture proportions of the investigated SCC mixtures

<b>Materials</b>	<b>SCC-R</b>	<b>SCC-1</b>	<b>SCC-2</b>
	<b>Content in kg/m<sup>3</sup></b>		
<b>Cement</b>	450	405	360
<b>TMS</b>	-	45.0	90.0
<b>CA<sub>1</sub></b>	147		
<b>CA<sub>2</sub></b>	578		
<b>Sand</b>	893		
<b>Water (w/cm = 0.4)</b>	180	162	144
<b>Added water (w/s =0.35)</b>	-	15.7	31.5

## 197 4. Results and discussion

### 198 4.1. Fresh properties

199 Before casting, fresh property tests (e.g.,  $S_{\text{flow}}$  diameter,  $T_{500}$ ,  $V_{\text{funnel}}$  time,  $J_{\text{Ring}}$  diameter, L-  
200 box, resistance to static segregation, and air content) were performed according to the EFNARC  
201 guidelines. The fresh properties of the investigated SCC mixtures are summarized in Table 3. The  
202 SCC mixtures achieved a  $S_{\text{flow}}$  ranging between 685 and 695 mm, which corresponded to SCC class  
203 2 (SF2) based on EFNARC. A slight decrease in  $S_{\text{flow}}$  was observed when the TMS content  
204 increased, which could be explained by the increase in their specific surface area. Laidani et al.  
205 (2020) found that fine calcined bentonite particles decreased the  $S_{\text{flow}}$  because of the increased  
206 particle surface area of bentonite compared to OPC. All the SCCs exhibited a similar  $T_{500}$  value of 2  
207 s, and the  $V_{\text{funnel}}$  flow time ranged between 6 and 8 s, which agrees with the fresh density (which  
208 increased with increasing TMS content). The  $H_2/H_1$  ratio was determined to evaluate the self-  
209 leveling properties, blocking, and stability of concrete (Khayat et al., 2004). The results of L-box  
210 flow show that the  $H_2/H_1$  ratio ranged between 0.85 and 0.98, and the  $\Delta$  between  $S_{\text{flow}}$  and  $J_{\text{Ring}}$  flow  
211 was < 50 mm. The findings indicate that all mixtures have a good passing ability and were within

212 the targeted range. The segregation index  $\leq 10\%$  and the  $\Delta$  between initial and final  $V_{\text{funnel}}$  flow  $\leq$   
 213  $10\%$  indicates the good stability of the studied SCCs.

214 *Table 3: Results of fresh properties of SCCs*

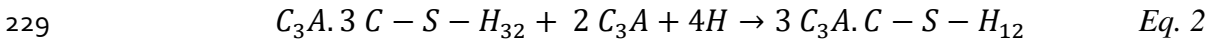
<i>Properties</i>		<i>SCC-R</i>	<i>SCC-1</i>	<i>SCC-2</i>
<i>Filling ability</i>	<i>S<sub>flow</sub> (mm)</i>	695	690	685
	<i>T<sub>500</sub> (s)</i>	2.00	2.03	2.06
	<i>Initial V<sub>funnel</sub> time (s)</i>	8.10	7.41	6.66
<i>Passing ability</i>	<i>J<sub>Ring</sub> flow (mm)</i>	655	662	665
	<i>L-box flow</i>	0.98	0.94	0.85
<i>Stability</i>	<i>Δ<sub>t</sub> initial &amp; final V<sub>funnel</sub> (%)</i>	5.43	3.37	9.31
	<i>Sieve segregation (%)</i>	6.59	8.47	6.23
<i>Air content (%)</i>		3.90	2.20	1.38
<i>Fresh density (kg/m<sup>3</sup>)</i>		2310	2366	2376

215 **4.2. Physical properties**

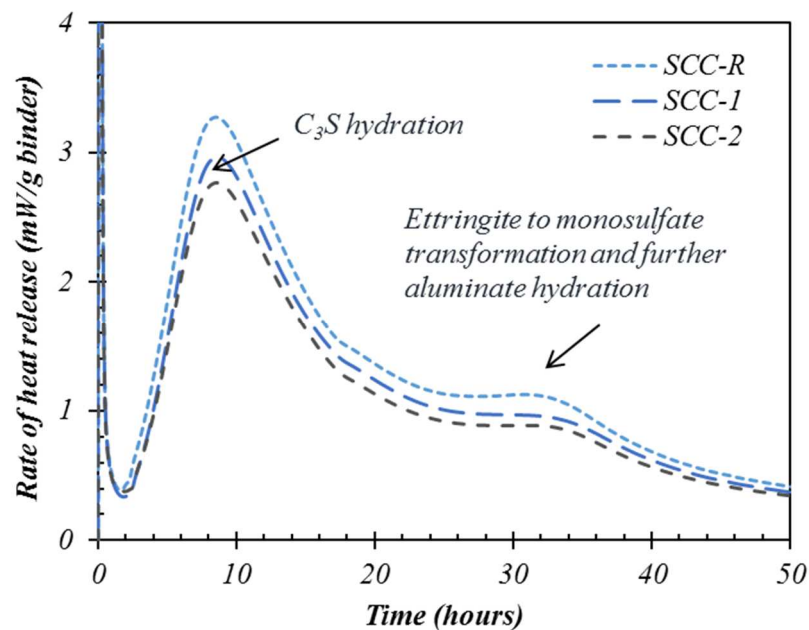
216 **4.2.1. Thermal calorimetry kinetics**

217 Paste mixtures containing 0, 10, and 20% of TMS and a fixed water-to-binder ratio of 0.50  
 218 were prepared and used to evaluate the calorimetric flux using an isothermal calorimeter (TAM Air)  
 219 within 50 h according to ASTM C1679 (2017). The hydration heat evolution curves of cement-  
 220 sediment binders are shown in Fig. 5. Right after mixing the binder with water and because of the  
 221 rapid dissolution of sulfate and aluminate, a sharp exothermic peak was observed which was caused  
 222 by the formation of Aft (alumina, ferric oxide, tri-sulfate) linked to a rapid decrease in heat  
 223 emission. Addition of TMS retarded the hydration of the binder. As can be observed, the SCC-R  
 224 paste generated 3.27 mW/g of heat after 8.53 h, whereas SCC-1 and SCC-2 pastes released 2.96 and  
 225 2.77 mW/g heat after 8.63 and 8.58 h, respectively. The first endothermic peak was mainly related  
 226 to C<sub>3</sub>S and C<sub>3</sub>A hydration which formed C-S-H and ettringite, respectively. The second peak was

227 associated with a certain surplus or delay in  $C_3A$  hydration due to the increase in gypsum content;  
 228 therefore, the ettringite transformed to monosulfate according to the following equation:



230 Unlike the results reported by Benzerzour et al. (2017), the incorporation of TMS reduced the heat  
 231 of hydration. This contradiction can be explained by the fineness of TMS used in this study, which  
 232 was coarser ( $d_{50}$  of TMS particles is 17  $\mu m$ ) than that ( $d_{50}$  of TMS particles is 5  $\mu m$ ) used by  
 233 Benzerzour et al. (2017). This resulted in higher hydration kinetics of sediments. In contrast, the  
 234 retarding effect was caused by the reduction of the  $Ca^{2+}$  ions, which affected the hydration kinetics,  
 235 delayed  $Ca(OH)_2$  nucleation, and generated an unstable C-S-H gel with low Ca/Si ratio, which  
 236 slowly changed into a stable C-S-H gel (Han et al., 2014). A second possible cause could be the  
 237 agglomeration of sediments, which entrapped some mixing water between flocs and reduced the  
 238 available water for hydration (Oey et al., 2013).



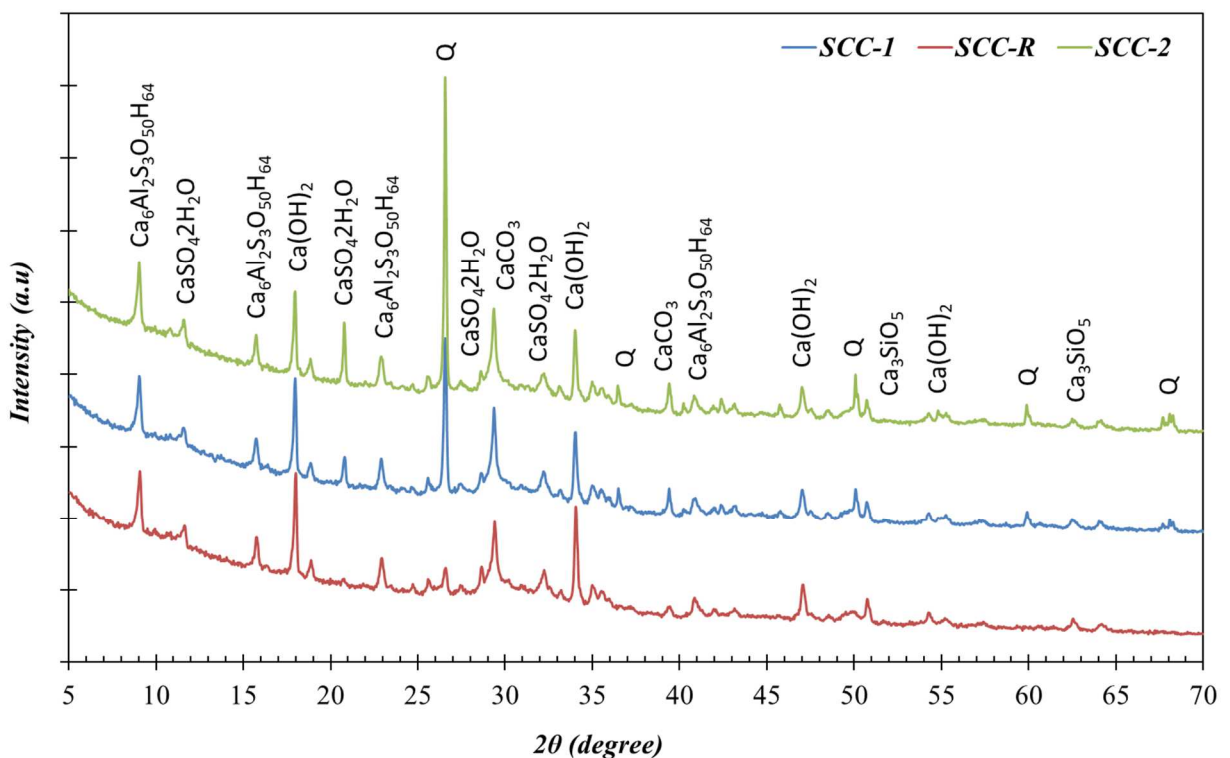
239  
 240 *Fig. 5: Evolution of heat hydration for the different SCC mixtures*

241 **4.2.2. X-ray diffraction analysis**

242 X-ray diffraction analyses were conducted at an age of 40 days on dried and ground pastes.  
 243 Analyses were conducted using a Panalytical X'Pert Pro MPD Diffractometer equipped with  $CuK\alpha$

244 radiation ( $\lambda = 1.54 \text{ \AA}$ ). Fig. 6 shows that quartz (Q), ettringite ( $\text{Ca}_6\text{Al}_2\text{S}_3\text{O}_{50}\text{H}_{64}$ ), and portlandite  
 245 ( $\text{Ca}(\text{OH})_2$ ) were the main phases. Some minor phases, such as gypsum ( $\text{CaSO}_4\cdot 2\text{H}_2\text{O}$ ), alite  
 246 ( $\text{Ca}_3\text{SiO}_5$ ), calcite ( $\text{CaCO}_3$ ), and tricalcium aluminate ( $\text{Ca}_3\text{Al}_2\text{O}_6$ ), were also detected. The XRD  
 247 analysis results agreed with the results of the thermal calorimetry kinetics with respect to the  
 248 formation of ettringite. The early-age phase (9-20°) was a solid solution of carbonated ettringite  
 249 (hem carbonate) and  $\text{OH}^-$  substituted mono-sulfate ettringite (Van Bunderen et al., 2019).

250 The three samples made with different TMSs exhibited the same pattern; overall, there were no  
 251 significant differences in hydration products. The greater the TMS substitution rate, the higher the  
 252 intensity of diffraction peaks of quartz (21, 27, 37, 50, 60, and 68  $2\theta^\circ$ ). This result was consistent  
 253 with the results of XRF analysis: the high  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio indicates the presence of free silica.  
 254 Compared with the reference paste, the other pastes containing 10 and 20% TMS exhibited lower  
 255 intensity diffraction peaks of portlandite, which can be attributed to the pozzolanic activity of  
 256 sediments.



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Fig. 6: XRD profiles of the different SCC pastes



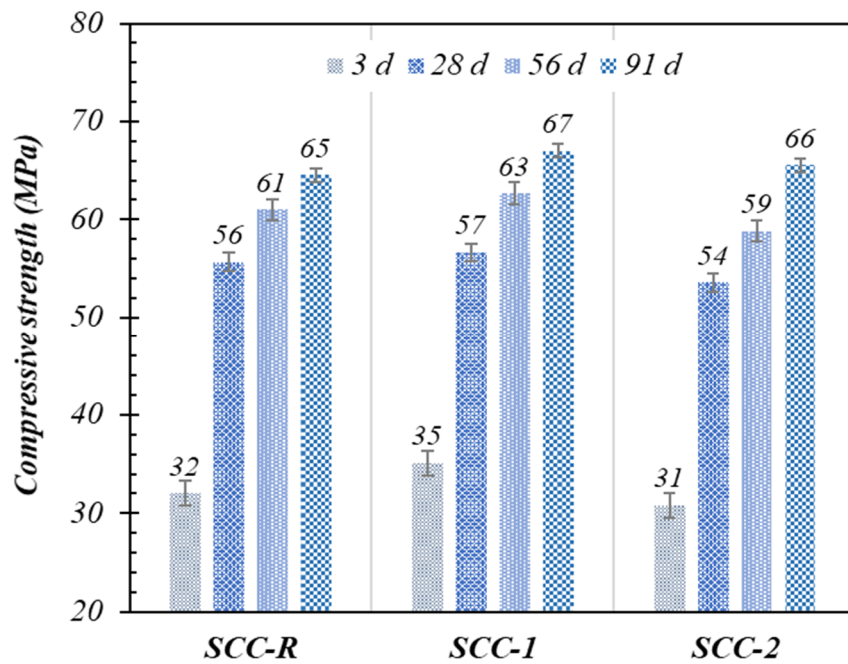
### 4.3. Hardened properties

#### 4.3.1. Compressive and splitting tensile strengths

Compressive strength (ASTM C39, 2018) and splitting tensile strength (ASTM C496, 2017) were determined from 100 Ø 200 mm cylindrical specimens (3 samples for each test), at 3, 28, 56, and 90 days. The results are presented in Fig. 7 and Fig. 8. The investigated SCC mixtures showed a similar tendency of increase in strength with curing time.

The compressive strengths of the SCC-1 mixture made with a replacement rate of 10% increased by approximately 10%, 2%, 3%, and 4% after 3, 28, 56, and 91 days, respectively. However, for a replacement rate of 20%, the early-age compressive strength of the SCC-2 mixture was lower than that of the control mixture but was comparable at later ages. These results are consistent with data obtained in the case of SCMs, which required more curing time to develop their strength. For the splitting tensile strength, the SCC-1 mixture showed a similar behavior to that of SCC-R. However, at 20% replacement, the SCC-2 mixture showed a slightly lower strength at an early age, which was still acceptable considering the standard deviation. According to Çakır (2014), the development of the tensile splitting strength was mainly attributed to the binder rather than the other ingredients. Therefore, the increase in the TMS content resulted in lower amount of hydration products, which can lead to higher porosity, lower bonding strength, and weaker ITZ (Wong et al., 2009).

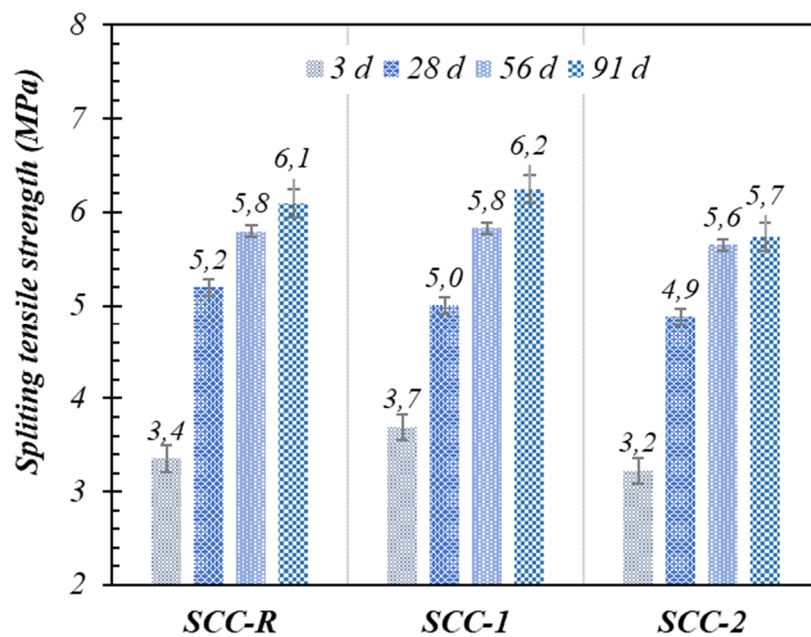
The optimization of the PD and the type of aggregate had an important role on strength development. Nevertheless, replacing more than the above-mentioned contents of TMS would not result in a pozzolanic reaction but would deteriorate the mechanical properties.



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Fig. 7: Compressive strength results of investigated SCC mixtures



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Fig. 8: Splitting tensile strength results of investigated SCC mixtures

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#### 4.3.2. Ultrasonic pulse velocity (UPV) and dynamic modulus of elasticity

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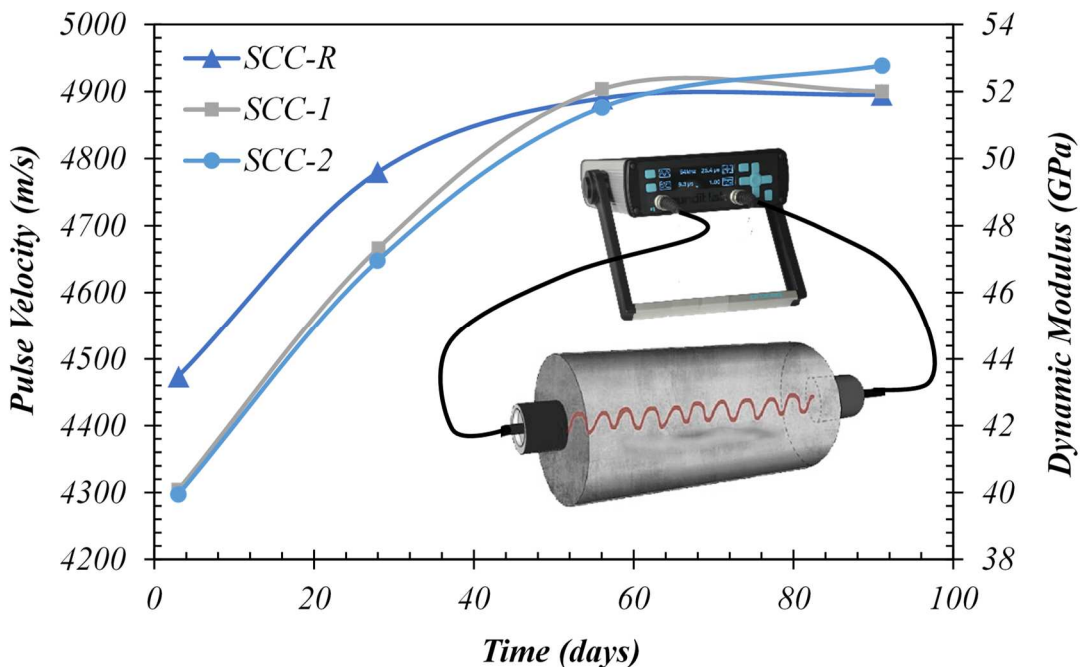
287

The UPV is a non-destructive test that assesses the internal microstructural soundness, micro-cracking, homogeneity, and compactness (Skaropoulou et al., 2013). UPV is associated with micro-crack formation in concrete (Ashrafian et al., 2018). UPV was performed according to

288 ASTM C597 (2016) on the specimens tested for compressive strength. The dynamic modulus of  
 289 elasticity ( $E_{dyn}$ ) was determined theoretically from the UPV values using the following equation:

$$290 \quad E_{dyn} = \frac{\rho (1+\mu)(1-2\mu)}{1-\mu} V^2 \quad Eq. 3$$

291 where  $\rho$  is the density ( $\text{kg/m}^3$ ),  $\mu$  is the dynamic Poisson's ratio, and  $V$  is the pulse velocity (m/s) of  
 292 concrete. The results of UPV and the calculated  $E_{dyn}$  values are shown in Fig. 9. The SCC-R  
 293 mixture had a velocity of 4460 m/s (44 GPa) at 3 days, which increased to 4900 m/s (52 GPa) at 91  
 294 days. The SCC-1 and SCC-2 mixtures showed nearly the same behavior: 4300 m/s (40 GPa) at 3  
 295 days followed by a low increase until the velocity reached that of the SCC-R mixture at 56 days.  
 296 The stabilization after 56 days reflects the total CH consumed by the pozzolanic reaction. The  
 297 evolution of velocity followed the ongoing hydration reactions of binder materials that formed a  
 298 denser and interlocked microstructure. Overall, the UPV results indicated the good quality of  
 299 concrete for the investigated SCC mixtures and agreed with the strength measurements.



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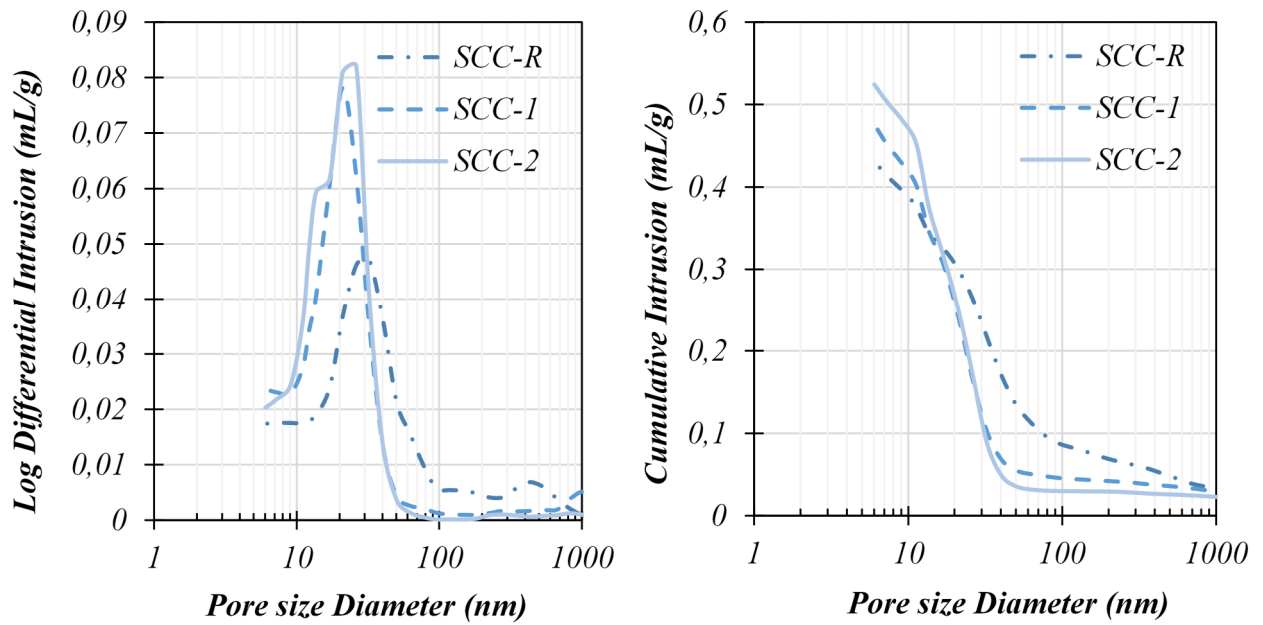
Fig. 9: UPV and  $E_{dyn}$  of the investigated SCC mixtures

## 4.4. Microstructure characterization

### 4.4.1. Mercury porosimetry

Microstructural and pore size distribution of the SCC mixtures were investigated. Fragments taken from SCC mortar portions were dried at 105 °C for 24 h before mercury intrusion porosimetry testing. The differential pore size distributions and the cumulative adsorption were calculated and are illustrated in Fig. 10. The three SCC mixtures showed approximately the same pattern with apparent diameters of most pores in the range of 5 to 50 nm. According to Cheng et al. (2018), the pore structure can be divided into three major ranges: micro-pores (< 20 nm), meso-pores (20–200 nm), and macro-pores (> 200 nm), which is related to impermeability, less permeability, and high permeability, respectively. Increasing the TMS content generated a shift in the differential curve peak, which decreased the micro-pores from 32.3 nm to 21.1 and 26.3 nm for the SCC-1 and SCC-2 mixtures, respectively. This is shown clearly in the cumulative volume: the meso-pores increased, which denotes that the pores became finer and denser.

According to Table 4, the SCC-1 mixture had the highest volume of micro-pores and the lowest volume of meso-pores, followed by SCC-2 and SCC-R mixtures. A negligible volume of macro-pores was observed in the three concrete samples, but it was mainly observed in the SCC-R mixture. Although, the SCC-R mixture had the highest pore size, it exhibited a lower porosity (8.74%) compared with the SCC-1 (9.08%) and SCC-2 samples (9.57%). Nevertheless, the small particle size and high chemical activity of the TMS resulted in finer capillary porosity, which increased the density, improved the structure, and reduced the volume of meso-pores and micro-cracks. The porosity of the investigated mixtures indicates adequate durability because of the optimization of the PD and the pozzolanic reaction of TMS which refined and increased the density of ITZ.



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Fig. 10: Pore structure of SCC fragments at 91 days

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Table 4: Pore size distribution of the investigated mixtures

<i>Pore <math>\varnothing</math> (nm)</i>		<i>Pore size distribution (%)</i>		
		<i>SCC-R</i>	<i>SCC-I</i>	<i>SCC-2</i>
<i>Micro-pores</i>	<i>&lt; 20</i>	74.1	86.3	85.3
<i>Meso-pores</i>	<i>20 - 50</i>	22.7	13.2	14.5
	<i>50 - 200</i>	2.73	0.36	0.11
<i>Macro-pores</i>	<i>200 - 1000</i>	0.42	0.12	0.05
	<i>&gt; 1000</i>	0.05	0.01	0.00

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#### 4.4.2. Interfacial transition zone

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When the concrete is subjected to external forces, stresses are transferred to the ITZ. This factor is very important because the bonding between the cementitious paste and aggregates within the transition zone governs the concrete strength (Brandt, 1995). De Larrard and Belloc (1997) addressed a specific characteristic of the ITZ which is called the maximum paste thickness (MPT), which represents the mean distance between the contiguous CA as follows:

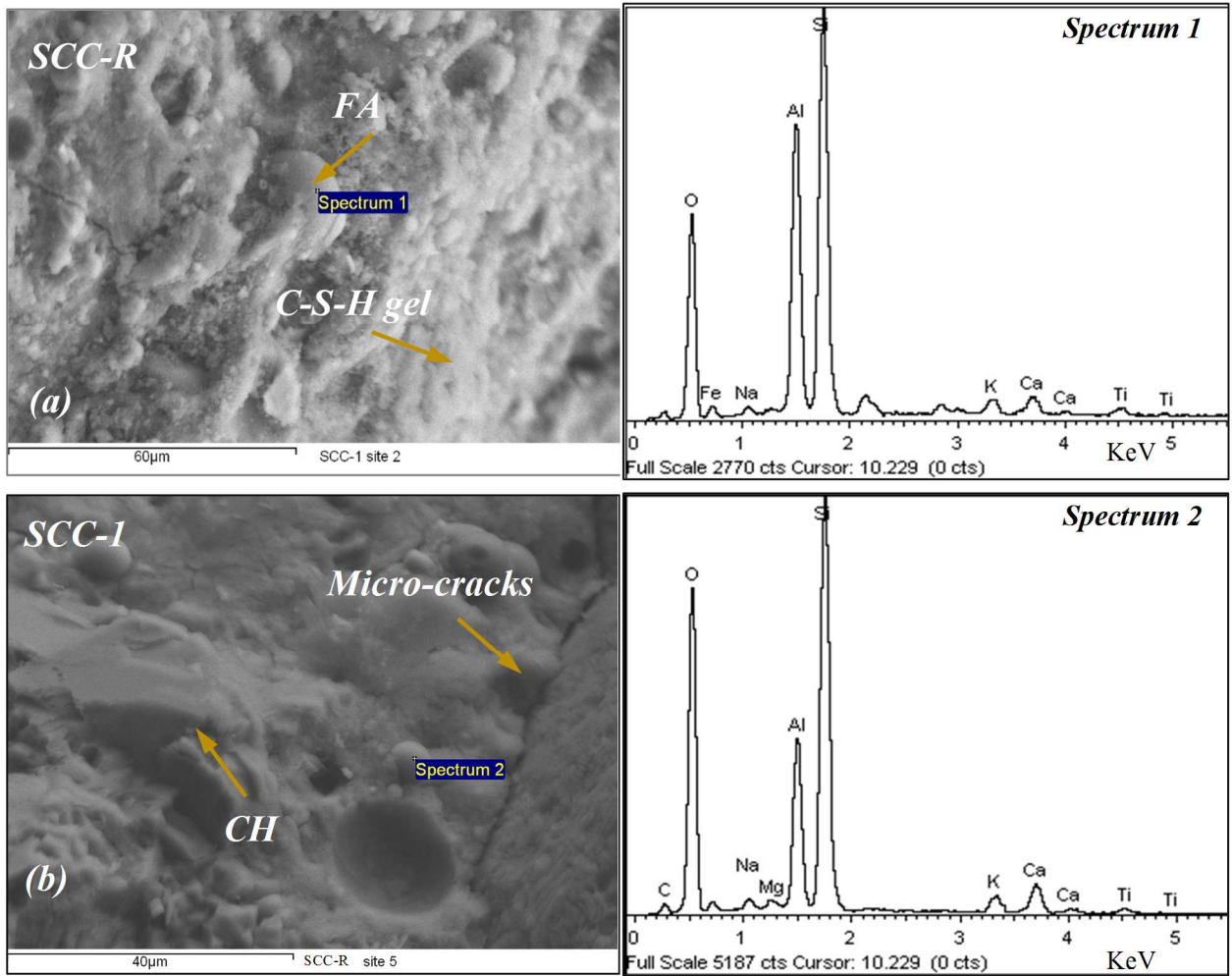
$$334 \quad MPT = D_{max} \left( \sqrt[3]{\frac{g^*}{g}} - 1 \right) \quad \text{where} \quad g^* = 1 - 0.45 \left( \frac{d_{min}}{d_{max}} \right)^{0.19} \quad Eq. 4$$

335 where  $D_{max}$  is the maximum size of aggregate (20 mm),  $g$  is the aggregate volume ( $0.266 \text{ m}^3/\text{m}^3$ ),  $g^*$   
 336 is equal to the PD of the aggregate, and  $d_{min}$  and  $d_{max}$  are the minimum and the maximum sizes of  
 337 aggregate corresponding to 10 (0.18 mm) and 90% (8.86 mm) passing, respectively. MPT is  
 338 considered a small-scale compressive strength test where the cementitious paste is the concrete  
 339 specimen and the two juxtaposed CA are the test machine platens (T. de Grazia et al., 2019). A low  
 340 MPT is associated with higher mechanical properties (Shadkam et al., 2017). The calculated MPT  
 341 in this study was 8.70 mm, which is lower than those observed in previous studies (Shadkam et al.,  
 342 2017; T. de Grazia et al., 2019).

343 To understand the formation of ITZ, the morphology characteristics of fragments of concrete  
 344 were analyzed using a Hitachi S-4700 scanning electronic microscope (SEM). Fig. 11 represents the  
 345 results of the coupled SEM-EDS analysis conducted on SCC-R and SCC-1 fragments. It was  
 346 observed that the C-S-H mineral phases were well-formed, and large amounts of hydration products  
 347 covered the particles forming a dense and homogeneous microstructure. However, for the SCC-2  
 348 mixture (Fig. 12) a disordered structure with a large amount of open pores was observed. In  
 349 addition, numerous sulfate phases were detected in the SCC-2 mixture. This suggests that the ratio  
 350 of calcium sulfate to tricalcium aluminate was high; otherwise, the ettringite tended to be replaced  
 351 by calcium aluminate monosulfate phases. In addition, increasing the TMS content increased the  
 352 calcium silicate content. Diverse CH crystallization was observed in the SCC-2, with thin  
 353 hexagonal and globular shaped portlandite crystals (Alite and Belite) (Fig. 12-c) and a likewise rice  
 354 particles of CH (Fig. 12-b). This fact confirms the low Ca/Si ratio at 20% of incorporation, which  
 355 refers to the poor quality of the blended binder.

356 SEM and XRD analyses of the different SCCs demonstrated that increasing the TMS  
 357 promoted the formation of ettringite, which may play an important role in the drying of the matrix  
 358 and stabilization. The well formation of C-S-H improved the microstructure by improving the solid-

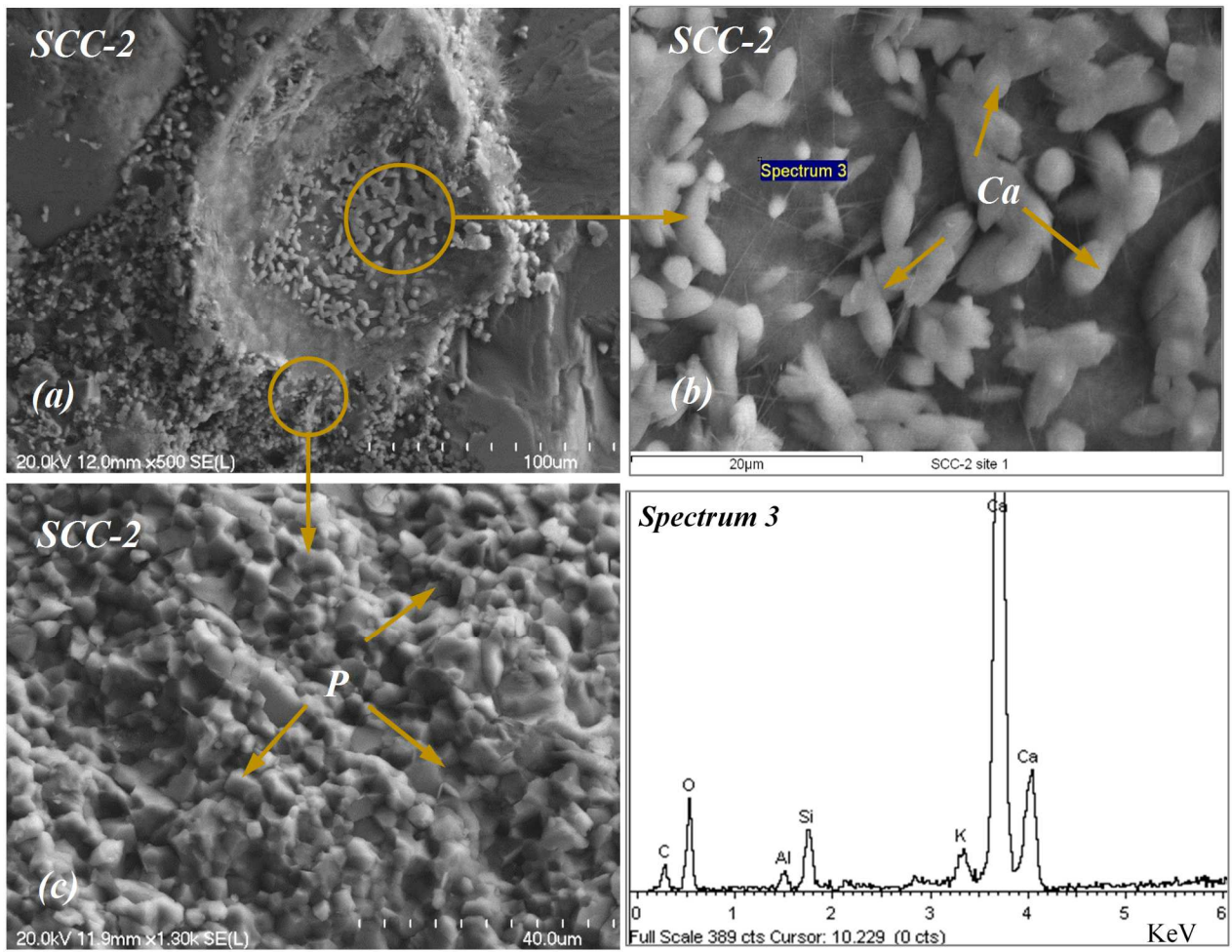
359 to-solid phase connectivity and reducing the pore-to-pore connectivity. Nevertheless, further  
360 investigation of the thermodynamic modeling of blended cement-sediments is highly recommended.



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Fig. 11: SEM-EDS with back-scattered electron analysis of SCC-R and SCC-1 at 91 days



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*Fig. 12: SEM-EDS with back-scattered electron analysis of SCC-2 at 91 days*

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#### 4.5. Leaching test

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To analyze the mobility of trace elements (pollution indicators), a leaching test was performed on ground fragments taken from the investigated SCC mixtures according to the NF EN 12457-2 (2002), using a liquid-to-solid ratio of 10:1 l/kg. The results are summarized in Table 5. Increasing the TMS decreased the barium content in the matrix, which is a highly reactive and toxic metal. A decrease in the concentrations of nickel, sulfates, and soluble fraction was noted as well. In contrast, increasing the TMS content increased the concentration of chrome and chlorides. Concentrations of As, Cd, Mo, Pb, Sb, Se, and Zn were below or close to the detection limits and were in the same order of magnitude, which indicates that there was no significant change in the chemistry and solubility. Overall, the concentration of trace elements in the ground powder of the



375 three SCCs was below the inert waste leaching limit according to the French Directive no. 0289

376 Published on December 14, 2014.

377 *Table 5: Mobility of heavy metals (mg/kg) in the different SCCs*

	<b>SCC-R</b>	<b>SCC-I</b>	<b>SCC-2</b>	<b>Limit</b>
<b>As</b>	< 0.08	< 0.08	< 0.08	0.50
<b>Ba</b>	9.33	2.99	2.78	20.0
<b>Cd</b>	< 0.01	< 0.01	< 0.01	0.04
<b>Cr</b>	0.01	0.19	0.22	0.50
<b>Cu</b>	0.03	0.06	0.05	2.00
<b>Mo</b>	< 0.06	0.08	0.08	0.50
<b>Ni</b>	0.14	< 0.06	< 0.06	0.40
<b>Pb</b>	< 0.03	< 0.03	< 0.03	0.50
<b>Sb</b>	< 0.06	< 0.06	< 0.06	0.06
<b>Se</b>	< 0.09	< 0.09	< 0.09	0.10
<b>Zn</b>	< 0.02	< 0.02	< 0.02	4.00
<b>Fluorides</b>	7.00	9.00	7.00	10.0
<b>Chlorides</b>	13.0	44.0	85.0	800
<b>Sulfates</b>	56.0	47.0	35.0	1000
<b>Soluble Fraction</b>	6057	5221	4835	4000

## 378 **5. Conclusion**

379 This study aimed to evaluate the influence of using treated marine sediments as SCM on the  
380 fresh, physical, hardened, microstructural, and environmental properties of SCC. Based on the  
381 experimental data, the following conclusions can be drawn:

- 382 1. The thermal treatment was efficient and led to an increase in the density and content of  
383 silicone dioxide (higher intensity of diffraction peaks of quartz) and a decrease in the  
384 content of sulfur trioxide.
- 385 2. The investigated SCC mixtures exhibited adequate self-consolidation characteristics with  
386 high filling ability, low  $T_{500}$ , high passing ability, and high stability for SCC production. The  
387 increase in TMS content did not significantly affect the workability.
- 388 3. TMS reduced the heat of binder hydration, which can be a positive effect in the case of mass  
389 concrete or a negative effect (early strength development) depending on the targeted  
390 application.
- 391 4. The investigated SCC mixtures showed similar strength development. The SCC-1 and SCC-  
392 2 mixtures exhibited a compressive strength of  $66 \pm 1$  MPa, which is similar to that of the  
393 reference mixture, at 91 days. Nevertheless, the SCC-2 mixture required a longer curing  
394 time to develop its mechanical properties. The results of the UPV agree with the strength  
395 findings and provided more insight into the development of internal microstructural micro-  
396 cracks in the investigated mixtures.
- 397 5. Microstructure development was improved by the incorporation of TMS. The SCC-1  
398 mixture exhibited the highest volume of micro-pores, and the lowest volume of meso-pores,  
399 followed by the SCC-2 and SCC-R mixtures. The average total porosity of the three  
400 mixtures was approximately 9% with a standard deviation of 0.42%. This low porosity was  
401 mainly due to the pozzolanic reaction of the TMS which refined and increased the density of  
402 the ITZ.
- 403 6. The microscopic examinations indicated that an increase of TMS increased the content of  
404 calcium sulfate and calcium silicate. The SCC-1 and SCC-R mixtures showed similar well-  
405 formed ITZ. However, the microstructure of the SCC-2 mixture was disordered with a large  
406 amount of open pores. Increasing the content of TMS over 20% led to a negative  
407 microstructural effect.

408 7. The above findings, in addition to the leaching test result, should promote the valorization of  
409 dredged sediments in SCC as alternative SCM, thus contributing to the reduction of the  
410 carbon footprint of concrete production.

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## 417 **Declaration of Competing Interest**

418 The authors declare that they have no known competing financial interests or personal relationships  
419 that could have appeared to influence the work reported in this paper.

## 420 **CRedit authorship contribution statement**

421 **A. M. Safhi:** Conceptualization, Data Curation, Formal analysis, Methodology, Validation,  
422 Visualization, Writing - Original Draft, Writing - Review & Editing. **P. Rivard:** Validation,  
423 Resources, Supervision, Project administration, Funding acquisition, Writing - Review & Editing.  
424 **A. Yahia:** Validation, Resources, Supervision, Writing - Review & Editing. **M. Benzerzour:**  
425 Resources, Project administration, Funding acquisition. **K. H. Khayat:** Validation, Writing -  
426 Review & Editing.

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