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1 **Production of Portland cement clinker from French Municipal Solid Waste Incineration**
2 **Bottom Ash**

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11

12 **Abstract**

13 The amount of bottom ashes generated from the municipal solid waste incineration is increasing
14 considerably. Therefore their recycling is necessary in order to limit the landfilling that presents
15 environmental issues. The aim of this paper is to recycle Municipal Solid Waste Incineration Bottom
16 Ash from the north of France; as a secondary raw material for Portland cement clinker production. In
17 this order Municipal Solid Waste Incineration Bottom Ash is characterized and then used to substitute
18 raw materials for clinker production at laboratory scale. Substitutions rates up to 12.4% have been
19 tested. The produced clinkers have been characterized by X-Ray Diffraction and the crystalline phases
20 were quantified by Rietveld analysis. The microstructure has been studied by scanning electronic
21 microscopy. After adding gypsum, the compressive strengths of the corresponding cements were
22 measured at 1, 7 and 28 days. The mineralogical composition of the 28 days hydrated cement pastes
23 were studied by X-Ray Diffraction. The results show that up to 12.4% of Portland cement raw
24 materials can be replaced by the Municipal Solid Waste Incineration Bottom Ash without changing the
25 mineralogical composition. In addition the compressive strengths of the cement pastes produced with
26 Municipal Solid Waste Incineration Bottom Ash are equivalent to the one without bottom ash.

27 **Keywords:** Clinker; Compressive strength, Municipal Solid Waste Incineration Bottom Ash, Recycling

28 **1 Introduction**

29 The cement industry is responsible for more than 6% of the total anthropogenic greenhouse gases
30 emissions. In fact the production of Ordinary Portland Cement "OPC" generates 842 kg CO₂/t of
31 clinker [1][2][3][4][5]. In addition to its huge energy consumption, the cement industry uses large
32 amounts of raw materials for cement production [6]. Therefore the replacing of raw materials in the
33 clinker raw meal by waste as secondary material, gains a lot of attention [2][7][8][9]. This replacement
34 allows not only to preserve the natural resources and to recover wastes, but also to reduce the CO₂
35 emissions. Different types of wastes have been studied in the literature in order to produce cement,
36 and mainly OPC cement, such as red mud [10], steel slag [11], sediments [12][13], recycled concrete
37 aggregate [14], oil based mud cutting [15], oyster and scallop shell [16] etc.

38 In the European Union, the Municipal Solid Waste (MSW) production is significantly increasing. In
39 2015, each inhabitant generated about 477 kg of MSW per year [17] [18]. The incineration presents
40 the most common method to treat these wastes as an alternative to landfilling[19] and reduces
41 between 70 and 90% the volume of wastes [20][21]. The MSWI ash is a by-product from the
42 combustion of MSW [22][21][23][24][25]. Two types of ashes are produced following the incineration
43 operation: bottom ash and fly ash. The recycling of these ashes is a concern especially that their
44 quantities are increasing considerably and due to their highly variable composition.

45 The MSWI bottom ash (MSWI BA) represents 75-80% of the produced ashes [26][27][28]. Nowadays,
46 the landfilling presents the main option for MSWI BA disposal [29][30]. However, the landfill causes the
47 pollution of the groundwater and contaminates the soil in addition to the limited land space and the
48 problems related to the disposal cost and the regulation [31][32][33]. The literature describes different
49 methods to recycle the MSWI BA in civil engineering. The latter has been used as supplementary
50 cementitious materials and pozzolanic material [29][27][34][35], as an aggregate replacement
51 [19][28][36][37] or in alkali activated materials as a precursor [20][38][39], as a road based material
52 and in road construction [40][41][42].

53 The chemical composition of MSWI BA contains CaO, SiO₂, Al₂O₃ and Fe₂O₃ [43][44]. This
54 composition makes the use of MSWI BA, as a secondary raw material, for clinker production
55 possible[45]. Li et al. analyzed different fraction from MSWI BA from Beijing to produce a type II
56 Portland cement and shows that 9% of this waste could be added to the raw meal [46]. Pan et al. also
57 used MSWI fly ash and bottom ash, after washing with water/acid as a pre-treatment, to produce
58 clinker by incorporating 1.75 and 3.5% respectively in the raw meal [47]. The percentages of
59 incorporation were determined based on the chloride content in the ashes in order to respect the limit
60 of chloride of 100 ppm required by the cement plant in Taiwan. They showed that this incorporation did
61 not affect the compressive strength and the cement produced is conformed to the Chinese National
62 Standard of type II Portland cement [47].

63 It is important to note that the MSWI BA has a very heterogeneous composition depending on several
64 factors such as the waste origin, the incineration process, the incinerator used, the pre-treatment
65 applied, etc. [48][49][50]. Therefore the percentage of substitution in the clinker raw meal differs from
66 one to another MSWI BA. Until today, there are no studies dealing with the use of MSWI BA generated
67 in France, as a secondary raw material for clinker production. Therefore the aim of this study is to use
68 Municipal Solid Waste Incineration Bottom Ash from the north of France, as a raw material in the OPC
69 clinker production. First a reference OPC clinker is produced without adding MSWI BA. Then 2
70 substitution rates of MSWI BA are applied: 5% and a second one corresponding to the maximum
71 amount of MSWI BA that could be incorporated while respecting the targeted composition of the
72 reference clinker. The mechanical properties of the resulting cement are compared to reference OPC
73 cement produced as well as the hydration products.

74 **2 Materials and methods**

75 The materials used in this study for the clinker production are the chemical reagents: CaCO₃, SiO₂,
76 Al₂O₃, Fe₂O₃ and the MSWI BA from the north of France.

77 **2.1 MSWI BA characterization**

78 The MSWI-BA used in this study originates from the recycling platform RMN-PréFerNord located in
79 Fretin in the region of "Hauts-de-France" in France. This platform treats between 150,000 and 180,000
80 tons yearly and is considered to be among the largest in France in terms of quantities and the
81 techniques used for the process of preparation of the MSWI-BA. MSWI-BA was subjected to a

82 maturation period of two to four months in open air and was considered recyclable in the field of roads'
 83 construction according to the French regulation of 18 November 2011 [51]. The fraction 0-2 mm of
 84 Municipal Solid Waste Incineration Bottom Ash (MSWI BA) is crashed and then sieved at 100 μm . The
 85 mineralogical characterization was made by X-ray diffraction (XRD) using a diffractometer Bruker D2
 86 with Cu K α radiation. The X-ray patterns were acquired in the 2θ (10-80 $^\circ$) with a step of 0.02 $^\circ$ and 1 s
 87 per step. The chemical composition was analyzed by X-ray fluorescence (XRF). The particle size
 88 distribution was determined using the laser granulometer COULTER type LS 230.

89 2.2 Clinker production at laboratory scale

90 Three Portland clinkers are produced in the laboratory with different percentage of substitution by
 91 MSWI BA. The first is a reference clinker referred by OPC0 produced using analytical reagents
 92 (CaCO_3 , SiO_2 , Al_2O_3 and Fe_2O_3) only. The theoretical mineralogical composition of OPC0 is shown in
 93 table 1. This composition was chosen according to the study of Gineys et al [8] in order to produce an
 94 Ordinary Portland Cement.

95 **Table 1 : Theoretical composition of the targeted Portland clinker**

C_3S (wt. %)	C_2S (wt. %)	C_3A (wt. %)	C_4AF (wt. %)	C (wt. %)
65%	18%	8%	8%	1%

96

97 For the MSWI BA based clinker, the solver option in Microsoft Excel was used to adjust and calculate
 98 the amount of the different raw materials [14] when substituting with MSWI BA. This option uses the
 99 chemical composition, determined by XRF for the different raw materials and MSWI BA (% wt. of Ca,
 100 Si, Al, Fe), with the aim to respect the targeted cement criteria represented in table 1.

101 The first substitution rate in the clinker raw meal by MSWI BA corresponds to 5% (OPC5). The second
 102 one corresponds to the maximal amount of MSWI BA that could be incorporated in the raw meal while
 103 respecting the theoretical composition of the targeted Portland cement.

104 Table 2 shows the mixes designs of the 3 clinker raw meals obtained by solver and the percentage of
 105 each component to add, in order to reach the theoretical composition of table 1. As it can be seen from
 106 the table 2, the maximum amount of MSWI BA that could be incorporated in the raw meal is controlled
 107 by the Fe_2O_3 content (OPC12.4). Adding more than 12.4% of MSWI BA (maximum value) in the raw
 108 meal provides more Fe_2O_3 than needed and consequently the resulting clinker composition differs
 109 from the theoretical composition of the targeted Portland clinker.

110

Table 2: Mixes designs for the 3 produced clinker raw meals

Constituents	CaCO_3 (wt. %)	SiO_2 (wt. %)	Al_2O_3 (wt. %)	Fe_2O_3 (wt. %)	MSWI BA (wt. %)
OPC0	79.98	15.35	2.92	1.75	0
OPC5	77.45	14.11	2.39	1.05	5
OPC12.4	73.71	12.28	1.61	0	12.4

111

112 The method to produce clinker consists to mix all materials together by adding water to better
 113 homogenize. The mix is then dried at 105 $^\circ\text{C}$ and pressed at 5 kN into pellets to obtain a better
 114 clinking process. Then the pellets are fired up 30 min at 200 $^\circ\text{C}$ then to 1450 $^\circ\text{C}$ with a rate of 10 $^\circ\text{C}$.
 115 After 15 min of burning at the clinking temperature, the clinker was cooled in the furnace. The
 116 obtained clinkers are ground until a Blaine specific area of 4500 cm^2/g . To produce cements, gypsum
 117 was added to the clinker using equation 4 [52]:

118 Amount of gypsum used = Al_2O_3 content * [0.6/101.96] * [80/0.461] (equation 4)

119 Where, 101.96 = mol. wt. Al_2O_3 ; 80 = mol. wt. SO_3 ; 0.461 = mol. ratio SO_3 /natural gypsum used (for
 120 pure gypsum 80/172 is 0.466)

121 2.3 Clinker and cement paste characterization

122 The mineralogy of the synthesized clinkers was studied using X-ray diffraction (XRD). A Bruker D2
 123 with Cu K α radiation was used. The X-ray patterns were acquired in the 2θ (10-80 $^\circ$) with a step size
 124 of 0.02 $^\circ$ and 1 s per step. A Rietveld analysis was also conducted using TOPAS software in order to
 125 quantify the crystalline phases.

126 The Microscopic observations were conducted on polished section of clinkers using SEM type Hitachi
 127 S-4300SE/N. Local chemical analyses were conducted using energy dispersive X-ray spectrometer
 128 (EDS) with a Thermo Scientific Ultra Dry EDS detector coupled with the SEM. The polished sections
 129 were made by vacuum impregnating a clinker monolith in an epoxy resin. After hardening, the samples
 130 were polished with ethanol and then coated with carbon.

131 The reactivity of the cement pastes was followed by isothermal calorimetry performed at 20°C. The
 132 water/cement ratio used was 0.5. The calorimeter was a home-made calorimeter using fluxmeters that
 133 allowed the calorimeter to equilibrate in less than 5 min [53].

134 The hydrates of the 4 different cement pastes are identified by XRD analysis at 28 days after stopping
 135 the hydration by immersing the cement pastes - already grounded into powder – in ethanol. After few
 136 minutes of mixing with ethanol, a filtration is done using the Buchner filter. The powder is then
 137 collected and dried in the oven at 50°C.

138 The compressive strength was determined on cement paste small cubes 1*1*1 cm (due to the limited
 139 quantities of cement produced in the laboratory according to [51]) made of cement pastes with a
 140 water/cement ratio (w/c) of 0.5. Cement and water were manually mixed. The filling of the molds
 141 (polytetrafluoroethylene (PTFE) molds) was performed in two steps on a vibration table. The molds
 142 were kept for 24 h at 100% relative humidity at 20 °C. The paste cubes were demolded after 24 h and
 143 curing was continued at 100% relative humidity at 20 °C. The compressive strength was measured
 144 after 1, 7 and 28 days [53], [54].

145 3 Results and discussions

146 3.1 MSWI BA characterization

147 Table 3 presents the bottom ash composition as well as the soluble salts. The results show that MSWI
 148 BA contains all the necessary major elements for the clinker production (Ca, Si, Al and Fe).

149 **Table 3: Bottom ash composition**

Element	LOI 950°C	O	Ca	Fe	Al	Si	P	S	Cl	Pb
Percentage (%)	13.99	34.06	18.91	9.45	5.57	11.06	0.76	0.94	0.25	0.17
Element	K	Na	Ti	Cr	Mn	Mg	Cu	Zn	Ba	
Percentage (%)	0.6	1.25	0.44	0.1	0.19	1.13	0.24	0.63	0.11	
soluble salts (mg/kg)										
Fluoride					<20					
Chloride					2740					
Sulphate					5059					

150

151 The X-ray diffraction (figure 1) for MSWI BA shows that the principal phases are quartz, calcite, with
 152 some minor phases such as hematite, gehlenite etc.

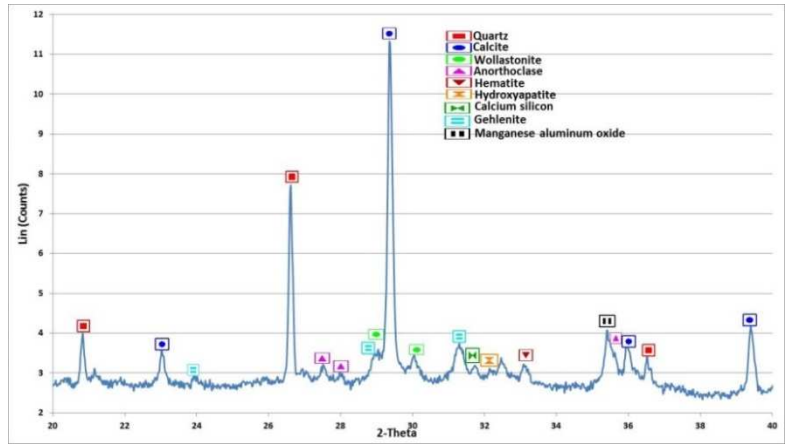


Figure 1: X-ray pattern for MSWI BA

153
154

155 The particle size distribution of MSWI BA is represented in figure 2. The results show that 90% of
156 particles are inferior to 70 μm . In fact the finesse of the raw materials is important in order to have a
157 better reactivity.

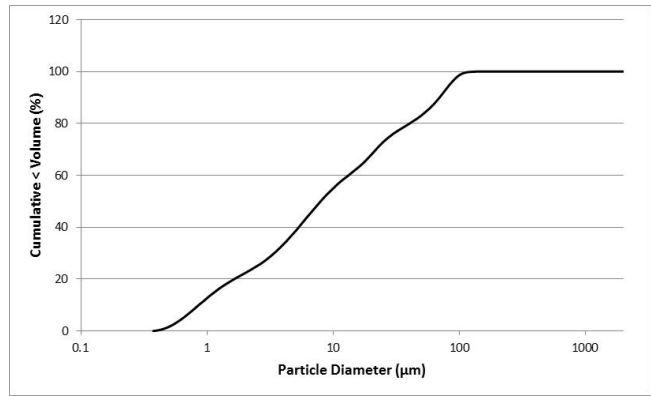


Figure 2: Particle size distribution for MSWI BA

158
159

160 3.2 Clinkers characterization

161 3.2.1 Mineralogical and chemical analyzes

162 The OPC0 reference clinker characterization by XRD is presented in figure 3. The result shows the
163 presence of the 4 crystalline phases of a Portland clinker, C_3S , C_2S , C_4AF and C_3A . The pattern shows
164 also that no secondary mineral phases are produced. Therefore the raw meal composition, the clinker
165 preparation process at the laboratory scale and the burning temperature are suitable for this study.

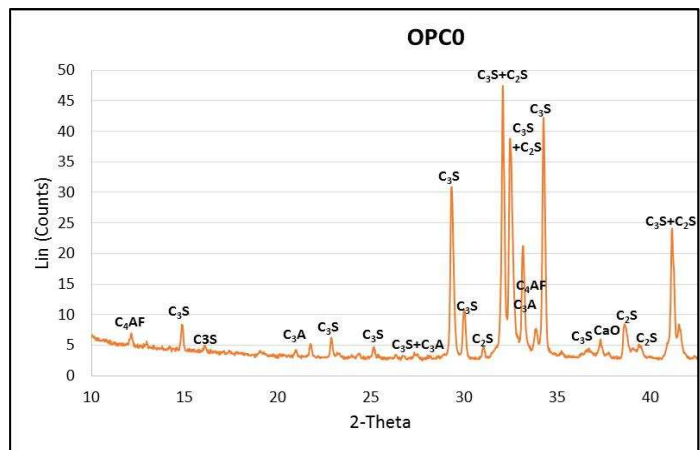
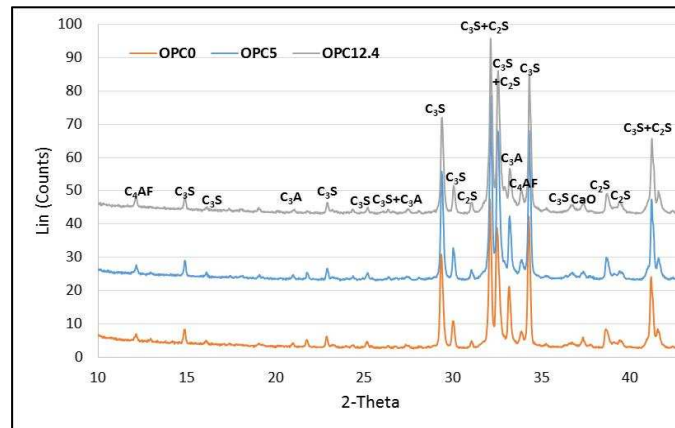


Figure 3: XRD patterns of the reference clinker OPC0

166
167

168 The MSWI BA based clinkers OPC5 and OPC12.4 are characterized and compared to the reference
 169 clinker. The results are shown in the figure 4. As it can be seen from the patterns, the 2 MSWI BA
 170 based clinkers, OPC5 and OPC12.4 present the same mineralogical phases as the reference clinker
 171 OPC0. Consequently the partial substitution of the raw materials by MSWI BA does not modify the
 172 mineralogical composition of the produced clinkers.



173
 174 **Figure 4: XRD results for the reference clinker OPC0 and for the MSWI BA based clinker OPC5 and OPC12.4**

175 The three produced clinkers were subjected to an elementary analyzes by XRF in order to detect the
 176 presence of heavy metals and alkalis (Table 4). The results show that some heavy metals (Zn) and
 177 alkali (Na) slightly increases while increasing the MSWI BA substitution. The presence of these heavy
 178 metals could have an influence on the hydration mechanisms and on the setting time of the cement by
 179 forming a heavy metal hydroxide on the surface of the anhydrous cement grains, as suggested by
 180 Gineys et al. studies [55][56].

181 **Table 4: XRF analyzes for the 3 produced clinkers**

Element (%)	L.O.I (%)	O	Ca	Si	Fe	Al	Na	Mg	P	Ti	Zn
OPC0	3	38.8	47.3	9.9	1.8	2.2	-	-	-	-	-
OPC5	2.7	38.4	47.3	9.9	1.9	2.0	0.2	0.1	<0.1	<0.1	<0.1
OPC12.4	2.6	38.7	46.4	10.1	1.9	2.0	0.2	0.2	0.1	0.1	0.1

182 In order to quantify the crystalline phases in the 3 synthesized clinkers, Rietveld analysis was
 183 conducted. The results are represented in the table 5 and show that adding MSWI BA in the raw meal
 184 produces a clinker with slightly less C₃S comparing to the reference clinker. This could be due to the
 185 consideration in the calculation method that the four clinker phases are in their stoichiometric state
 186 which is not quite realistic especially when incorporating wastes into the raw meal where the phases
 187 can accommodate substitute ions [57]. Same results was obtained by Clavier et al. 2021 [43] when
 188 incorporating bottom ash in the clinker. This was attributed to the operational variations associated
 189 with lab-scale clinker production. In fact the C₃S tends to revert to C₂S when clinker is cooled too
 190 slowly and therefore the speed of cooling showed be considered. In addition the Portland cement
 191 chemistry is complex, therefore this complexity increases when a replacing the raw materials by a
 192 heterogeneous material such as MSWI BA. The latter contains, as shown in table 3, some metallic
 193 elements (i.e. Cu, Zn, Mg, Cl...) that could cause a reduction in the mineralization temperature,
 194 favoring therefore the formation of C₂S over the C₃S phase[58]. Concerning the residual lime, its
 195 content is between 0.05% and 0.21% (table 5) which confirms that the clinkering was complete
 196 because these values are lower than what is specified in the standard NF EN 196-2. In addition,
 197 according to the standard there is no risk due to these low free lime contents (<0.5%).

198 **Table 5: Rietveld analysis for the 3 clinkers: OPC0, OPC5 and OPC12.4**

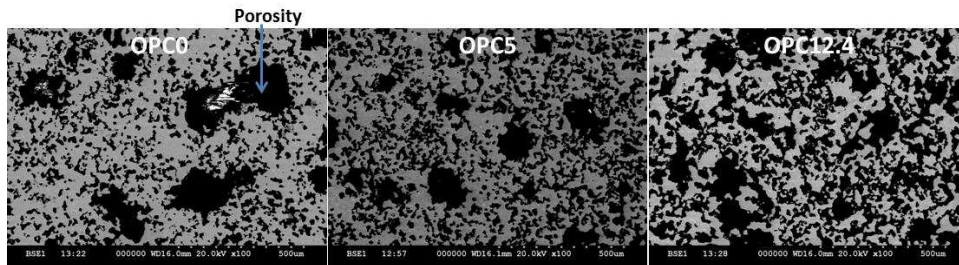
	C ₃ S (wt. %)	C ₂ S (wt. %)	C ₄ AF (wt. %)	C ₃ A (wt. %)	CaO (wt. %)	GoF
OPC0	76.14	12.27	3.5	7.88	0.21	1.65
OPC5	74.01	15.21	4.07	6.66	0.05	1.44
OPC12.4	72.12	19.60	4.13	4.09	0.06	1.45

200 **3.2.2 Microstructure observation**

201 The microstructure of the 3 produced clinkers was observed by SEM/EDS on polished sections (figure
202 5). By comparing the 3 samples, it can be seen that the porosity (black areas) of the clinker matrix
203 increases while increasing the incorporation percentage of MSWI BA. According to the literature, this
204 behavior could be due to the presence of heavy metals (i.e. Zn) that could change the viscosity of the
205 interstitial liquid phase of clinkers during the sintering process. In fact, zinc acts as a mineralizer [59],
206 therefore the porosity of the clinkers could be influenced [60] [61].

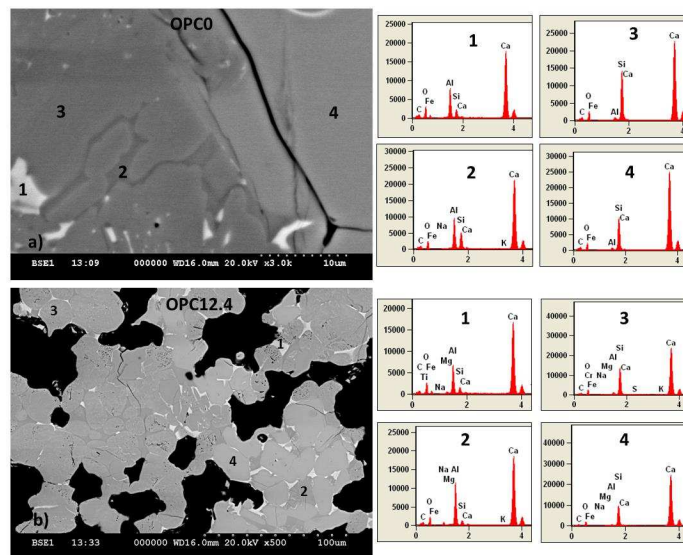
207

Figure 5: Microstructural observations of the 3 clinkers: OPC0, OPC5 and OPC12.4



208

209 In order to identify the mineralogical phases of clinkers, elemental analyzes by EDS were conducted
210 on OPC0 and OPC12.4 (figure 6). In both clinkers, C₄AF (point 1), C₃A (point 2), C₂S (point 3) and C₃S
211 (point 4) were identified. The alite grains appeared to be angular whereas the belite one round. The
212 clinkers, with and without BA, present in general almost a similar microstructures.



213

214

Figure 6: SEM/EDS spectra of the 2 clinkers OPC0 and OPC12.4

215 **3.3 Calorimetric study**

216 The hydration reactions of the cement paste are exothermic and can be monitored by following the
217 rate of heat release using an isothermal calorimeter. Figure 8 shows the calorimetric curves for the
218 three cement pastes with and without BA incorporation: OPC0, OPC5 and OPC12.4. It can be seen
219 from the results that the three cement pastes have a similar curve shape. Three peaks were formed
220 during the first 36 hours of hydration. The first peak corresponds to the dissolution of the anhydrous
221 grains and appears immediately after adding water to cement. Then an induction period with low
222 thermal activity was observed for the three cement paste before appearance of the second peak. The
223 latter corresponds to the acceleration of the silicates and aluminates hydration and the formation of C-
224 S-H and ettringite. A third peak is formed after the gypsum depletion and the transformation of
225 ettringite to monosulfoaluminate hydrate.

226 By comparing the three curves a main difference can be detected related to the initial setting time. In
227 fact the setting of OPC0 begins approximately 1 hour before OPC5 and OPC12.4. This could be due
228 to the trace elements (i.e. Zn) that are present in the MSWI BA based clinkers as already shown in the
229 XRF results (table 4). According to previous studies, Zn is well known to have a retarder effect on the

230 cement paste hydration [56][55]. In addition, the presence of alkalis such as Na could also induce a
231 retardation effect [62].

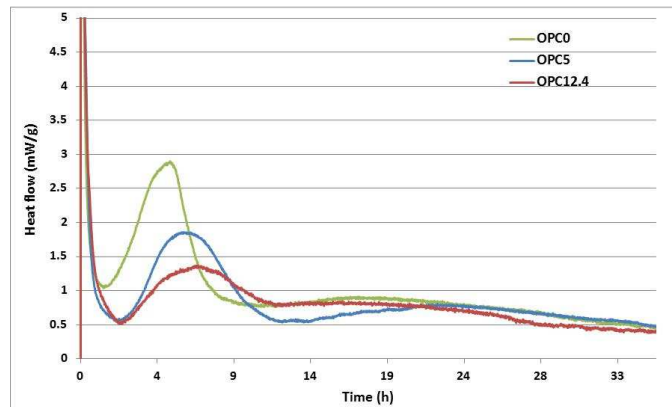


Figure 7 : The heat release for the three cement pastes: OPC0, OPC5 and OPC12.4

232
233

234 3.4 Mechanical performances

235 The compressive strength is measured in order to evaluate the mechanical performances of the
236 produced cement. Figure 7 represents the compressive strength at 1, 7 and 28 days for the three
237 cement pastes: OPC0, OPC5 and OPC12.4. These results show that at early age (1 day) the
238 compressive strengths is lower when MSWI BA is added (19.5, 14 and 10.7 MPa for OPC0, OPC5
239 and OPC12.4 respectively). This could be related to the retardation effect observed by isothermal
240 calorimetry at 1 day caused by the heavy metals and alkalis as previously shown in figure 8. This
241 could be related also to the amount of C_3S in the clinker (table 4) that is a slightly higher in the clinker
242 containing less MSWI BA. In addition the presence of alkali such as sodium and potassium could also
243 have a retardation effect on the hydration and influencing therefore the compressive strength [63].
244 This difference in the compressive strength of the three cement pastes is less observed from day 7
245 because the effect of the delay of the hydration appears at very short term. It can be concluded that
246 using MSWI BA as a substitution for the raw materials for clinker production does not change
247 significantly the compressive strength of the cement pastes especially after 7 days.

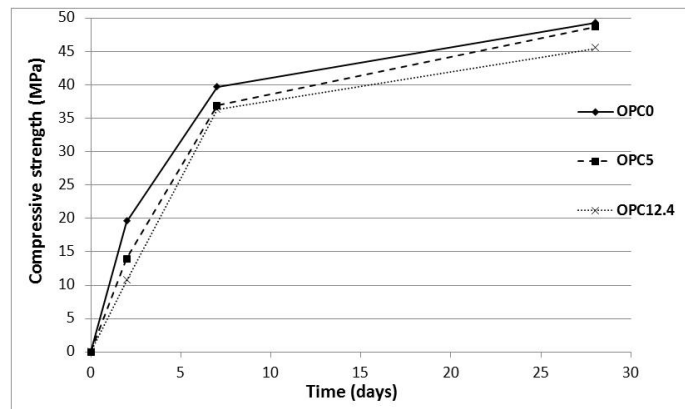


Figure 8: Compressive strength of the 3 cement pastes at 1, 7 and 28 days

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249

250 The mineralogical assemblages of the hydrated cement pastes at 28 days were studied by XRD
251 analysis after stopping the hydration (figure 9). The results show that the Portlandite CH is the main
252 crystalline hydrate. This phase comes from the hydration of C_3S and C_2S . The patterns show also the
253 presence of Ettringite AFt coming from the reaction of C_3A with gypsum.

254 The diffractograms of the 3 cement pastes (reference and the 2 cement pastes with MSWI BA) are
255 similar. Therefore adding MSWI BA in the raw meal to produce Portland cement does not modify the
256 mineralogical assemblage of the hydrated cement pastes.

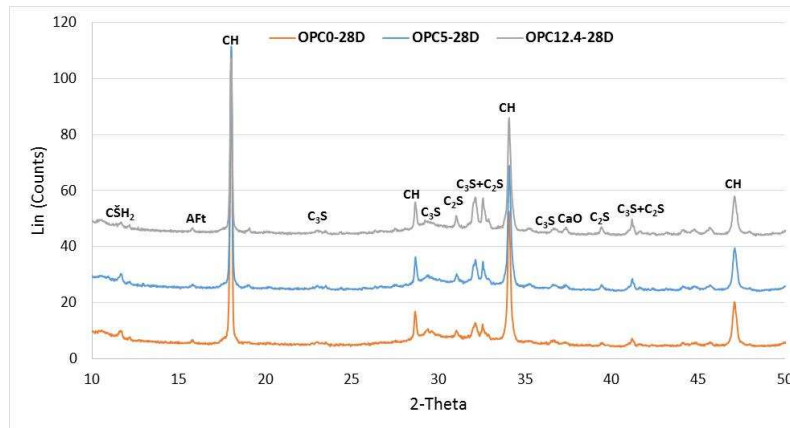


Figure 9: XRD analysis for the 3 cement pastes OPC0-28D, OPC5-28D and OPC12.4-28D at 28 days

257
258

259 4 Conclusion

260 The aim of this paper is to recycle the MSWI BA from the north of France, in the raw meal to produce
 261 Portland clinker. To reach this target, MSWI BA was grinded and characterized in order to identify its
 262 composition. In addition to the reference clinkers made with chemical reagents, two other clinkers
 263 were synthesized: OPC5 with 5% of MSWI BA and OPC12.4 with 12.4% that represents the maximum
 264 amount of substitution. It has been shown in this study that:

- 265 - MSWI BA contains the main oxides - CaO, SiO₂, Al₂O₃, Fe₂O₃ - needed for the clinker
 266 production
- 267 - For the clinker composition chosen, the maximum amount of MSWI BA that could be
 268 incorporated in the raw meal is controlled by the Fe₂O₃ amount. The addition of 12.4% of
 269 MSWI BA provides the total amount of iron oxide needed for the clinker production
- 270 - Adding MSWI BA in the raw meal for the clinker production does not affect the mineralogical
 271 composition comparing to the reference clinker. The main phases of Portland clinker – C₃S,
 272 C₂S, C₄AF and C₃A in addition to free lime are obtained for all produced clinkers
- 273 - The microstructure of the clinker with and without BA is quite similar despite the presence of
 274 more porosity in the MSWI BA based clinker which could be due to the clinkering process and the
 275 presence of some heavy metals
- 276 - The partial substitution of raw materials by MSWI BA provoked a delay of less than 1 hour in
 277 the setting time which could be attributed to the presence of heavy metals and alkalis.
- 278 - The difference between the compressive strength of MSWI BA-based clinkers and the
 279 reference are slight after 7 days of hydration.

280 Finally, this study reveals that the bottom ash could be recycled as a secondary raw material for
 281 clinker production. The amount of bottom ash that could be recycled depends mainly on its
 282 composition and the type of cement targeted.

283 In our case we conclude that the French bottom ash has different chemical composition from the
 284 ones studied in the literature (coming from Taiwan and Beijing [46] [47]). With this type of bottom
 285 ash, it is possible to recycle 12.4% in CEM I Portland cement after adapting the raw meal
 286 composition and without applying a treatment for the bottom ash.

287 Given the large variability of the bottom ash composition, due to the type and the origin of wastes
 288 that differs from a country to another, it is important to extend the study to investigate the
 289 variability of the bottom ash composition from a batch to another and the effect that could have on
 290 its possible use as a raw material for cement production. In addition it is interesting to follow in
 291 further studies the durability of the cement pastes made from MSWI BA based clinkers in order to
 292 have a complete vision on the behavior of these clinkers.

293 Declaration of competing interest

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

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References

- [1] E. Gartner, "Industrially interesting approaches to 'low-CO₂' cements," *Cem. Concr. Res.*, vol. 34, no. 9, pp. 1489–1498, 2004, doi: 10.1016/j.cemconres.2004.01.021.
- [2] E. Gartner and H. Hirao, "A review of alternative approaches to the reduction of CO₂ emissions associated with the manufacture of the binder phase in concrete," *Cem. Concr. Res.*, vol. 78, pp. 126–142, 2015, doi: 10.1016/j.cemconres.2015.04.012.
- [3] L. H. J. Martin, F. Winnefeld, C. J. Muller, and B. Lothenbach, "Contribution of limestone to the hydration of calcium sulfoaluminate cement," *Cem. Concr. Compos.*, vol. 62, pp. 204–211, 2015, doi: 10.1016/j.cemconcomp.2015.07.005.
- [4] M. C. G. Juenger, F. Winnefeld, J. L. Provis, and J. H. Ideker, "Advances in alternative cementitious binders," *Cem. Concr. Res.*, vol. 41, no. 12, pp. 1232–1243, 2011, doi: 10.1016/j.cemconres.2010.11.012.
- [5] K. L. Scrivener, V. M. John, and E. M. Gartner, "Eco-efficient cements: Potential economically viable solutions for a low-CO₂ cement-based materials industry," *Cem. Concr. Res.*, vol. 114, no. March, pp. 2–26, 2018, doi: 10.1016/j.cemconres.2018.03.015.
- [6] H. X. Chen, X. Ma, and H. J. Dai, "Reuse of water purification sludge as raw material in cement production," *Cem. Concr. Compos.*, vol. 32, no. 6, pp. 436–439, 2010, doi: 10.1016/j.cemconcomp.2010.02.009.
- [7] H. X. Chen, X. Ma, and H. J. Dai, "Reuse of water purification sludge as raw material in cement production," *Cem. Concr. Compos.*, vol. 32, no. 6, pp. 436–439, 2010, doi: 10.1016/j.cemconcomp.2010.02.009.
- [8] N. Gineys, G. Aouad, F. Sorrentino, and D. Damidot, "Incorporation of trace elements in Portland cement clinker: Thresholds limits for Cu, Ni, Sn or Zn," *Cem. Concr. Res.*, vol. 41, no. 11, pp. 1177–1184, 2011, doi: 10.1016/j.cemconres.2011.07.006.
- [9] M. Schneider, M. Romer, M. Tschudin, and H. Bolio, "Sustainable cement production-present and future," *Cem. Concr. Res.*, vol. 41, no. 7, pp. 642–650, 2011, doi: 10.1016/j.cemconres.2011.03.019.
- [10] P. E. Tsakiridis, S. Agatzini-Leonardou, and P. Oustadakis, "Red mud addition in the raw meal for the production of Portland cement clinker," *J. Hazard. Mater.*, vol. 116, no. 1–2, pp. 103–110, 2004, doi: 10.1016/j.jhazmat.2004.08.002.
- [11] P. E. Tsakiridis, G. D. Papadimitriou, S. Tsvilis, and C. Koroneos, "Utilization of steel slag for Portland cement clinker production," *J. Hazard. Mater.*, vol. 152, no. 2, pp. 805–811, 2008, doi: 10.1016/j.jhazmat.2007.07.093.
- [12] G. Aouad, A. Laboudigue, N. Gineys, and N. E. Abriak, "Dredged sediments used as novel supply of raw material to produce Portland cement clinker," *Cem. Concr. Compos.*, vol. 34, no. 6, pp. 788–793, 2012, doi: 10.1016/j.cemconcomp.2012.02.008.
- [13] A. Faure *et al.*, "Beneficial reuse of dam fine sediments as clinker raw material," *Constr. Build. Mater.*, vol. 218, pp. 365–384, 2019, doi: 10.1016/j.conbuildmat.2019.05.047.
- [14] C. Diliberto, A. Lecomte, J. M. Mechling, L. Izoret, and A. Smith, "Valorisation of recycled concrete sands in cement raw meal for cement production," *Mater. Struct. Constr.*, vol. 50, no. 2, pp. 1–12, 2017, doi: 10.1617/s11527-017-0996-8.
- [15] H. S. Al Dhamri, S. A. Abdul-Wahab, C. Velis, and L. Black, "Oil-based mud cutting as an additional raw material in clinker production," *J. Hazard. Mater.*, vol. 384, no. February 2019, p. 121022, 2020, doi: 10.1016/j.jhazmat.2019.121022.
- [16] S. Her, T. Park, E. Zalnezhad, and S. Bae, "Synthesis and characterization of cement clinker using recycled pulverized oyster and scallop shell as limestone substitutes," *J. Clean. Prod.*, vol. 278, p. 123987, 2021, doi: 10.1016/j.jclepro.2020.123987.
- [17] Eurostat, "Eurostat. 477 kg of municipal waste generated per person in the EU," <https://ec.europa.eu/eurostat/web/products-eurostat-news/-/DDN-20170130-1>.
- [18] J. Malinauskaite *et al.*, "Municipal solid waste management and waste-to-energy in the context of a circular economy and energy recycling in Europe," *Energy*, vol. 141, pp. 2013–2044, 2017, doi: 10.1016/j.energy.2017.11.128.
- [19] M. Antoun, F. Becquart, N. Gerges, and G. Aouad, "The use of calcium sulfo-aluminate cement as an alternative to Portland Cement for the recycling of municipal solid waste incineration bottom ash in mortar," *Waste Manag. Res.*, vol. 38, no. 8, pp. 868–875, 2020, doi: 10.1177/0734242X20925170.
- [20] B. Jurič, L. Hanžič, R. Ilič, and N. Samec, "Utilization of municipal solid waste bottom ash and recycled aggregate in concrete," *Waste Manag.*, vol. 26, no. 12, pp. 1436–1442, 2006, doi: 10.1016/j.wasman.2005.10.016.
- [21] H. F. Hassan, "Recycling of municipal solid waste incinerator ash in hot-mix asphalt concrete," *Constr. Build. Mater.*, vol. 19, no. 2, pp. 91–98, 2005, doi: 10.1016/j.conbuildmat.2004.05.010.
- [22] R. Siddique, "Utilization of municipal solid waste (MSW) ash in cement and mortar," *Resour. Conserv. Recycl.*, vol. 54, no. 12, pp. 1037–1047, 2010, doi: 10.1016/j.resconrec.2010.05.002.
- [23] K. A. Clavier, B. Watts, Y. Liu, C. C. Ferraro, and T. G. Townsend, "Risk and performance assessment of cement made using municipal solid waste incinerator bottom ash as a cement kiln feed," *Resour. Conserv. Recycl.*, vol. 146, no. January, pp. 270–279, 2019, doi: 10.1016/j.resconrec.2019.03.047.
- [24] R. V. Silva, J. de Brito, C. J. Lynn, and R. K. Dhir, "Environmental impacts of the use of bottom ashes from municipal solid waste incineration: A review," *Resour. Conserv. Recycl.*, vol. 140, no. June 2018, pp. 23–35, 2019, doi: 10.1016/j.resconrec.2018.09.011.
- [25] A. Mary Joseph, R. Snellings, P. Nielsen, S. Matthys, and N. De Belie, "Pre-treatment and utilisation of

- 364 municipal solid waste incineration bottom ashes towards a circular economy," *Constr. Build. Mater.*, vol.
365 260, p. 120485, 2020, doi: 10.1016/j.conbuildmat.2020.120485.
- 366 [26] L. M. Sarmiento, K. A. Clavier, J. M. Paris, C. C. Ferraro, and T. G. Townsend, "Critical examination of
367 recycled municipal solid waste incineration ash as a mineral source for portland cement manufacture – A
368 case study," *Resour. Conserv. Recycl.*, vol. 148, no. April, pp. 1–10, 2019, doi:
369 10.1016/j.resconrec.2019.05.002.
- 370 [27] Z. Chen and E. H. Yang, "Early age hydration of blended cement with different size fractions of municipal
371 solid waste incineration bottom ash," *Constr. Build. Mater.*, vol. 156, pp. 880–890, 2017, doi:
372 10.1016/j.conbuildmat.2017.09.063.
- 373 [28] P. J., L. Coutaz, J. Ambroise, and M. Chababbet, "Use of incinerator bottom ash in concrete," vol. 27, no.
374 1, pp. 1–5, 1997.
- 375 [29] A. Abdulmatin, W. Tangchirapat, and C. Jaturapitakkul, "An investigation of bottom ash as a pozzolanic
376 material," *Constr. Build. Mater.*, vol. 186, pp. 155–162, 2018, doi: 10.1016/j.conbuildmat.2018.07.101.
- 377 [30] X. Dou *et al.*, "Review of MSWI bottom ash utilization from perspectives of collective characterization,
378 treatment and existing application," *Renew. Sustain. Energy Rev.*, vol. 79, no. May 2016, pp. 24–38,
379 2017, doi: 10.1016/j.rser.2017.05.044.
- 380 [31] M. Herrero, A. Laca, and M. Díaz, "Life cycle assessment focusing on food industry wastes," *Food Ind.
381 Wastes*, pp. 265–280, 2013, doi: 10.1016/B978-0-12-391921-2.00015-9.
- 382 [32] P. Kjeldsen, M. A. Barlaz, A. P. Rooker, A. Baun, A. Ledin, and T. H. Christensen, "Present and long-term
383 composition of MSW landfill leachate: A review," *Crit. Rev. Environ. Sci. Technol.*, vol. 32, no. 4, pp. 297–
384 336, 2002, doi: 10.1080/10643380290813462.
- 385 [33] S. Mukherjee, S. Mukhopadhyay, M. A. Hashim, and B. Sen Gupta, "Contemporary environmental issues
386 of landfill leachate: Assessment and remedies," *Crit. Rev. Environ. Sci. Technol.*, vol. 45, no. 5, pp. 472–
387 590, 2015, doi: 10.1080/10643389.2013.876524.
- 388 [34] C. Jaturapitakkul and R. Cheerarot, "Development of bottom ash as pozzolanic material," *J. Mater. Civ.
389 Eng.*, vol. 15, no. 1, pp. 48–53, 2003, doi: 10.1061/(ASCE)0899-1561(2003)15:1(48).
- 390 [35] P. Tang, M. V. A. Florea, P. Spiesz, and H. J. H. Brouwers, "Application of thermally activated municipal
391 solid waste incineration (MSWI) bottom ash fines as binder substitute," *Cem. Concr. Compos.*, vol. 70, pp.
392 194–205, 2016, doi: 10.1016/j.cemconcomp.2016.03.015.
- 393 [36] S. Sorlini, A. Abbà, and C. Collivignarelli, "Recovery of MSWI and soil washing residues as concrete
394 aggregates," *Waste Manag.*, vol. 31, no. 2, pp. 289–297, 2011, doi: 10.1016/j.wasman.2010.04.019.
- 395 [37] T. Zhang and Z. Zhao, "Optimal Use of MSWI Bottom Ash in Concrete," *Int. J. Concr. Struct. Mater.*, vol.
396 8, no. 2, pp. 173–182, 2014, doi: 10.1007/s40069-014-0073-4.
- 397 [38] Z. Zhang, G. Q. Li, Y. X. Yang, and Y. P. Xiao, "Thermodynamics of the vitrified bottom ash slag from
398 municipal solid waste incinerators-phase relations of CaO-SiO₂-Na₂O oxide system," *Adv. Mater. Res.*,
399 vol. 881–883, pp. 574–578, 2014, doi: 10.4028/www.scientific.net/AMR.881-883.574.
- 400 [39] Z. Chen, Y. Liu, W. Zhu, and E. H. Yang, "Incinerator bottom ash (IBA) aerated geopolymer," *Constr.
401 Build. Mater.*, vol. 112, pp. 1025–1031, 2016, doi: 10.1016/j.conbuildmat.2016.02.164.
- 402 [40] R. Forteza, M. Far, C. Seguí, and V. Cerdá, "Characterization of bottom ash in municipal solid waste
403 incinerators for its use in road base," *Waste Manag.*, vol. 24, no. 9, pp. 899–909, 2004, doi:
404 10.1016/j.wasman.2004.07.004.
- 405 [41] I. Vegas, J. A. Ibañez, J. T. San José, and A. Urzelai, "Construction demolition wastes, Waelz slag and
406 MSWI bottom ash: A comparative technical analysis as material for road construction," *Waste Manag.*,
407 vol. 28, no. 3, pp. 565–574, 2008, doi: 10.1016/j.wasman.2007.01.016.
- 408 [42] T. Van Gerven, D. Geysen, L. Stoffels, M. Jaspers, G. Wauters, and C. Vandecasteele, "Management of
409 incinerator residues in Flanders (Belgium) and in neighbouring countries. A comparison," *Waste Manag.*,
410 vol. 25, no. 1, pp. 75–87, 2005, doi: 10.1016/j.wasman.2004.09.002.
- 411 [43] K. A. Clavier, J. M. Paris, C. C. Ferraro, E. T. Bueno, C. M. Tibbetts, and T. G. Townsend, "Washed
412 waste incineration bottom ash as a raw ingredient in cement production: Implications for lab-scale clinker
413 behavior," *Resour. Conserv. Recycl.*, vol. 169, no. December 2020, p. 105513, 2021, doi:
414 10.1016/j.resconrec.2021.105513.
- 415 [44] N. Saikia, S. Kato, and T. Kojima, "Production of cement clinkers from municipal solid waste incineration
416 (MSWI) fly ash," *Waste Manag.*, vol. 27, no. 9, pp. 1178–1189, 2007, doi:
417 10.1016/j.wasman.2006.06.004.
- 418 [45] K. A. Clavier, J. M. Paris, C. C. Ferraro, and T. G. Townsend, "Opportunities and challenges associated
419 with using municipal waste incineration ash as a raw ingredient in cement production – a review," *Resour.
420 Conserv. Recycl.*, vol. 160, no. April, p. 104888, 2020, doi: 10.1016/j.resconrec.2020.104888.
- 421 [46] Y. Li, L. Hao, and X. Chen, "Analysis of MSWI Bottom Ash Reused as Alternative Material for Cement
422 Production," *Procedia Environ. Sci.*, vol. 31, pp. 549–553, 2016, doi: 10.1016/j.proenv.2016.02.084.
- 423 [47] J. R. Pan, C. Huang, J. J. Kuo, and S. H. Lin, "Recycling MSWI bottom and fly ash as raw materials for
424 Portland cement," *Waste Manag.*, vol. 28, no. 7, pp. 1113–1118, 2008, doi:
425 10.1016/j.wasman.2007.04.009.
- 426 [48] O. Hammoud *et al.*, "Influence of chemical pre-treatment on the leaching behaviour of bottom ash," 2019.
- 427 [49] A. Vaitkus, J. Gražulytė, V. Vorobjovas, O. Šernas, and R. Kleizienė, "Potential of MSWI bottom ash to be
428 used as aggregate in road building materials," *Balt. J. Road Bridg. Eng.*, vol. 13, no. 1, pp. 77–86, 2018,
429 doi: 10.3846/bjrbe.2018.401.
- 430 [50] A. M. Joseph, R. Snellings, P. Van den Heede, S. Matthys, and N. De Belie, "The use of municipal

- 431 solidwaste incineration ash in various building materials: A Belgian point of view," *Materials (Basel)*, vol.
 432 11, no. 1, 2018, doi: 10.3390/ma11010141.
- 433 [51] "Government of France (2011) Arrêté ministériel du 18 novembre 2011 relatif au recyclage en technique
 434 routière des mâchefers d'incinération de déchets non dangereux, 18 novembre 2011. Version consolidée,
 435 8 pages,"
 436 <https://www.legifrance.gouv.fr/affichTexte.do?cidTexte=JORFTEXT000024873229&categorieLien=id> .
- 437 [52] Bhatti and I. Javed, *Effect of Minor Elements on Clinker and Cement Performance*. 2006.
- 438 [53] J. Kleib, G. Aouad, N. Khalil, and M. Zakhour, "Incorporation of zinc in calcium sulfoaluminate cement
 439 clinker," *Adv. Cem. Res.*, 2021, doi: <https://doi.org/10.1680/jadcr.19.00125>.
- 440 [54] N. Gineys, G. Aouad, F. Sorrentino, and D. Damidot, "Effect of the clinker composition on the threshold
 441 limits for Cu, Sn or Zn," *Cem. Concr. Res.*, vol. 42, no. 8, pp. 1088–1093, 2012.
- 442 [55] N. Gineys, G. Aouad, and D. Damidot, "Managing trace elements in Portland cement - Part I: Interactions
 443 between cement paste and heavy metals added during mixing as soluble salts," *Cem. Concr. Compos.*,
 444 vol. 32, pp. 563–570, 2010.
- 445 [56] N. Gineys, G. Aouad, and D. Damidot, "Managing trace elements in Portland cement - Part II:
 446 Comparison of two methods to incorporate Zn in a cement," *Cem. Concr. Compos.*, vol. 33, no. 6, pp.
 447 629–636, 2011, doi: 10.1016/j.cemconcomp.2011.03.008.
- 448 [57] I. A. Chen and M. C. G. Juenger, "Incorporation of waste materials into portland cement clinker
 449 synthesized from natural raw materials," *J. Mater. Sci.*, vol. 44, no. 10, pp. 2617–2627, 2009, doi:
 450 10.1007/s10853-009-3342-x.
- 451 [58] D. Herfort, G. K. Moir, V. Johansen, F. Sorrentino, and H. B. Arceo, "The chemistry of Portland cement
 452 clinker," *Adv. Cem. Res.*, vol. 22, no. 4, pp. 187–194, 2010.
- 453 [59] K. G. Kolovos, S. Barafaka, G. Kakali, and S. Tsivilis, "CuO and ZnO addition in the cement raw mix:
 454 Effect on clinkering process and cement hydration and properties," *Ceram. - Silikaty*, vol. 49, no. 3, pp.
 455 205–212, 2005.
- 456 [60] H. M. Ludwig and W. Zhang, "Research review of cement clinker chemistry," *Cem. Concr. Res.*, vol. 78,
 457 pp. 24–37, 2015, doi: 10.1016/j.cemconres.2015.05.018.
- 458 [61] Y. Lin, S. Zhou, F. Li, and Y. Lin, "Utilization of municipal sewage sludge as additives for the production of
 459 eco-cement," *J. Hazard. Mater.*, vol. 213–214, pp. 457–465, 2012, doi: 10.1016/j.jhazmat.2012.02.020.
- 460 [62] S. T. Witzleben, "Acceleration of Portland cement with lithium, sodium and potassium silicates and
 461 hydroxides," *Mater. Chem. Phys.*, vol. 243, no. July 2019, p. 122608, 2020, doi:
 462 10.1016/j.matchemphys.2019.122608.
- 463 [63] S. T. Witzleben, "Acceleration of Portland cement with lithium, sodium and potassium silicates and
 464 hydroxides," *Mater. Chem. Phys.*, vol. 243, no. July 2019, p. 122608, 2020, doi:
 465 10.1016/j.matchemphys.2019.122608.
- 466