1	Cold-bonding process for treatment and reuse of waste materials: technical
2	designs and applications of pelletized products
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#### 43 Abstract

This work provides a comprehensive review of research on the cold-bonding pelletization 44 process used to produce lightweight aggregates (LWAs) using waste materials, to valorise 45 the waste and, at the same time, minimize risks related to disposal. Research investigating 46 various aspects of the cold-bonding process highlight: i) feasible mix-designs for pellet 47 production; ii) the most relevant operating parameters affecting the process; and iii) the 48 49 potential applications of the LWAs produced. The analysis gives a wide overview of the fundamental key-points that control the cold-bonding process. Data comparison provides a 50 51 useful way to identify the optimal process conditions to allow development of optimum products. This involves the selection of the correct mix-design, including suitable binders 52 and potential additives, and the selection of appropriate operating conditions, which are a 53 54 function of the waste investigated, and/or waste mix characteristics. The review proposes 55 an optimised approach to experimental studies on cold-bonding processes that has potential to enhance future process performance. Moreover, the present work provides a complete 56 57 framework useful for decision-making for both manufacturers and researchers working to use this promising technique. 58

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61 Key words: Cold-bonding process; Pelletization efficiency; Lightweight aggregates;
62 Lightweight concrete; Aggregate mix-designs

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## 66 **1.** Introduction

Stabilization/solidification (S/S) has been widely used in the last few decades on many types of 67 waste materials as a remediation solution, which allows pollutant containment and prevents 68 leaching of hazardous compounds (Senneca et al., 2020). While the initial applications of S/S 69 were focused on the production of a stabilized waste for environmentally safe disposal, it is 70 now being used as a technology to allow the possible reuse of stabilized waste products as 71 72 aggregates to be used in the construction industry (Colangelo, Cioffi, Montagnaro, & Santoro, 2012; Narattha & Chaipanich, 2018). An interesting application of this is the manufacture of 73 74 artificial lightweight aggregates (LWA) (Ayati, Ferrándiz-Mas, Newport, & Cheeseman, 2018; Velis et al., 2014). LWA can be produced through pelletization which is an agglomeration 75 phenomena that occurs to moist waste fines containing a binder (Baykal & Döven, 2000). The 76 77 pelletized LWA product can then be obtained by sintering, which involves heating the aggregates after pelletization, or by cold-bonding which allows LWA pellets to form at ambient 78 temperature (Ayati, Molineux, Newport, & Cheeseman, 2019; Bui, Hwang, Chen, & Hsieh, 79 2012). Despite the rapid production of a readily usable product by sintering, the high 80 temperatures (900-1400 °C) needed are a disadvantage compared to the cold-bonding process 81 which requires less energy consumption and reduces secondary pollution formation (Thomas 82 & Harilal, 2015). In recent years several studies have investigated cold-bonding pelletization 83 processes to determine optimal operating conditions (Frankovič, Bosiljkov, & Ducman, 2017; 84 85 Gesoğlu, Güneyisi, & Öz, 2012). These have determined suitable mix-designs for the production of LWA with appropriate properties. It is evident that the final characteristics of the 86 stabilized products have wide variability depending on numerous factors affecting the process. 87 88 From this perspective, the present work aims to provide a comprehensive and technical overview of scientific studies focused on cold-bonding pelletization reported in the scientific 89 literature. This can provide a significant contribution by analysing and identifying useful and 90

complete information to support the operational practices of both researchers and industrial manufacturers working on the cold-bonding process. For this aim, this work focuses on studies concerning the effect of operating parameters on the process performance including the waste/binder ratio, the waste properties, angle and rotation speed of the pelletizer, the process duration, and the aggregate curing method. Finally, studies related to the possible applications of pelletized materials are analysed to highlight the benefits resulting from using cold-bonding processes.

98

# 99 2. Cold-bonding processing

100 The cold-bond process uses a rotating plate, which causes pellet formation for moist materials 101 because of the plate rotation effect. This allows the formation of granular structures with enhanced bonding forces due to centrifugal and gravitational forces (Arslan & Baykal, 2006). 102 The pelletization process is characterized by three stages. These are the i) the pendular state 103 104 (water on the grains contact point), ii) the funicular state (water filling some pores) and, iii) the capillary state (water filling all intergranular spaces) which represents the best pelletization 105 condition (Baykal & Döven, 2000). The first step of the pelletization process is represented by 106 107 the mixture wetted by water addition. The nucleation phase allows formation of granule nuclei, which are loosely packed. Depending on the agitation intensity and granule nuclei resistance, 108 109 the consolidation phase can take place through collisions of different nuclei. During collisions, the granules bond to form larger particles by coalescence. Strong bond formation among 110 granules should occur in order to achieve efficient particle formation (Hapgood, lveson, Litster, 111 & Liu, 2007). 112

113 The pelletization process is mainly driven by gravitational/centrifugal forces. Operating 114 parameters that control the process are the rotation velocity and inclination of the pelletizer plate as well as the amount of water in the mix. These parameters will be discussed in moredetail.

117 The overall processing scheme is summarized in Fig. 1. The main element of the pelletizer is 118 the rotating plate and this exerts the rotation effect on the mix particles. A speed regulator adjusts the rotational velocity of the plate, while plate inclination can also be controlled. Water 119 addition can be performed either manually or in automated mode. In this latter case, the 120 121 pelletizer is equipped with a nebulizer and related nozzles for spraying water on the mixture. Moreover, one or more scrapers is generally placed in different positions on the rotating plate. 122 The scrapers are used to remove the material adhering to both the surface and side of the rotating 123 124 plate to avoid unused particles in the process.

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## 126 **3.** Mix-design for cold-bonding pelletization

#### 127 **3.1** Mix-design for pellets using fly-ash as waste material

Coal and municipal solid waste incineration fly-ashes (FA) are probably the waste materials 128 most frequently used in the cold-bonding pelletization process (Table 1) because of the large 129 130 available amount of these wastes which can be stabilized/solidified and used as aggregate (G. Joseph & Ramamurthy, 2009a). Cold-bonded aggregates made from FA have mechanical 131 132 properties similar to commercial LWAs (Rivera, Martínez, Castro, & López, 2015). However, the correct mix-design to achieve good results is challenging, with LWA properties strongly 133 depending on the FA chemical composition. For example, high CaO content may lead to 134 reduced crushing strengths of LWA pellets (Gesoğlu, Özturan, & Güneyisi, 2007). 135

Cold-bonded pellets prepared using just FA have low quality compared to pellets produced
using additional binders (Baykal & Döven, 2000), particularly in terms of compressive strength,
specific gravity and water absorption capacity. Similarly, cold-bonded pellets prepared using

mechanically activated FA mixed with Na<sub>2</sub>SiO<sub>3</sub> have much worse mechanical properties (about
50% lower), compared to sintered pellets prepared using the same mix-design (Terzić, Pezo,
Mitić, & Radojević, 2015). The same occurs for cold-bonded pellets prepared using FA alkali
activated with NaOH and mixed with bentonite (Gomathi & Sivakumar, 2015). Cold-bonded
pellets produced using FA and phase change materials (PCM) had poor properties, particularly
as the percentage of PCM in the mix increased (Tuncel & Pekmezci, 2018).

Improved performance of cold-bonded pelletized FA aggregates was obtained by adding
Portland cement (Arslan and Baykal, 2006). FA pellets also increased in specific gravity, bulk
unit weight and particle strength and the water absorption capacity decreased when the cement
addition increased from 10 to 20% (Chi et al., 2003).

The addition of  $Ca(OH)_2$  to FA pellets can also produce pellets with good characteristics, although the samples formed were not as good in terms of pellet density, water absorption and crushing strength as samples made using cement. Further additions of  $Ca(OH)_2$  reduced the quality of the pellets formed (Narattha & Chaipanich, 2018).

The addition of more components in the cold-bonded pellet mix-design may change the 153 properties. For example, the three-component mixture containing FA, cement and granulated 154 blast furnace slag (GGBS) and the three-component mixture containing, FA, cement and rice 155 husk ash (RHS) have been investigated. These had very different properties. The crushing 156 strength was much higher for pellets containing GGBS compared to pellets containing RHS 157 (Bui, Hwang, Chen, & Hsieh, 2012). This is because the GGBS is alkali activated and this 158 produces additional C-S-H, the main binding phase in cementitious materials. The GGBS 159 samples also had decreased water absorption, due to the higher density of the aggregate 160 microstructure associated with increased formation of C-S-H gel (Bui, Hwang, Chen, & Hsieh, 161

162 2012). The positive effects of GGBS compared to RHS were also observed for cold-bonded163 pellets produced using two-component mixtures without cement.

An increase of strength, absorption capacity and unit weight is reported with increasing GGBS 164 165 content in a two-component mix containing FA+GGBS (from 15.5 to 15.7 MPa, from 7.8 to 8.3%, from 1003 to 1060 kg m<sup>-3</sup>, respectively). For the two-component mixtures containing 166 FA+RHS, values of strength, absorption capacity and unit weight varied in a range from 6 to 167 8.1 MPa, from 10.8 to 20.5%, and from 833 to 769 kg m<sup>-3</sup>, respectively. Improved enhancement 168 in properties is also observed compared to the three-component mixtures containing both 169 GGBS and RHS with FA (from 8.1 to 8.8 MPa, from 9.8 to 10.1%, from 855 to 894 kg m<sup>-3</sup>, 170 171 respectively) (Bui, Hwang, Chen, Lin, & Hsieh, 2012).

The partial replacement of FA (up to 40%) with expanded perlite powder also produces an 172 173 important improvement in aggregate mechanical properties (Tajra, Elrahman, Chung, & Stephan, 2018). Similarly, the partial replacement of FA with bentonite or kaolinite may 174 175 improve the mechanical characteristics of cold-bonded pellets. This clearly indicates the 176 beneficial effects on process performances through the use of clay minerals (Manikandan & Ramamurthy, 2007). It is reported that high swelling bentonite leads to a higher pelletization 177 efficiency compared to medium swelling bentonite at the same dosage. This is due to the higher 178 179 amount of interlayer cations in the high swelling bentonite causing the formation of more expanded montmorillonite platelet fibres available for the attraction FA particles (Manikandan 180 & Ramamurthy, 2009). 181

Improved mechanical strength and water absorption capacity are observed using an aggregate mixture containing FA+furnace slag, alkali activated using NaOH solution (Gomathi & Sivakumar, 2014). In this case, a crushing strength of 22.8 MPa, and a water absorption capacity of 13.0% were observed. The latter values are preferable compared to the crushing strength of 14.5 MPa and to the water absorption capacity of 16.4% obtained using a mixture alkali
activated of FA+bentonite, or 17.6 MPa, and 17.9% observed for mixture alkali activated of
FA+metakaolin. Similar results are obtained for this two-component mixture using other alkali
activators (Yliniemi, Nugteren, et al., 2016; Yliniemi, Tiainen, & Illikainen, 2016). However
the use of sodium silicate may not avoid leaching of anionic complexes of heavy metals in the
stabilized pellets (Yliniemi, Pesonen, et al., 2016).

192 The partial replacement of FA with different wastes for cold-bonded pellets production can be beneficial not only in terms of process efficiency, but also as a way to recycle waste. Tang & 193 Brouwers (2017) report an overall improvement of pellets properties through the addition of 194 195 different components such as bottom ash (BA) fines, paper sludge ash (PSA), and polypropylene fibre (PPF) to the aggregates mixture. LWA pellets production used different 196 mixtures of weathered FA, wastewater treatment sludge and desulfurization device sludge with 197 lime addition (Ferone et al., 2013). Indeed, binary mixtures of weathered FA+wastewater 198 treatment sludge or desulfurization device sludge, and ternary mixture of the same materials 199 200 show feasible properties for LWA production.

Hwang & Tran (2015) analysed the effect of hydrogen peroxide as foaming agent for the 201 production of foamed LWA from binary mixture of FA+GGBS and ternary mixture of 202 203 FA+GGBS+cement. Despite the positive effect on foamed LWA properties with optimal hydrogen peroxide percentage (7%), results show decreasing values of crushing strength with 204 increasing hydrogen peroxide concentrations for both FA+GGBS (80%+20%, respectively) and 205 FA+GGBS+cement (70%+20%+10%, respectively) mixtures. A further study focused on the 206 production of geopolymer aggregates from fluidized bed combustion FA and mine tailing 207 208 material (Yliniemi, Paiva, Ferreira, Tiainen, & Illikainen, 2017). In this case, results displayed better mechanical properties of FA geopolymer aggregates due to the higher crushing force 209 required compared to the mine tailing geopolymer aggregates. 210

Finally, Colangelo, Messina, & Cioffi (2015), implemented a double step cold-bonding process for pellets production using municipal solid waste incineration (MSWI) FA from rotary and stoker furnaces. In this case, tests were carried out on different mixtures containing MSWI FA+cement, MSWI FA+lime, and MSWI FA+coal FA, subjected to a second pelletization step using a 1:1 cement/coal FA binder. This approach increased the mechanical properties of the produced cold-bonded pellets, reducing, at the same time, the risk of metal leaching which can be significantly consistent for the MSWI FA (Ferraro et al., 2019).

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#### 219 **3.2 Mix-design for pellets with ore tailings as waste material**

220 The cold-bonding technique has been widely applied for the stabilization/solidification of ore fines tailings produced by mining and milling activities (Table 1). Research studies in this field 221 mainly refer to iron ore tailing (IOT). Pioneering investigations (Dutta et al., 1992; Dutta, 222 Bordoloi, & Borthakur, 1997) indicate that the most suitable mix-design to be used for cold-223 bonded pellets containing IOT, must include cement/cement clinkers and other components. 224 225 Indeed cold-bonded pellets produced with GGBS and Portland cement clinker in the ratio of 1:1, 2:3, and 3:2, had higher crushing strength compared to pellets containing larger amounts 226 of clinker (Dutta et al., 1992). Similarly, cold-bonded pellets with suitable mechanical 227 228 characteristics can be obtained adding silica fines (quartz silica or RHA) in the mixture. Moreover, it is reported that production of pellets with good characteristics can be possible also 229 with lowered (from 10% to 4-6%) amount of binder (by enhancing the binder surface area to a 230 value of about 4100 cm<sup>2</sup> g<sup>-1</sup> (Dutta et al., 1997). As an alternative to calcium silicate cements 231 (i.e. Portland cement), calcium aluminate cement, used at a similar level (7 wt%) also leads to 232 233 the production of iron ore pellets with enhanced strength (Aota, Morin, Zhuang, & Clements, 2006). 234

Other inorganic binders used for the production of cold-bonded pellets containing IOT, are bentonite and Ca(OH)<sub>2</sub>. Bentonite dosed at about 0.5%, gave positive results especially for high Blaine numbers (fineness level) of hematite iron ore pellets (Pal, Ghorai, Agarwal, et al., 2015). Ca(OH)<sub>2</sub> dosed between 10% and 14% (w/w) produced very high compressive strengths for siliceous IOT (McDonald, Roache, & Kawatra, 2016).

Organic binders such as molasses, have been used and added to the mixtures in percentages ranging between 20% and 50% (Cevik, Ahlatci, & Sun, 2013). This produced high compressive strengths in cold-bonded pellets, but at high dosages, it has an adverse effect on pellets porosity. A positive effect on pellets strength due to molasses addition can also be obtained coupling molasses with calcined lime (added, respectively in percentages of 4 wt% and 10 wt%), as reported by Pal, Ghorai, & Das (2015). In particular, they tested these mixtures for the production of cold-bonded pellets of IOT containing high amount of coal (38 wt%).

Good performances are reported in case of mixture design including slaked lime coupled with dextrose. The cold-bonded pellets produced are characterized by increasing compressive strength, as the amount in the mixture increases up to 10% (Sah & Dutta, 2010).

Finally, very high compressive strength were obtained for IOT cold-bonded pellets containing
dextrin in a range between 1 and 5% (Agrawal et al., 2000). However, carboxymethyl-cellulose,
dosed between 1-2%, produced a better performance than dextrin, even if this latter is coupled
with bentonite, or substituted by calcium-lignosulfonate (Nikai & Garbers-Craig, 2016).

The cold-bonding process has been tested also on pyrrhotite and gold mill tailings (Amaratunga, 1995; Amaratunga & Hmidi, 1997). For both wastes several mixtures have been tested, with various composition of Portland cement and gypsum  $\beta$ -hemihydrate, added at different percentages. The results indicate that, to achieve a suitable compromise between reduced Portland cement consumption and high pellets strength, the best solution has 10% binder dosage, containing gypsum  $\beta$ -hemihydrate and Portland cement in a ratio of 40:60 for pyrrhotite tailings (Amaratunga, 1995). In the case of gold mill tailings, the best result was observed for
4% binder dosage, containing gypsum β-hemihydrate and Portland cement in a ratio of 20:80
(Amaratunga & Hmidi, 1997).

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#### **3.3 Mix-design for pellets with different waste materials**

Although FA are more frequently used for the production of cold-bonded pellets, BA is also 265 266 used (Table 1). This can positively affect the characteristics of the final LWA, especially in terms of crushing resistance (Tang, Florea, & Brouwers, 2017). Both cement and lime can be 267 used as binders to produce cold-bonded pellets containing BA. However the use of cement 268 269 produces higher aggregate strength (Geetha & Ramamurthy, 2010a). To improve the pellets strength, is also possible to use chemical activation of the binder through Na<sub>2</sub>SO<sub>4</sub> addition, 270 which accelerates early pozzolanic activity (Geetha & Ramamurthy, 2010a). The addition of 271 Ca(OH)<sub>2</sub> to the mixtures increases pelletization efficiency and reduces pelletization times. In 272 terms of pelletization efficiency, higher values correspond to binder percentages ranging around 273 274 14% for cement, lime and high swelling bentonite. Different values of binder percentages are reported around 25% for medium swelling bentonite and around 30% for clay (with plasticity 275 index of 78 and 108), kaolin, and metakaolin (Geetha & Ramamurthy, 2010b). 276

Ca(OH)<sub>2</sub> and Na<sub>2</sub>SO<sub>4</sub> have also been used as pelletization and strength enhancing admixtures for cold-bonding of lignite and bituminous pond ash (Vasugi & Ramamurthy, 2014). For the pond ash overall properties were enhanced by increasing Ca(OH)<sub>2</sub> and Na<sub>2</sub>SO<sub>4</sub> content, especially when the used binder is cement instead of lime. In the case of lignite pond ash, the characteristics of cold-bonded pellets improved with increasing Ca(OH)<sub>2</sub> and increasing hardening admixture (80% cement, 9% Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, 6% Na<sub>2</sub>CO<sub>3</sub> and 5% CaO) dosage (Vasugi & Ramamurthy, 2014). Better results are obtained for both the pelletization efficiency and the mechanical characteristics of the produced pellets, including the bulk density, the waterabsorption capacity, the open porosity and the mechanical strength.

Referring to other waste typologies (Table 1), it has to be mentioned the production of cold-286 287 bonded pellets composed by 35% basic oxygen furnace coarse sludge, 20% basic oxygen fine sludge, 20% blast furnace dust, 10% briquette fines, 5% filter dust, and 10% of Portland cement, 288 289 tested by Robinson (2005). In addition, it has to be considered either the production of cold-290 bonded pellets containing iron oxides, produced using blast furnace slag (6%) and clinker (4%) as binders (Camci, Aydin, & Arslan, 2002), and the use of various wastes as binders for quarry 291 fines pellets production (Gunning, Hills, & Carey, 2009). For these latter, the quarry 292 293 fines+waste mixtures are treated through accelerated carbonation during the pelletization phase. This approach allows the production of aggregates characterized by final physical and 294 mechanical properties, which are comparable or even better than the corresponding 295 296 characteristics of lightweight expanded clay aggregate.

The cold-bonding process has also been used to treat electric arc furnace dust (EAF), car fluff 297 298 and fluxed hematite concentrate (Colangelo, Messina, Di Palma, & Cioffi, 2017; Mantovani & Takano, 2000; Zhou, Wattanaphan, & Kawatra, 2017). The addition of 3 or 5 wt% of Portland 299 cement as binder produced good results in terms of cold compressive strength of pellets 300 containing EAF and coal fine mixtures (Mantovani & Takano, 2000). Further tests focused on 301 the determination of compressive strength after heating of EAF+coal fine pellets containing 5 302 wt% Portland cement and this indicated the need to reduce pellet size (max diameter=7 mm) to 303 obtain high strengths (Mantovani, Takano, & Büchler, 2002). Colangelo et al. (2017) suggest 304 305 the possibility of using up to 43.5% of car fluff to produce cold-bonded pellets.

306 It can be observed that higher crushing strength and reduced water absorption capacity result 307 from decreasing the water to binder ratio, when coal FA and cement are used as binders. Finally, 308 the study by Zhou et al. (2017) indicates that it is possible to produce cold-bonded pellets containing fluxed hematite concentrate, using humic acid as the binder. In this case, results
display good characteristics of the produced aggregate when the binder dosage is around 0.6
wt%.

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# **4.** Parameters affecting the cold-bonding process efficiency

The use of various waste types and mix-designs leads to different results in terms of aggregate quality and properties. However, cold-bonding process efficiency is also dependent on operational parameters (Table 2). The three parameters which mainly affect the pellets characteristics are the inclination angle and rotation speed of the pelletizer, which have the most important effect on the mechanical strength, and the moisture content of the mix which, influences of the pellet size (Harikrishnan & Ramamurthy, 2006).

Generally, increasing the inclination angle and the rotation speed of the pelletizer, or increasing
the rotation speed and the duration length of the process improves the ten percent fine values
due to the reduction of the pellets porosity (Manikandan & Ramamurthy, 2009).

323 However, the operational parameters have different effects on the pelletization efficiency depending on the pellets size (Table 2). Tests on cold-bonding pelletization of FA with high 324 swelling bentonite indicate that increasing the rotation speed, the inclination angle, the process 325 326 duration, the moisture content and the bentonite percentage results in beneficial effects for granules in the size range of 20-12.5 mm and 12.5-10 mm (Manikandan & Ramamurthy, 2009). 327 For pellets size ranging from 4.75 to 10 mm, the best process efficiency is obtained by 328 decreasing the rotation speed and the pelletizer inclination angle, or decreasing the process 329 duration length and increasing the bentonite content. 330

The effect of the rotation speed and the inclination angle seems to be less on final pellets characteristics when mixes contain a high percentage of cement (Tajra et al., 2018). Similarly, a reduced effect of these two parameters compared to the others (i.e. moisture and binder contents, and process duration) is observed for pellets with fineness of 257 m<sup>2</sup> kg<sup>-1</sup> and in the case of use of bentonite or kaolinite as binders (Manikandan & Ramamurthy, 2007). This effect becomes more important, for pellets with fineness of 414 m<sup>2</sup> kg<sup>-1</sup>, prepared without binder addition. Moreover, a good pelletization efficiency can be observed up to certain rotation speed limit because higher values lead to the prevalence of centrifugal force on gravitational ones with consequent particles adhesion on pelletizer sides and lowered process efficiency (Colangelo & Cioffi, 2013).

The typology of the aggregate and the curing method are essential in determining final pellets properties. Accelerated curing methods such as autoclaving or steam curing do not lead to production of pellets with improved properties compared to those obtained following a normal water cured method (Manikandan & Ramamurthy, 2008). Fig. 2 provides an example of qualitative variation trends for the aggregates mechanical performance at different values of the operating parameters.

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# 348 5. Application of the cold-bonded pellets

The main practical applications of LWAs are reviewed in the following subsections. The feasible LWAs reuse for manufacturing of products characterized by suitable properties is strongly dependent on several operating conditions. Table 3 summarizes the main LWAs applications, operating conditions and their consequent effects on the product properties. Moreover, illustrative representation of qualitative variation trends for mechanical performance of concretes made by artificial aggregates is reported in Fig. 2 as a function of the operating parameter values increase.

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#### **5.1 Cold-bonded pellets as aggregates for lightweight concrete**

Cold-bonded pellets are frequently employed as aggregates for the production of lightweight concrete (LWC). This application provides the possibility of massive waste reuse in concrete production, especially when LWC is prepared using FA both as a cementitious material and as cold-bonded aggregate (Rivera et al., 2015).

The partial replacement of natural aggregates by cold-bonded LWA in LWC results in a 362 concrete characterized by suitable properties for classical applications of the final product. For 363 364 instance, the use of FA pellets as LWC aggregates gives rise to good results in terms of compressive and split-tensile strength, static elastic modulus and shrinkage behavior, although 365 high volume ratios of added pellets cause an overall worsening of these properties (Gesoglu, 366 367 Özturan, & Güneyisi, 2004). Indeed, above certain values of the cement content, the volume fraction aggregate increase can lead to reduced concrete strength, due to the change of 368 predominant failure mode to aggregate fracture (G. Joseph & Ramamurthy, 2009b). The best 369 370 results in terms of concrete compressive strength enhancement are usually observed for LWC using cold-bonded FA pellets to replace the sand, (G. Joseph & Ramamurthy, 2009a). However, 371 372 despite the observable LWC properties enhancement achieved with high sand substitution rates, literature results indicate that normal weight concrete presents better characteristics compared 373 to light weight one containing FA pelletized aggregates (Kockal & Ozturan, 2011b; Patel, Patil, 374 375 Patil, & Vesmawala, 2018).

Negative effects on the compressive strength value of the final product can be obtained by increasing the FA pellets replacement level in LWC that are subjected to water or steam curing (Gesoğlu, Güneyisi, Ali, & Mermerdaş, 2013). Steam curing allows a total replacement of natural aggregates with FA cold-bonded pellets and the possibility of using the final product as structural concrete within 24 h. Steam curing is generally preferred to hot water curing to enhance LWC properties. Such a result can be observed at various replacement level (40, 50 and 62%) of cold-bonded aggregates made by FA+bentonite mixture after 3, 7, and 28 d of curing (Gomathi & Sivakumar, 2015). Similarly, LWC specimens subjected to oven curing at 200 °C present better mechanical properties than specimens subjected to water curing (Vijay & Singh, 2014). Nonetheless, experiments testing various curing methods (i.e. mist-cured, aircured, and sealed) on LWC made by FA aggregates do not indicate significant differences in terms of compressive strength and degree of hydration. This latter result can be ascribed to the autogenous curing in the cold-bonded aggregates (G. Joseph & Ramamurthy, 2011).

389 LWC properties strongly depend on the characteristics of the adopted cold-bonded pellets. For example, comparing LWC concrete made by FA and LWC concrete made by quarry dust 390 aggregates, it is showed a higher compressive strength of the former, and better values of the 391 392 compacting factor and porosity for the latter (Harilal & Thomas, 2013). According to the study of Thomas & Harilal (2015) a higher compressive strength can be obtained for LWC made by 393 quarry dust+FA aggregates, increasing the amount of FA. However, heating tests aimed at 394 395 determining the LWC residual compressive strength, indicate the suitable applicability of quarry dust alone for concrete manufacture. In fact, satisfactory results on the compressive 396 397 strength are reported up to 400 °C for the quarry dust-LWC, compared to the maximum value of 300 °C obtained in case of FA-LWC (N. Joseph, Harilal, Paul, & Thomas, 2013; Thomas & 398 Harilal, 2016). A further example is provided by the study of Gesoğlu, Özturan, & Güneyisi 399 (2006) examining the effect of four different aggregates addition (45% of concrete volume) on 400 LWC. This study shows that the use of 20% cement and 80% FA was the best solution in terms 401 of mechanical strength of the final product. 402

A possible enhancement of LWC properties can be obtained using LWAs containing
mechanically activated FA, which result in an improvement of compressive strength and a
substantial reduction in porosity and shrinkage of the concrete (Terzić et al., 2015). Another
possibility is the use of silica fume and/or steel fibers in the concrete preparation (Güneyisi,
Gesoğlu, & Ipek, 2013; Güneyisi, Gesoğlu, Pürsünlü, & Mermerdaş, 2013). Good results are

obtained preparing FA-LWC with the addition of silica fume at 10%, steel fibers at 0.75%, and 408 409 using a water to binder ratio of 0.35 (Gesoğlu, Güneyisi, Alzeebaree, & Mermerdas, 2013). In general, the addition of steel fibers improves the splitting tensile and flexural strength as well 410 as fracture energy and characteristic length (Güneyisi, Gesoglu, Özturan, & Ipek, 2015). Also, 411 the involvement of FA geopolymer aggregates in concrete production can lead to improved 412 mechanical properties compared to concrete made by addition of commercial lightweight 413 expanded clay aggregates (LECA) (Yliniemi et al., 2017). Referring to concrete shrinkage, 414 weight loss and age of cracking, positive effects are observed, by adding admixtures able to 415 reduce the shrinkage effect (from 0.75% to 3% by cement weight) in the LWC preparation 416 417 (Güneyisi et al., 2014). Good mechanical properties of the LWC are also obtained employing stoker furnace FA+cement pellets (70%+30%, respectively) which had a second-pelletization 418 419 step, adding, as binder a cement:coal FA ratio equal to 1:1 (Colangelo et al., 2015).

420 It has to be finally observed that, although LWC produced using cold-bonded FA aggregates has reduced mechanical properties compared to the equivalent LWC produced using sintered 421 422 FA aggregates (Kockal & Ozturan, 2010, 2011a), some exceptions exist. Chang & Shieh (1996) report that LWC made by cold-bonded FA aggregates has a higher compressive strength (33.9 423 MPa) compared to LWC made with sintered LWA (30.1 MPa) after 28 days curing. Frankovič 424 et al. (2017) indicate that granulated FA aggregate addition to LWC results in lower 425 compressive and tensile strength compared to LWC made from crushed FA aggregates or 426 crushed natural limestone aggregates even after 90 days curing. 427

428

#### 429 **5.2** Cold-bonded pellets as aggregates for self-compacting concrete

Cold-bonded aggregates can also be used for the production of self-compacting concrete (SCC).
Experiments aimed at investigating the effect of different replacement level of natural coarse
aggregates with cold-bonded aggregates mainly prepared from GGBS, show the possibility of

producing a stable material, even when the replacement is complete (i.e. 100%) (Gesoĝlu, 433 Günevisi, Mahmood, öz, & Mermerdas, 2012). Generally, different replacement levels of 434 coarse and/or fine FA-LWA, produce a SCC characterized by suitable deformability, passing 435 ability, and segregation resistance (Gesoğlu, Güneyisi, Özturan, Öz, & Asaad, 2014b). 436 Moreover, increasing the FA-LWA replacement level can entail the decrease of SCC density 437 438 and plastic viscosity as well as a lower high-range-water-reducing-admixture amount required 439 for a specific concrete workability (Gesoglu, Güneyisi, Ozturan, Oz, & Asaad, 2015). Despite this, the increasing replacement level of FA-LWA instead of natural aggregate in SCC can 440 cause lower splitting and compressive strength values due to the weakness of cold-bonded 441 aggregates (Gesoğlu, Güneyisi, Özturan, Öz, & Asaad, 2014a). Compared to control tests (only 442 Portland cement as binder), SCC made by FA-LWA as aggregates results in a decreased slump 443 flow and V-funnel flow times and increased L-box height ration. This latter result is achievable 444 445 with combined use of FA and silica fumes in ternary blends with Portland cement (Güneyisi, Gesoğlu, & Booya, 2012). The use of ternary blends with Portland cement, FA and silica fumes 446 447 for SCC preparation can lead to an improvement in the compressive strength achieving comparable values of binary blends with Portland cement and FA or only Portland cement 448 (Güneyisi, Gesoğlu, Booya, & Mermerdaş, 2015). The concurring use of SiO<sub>2</sub> nanoparticles 449 and cold-bonded FA-LWA treated by soluble sodium silicate solution appears to be very 450 interesting for SCC production (Güneyisi, Gesoglu, Azez, & Öz, 2015, 2016). Improved 451 concrete performance and reduced drying shrinkage compared to the untreated FA aggregates 452 produced with an optimal blend made from 5% of SiO<sub>2</sub> nanoparticles and treated FA 453 aggregates. Nonetheless, the use of these components may cause a significant decrease in the 454 SCC permeability, as reported by Güneyisi, Gesoglu, Azez, et al. (2015). 455

Further treatment of cold-bonded aggregates can be effective to produce a material suitable for the production of SCC. For instance, Hwang & Tran (2015) propose the use of  $H_2O_2$  as foaming agent for the production of LWA aggregate made by FA+GGBS. According to their results, the
adoption of foamed LWA aggregates can be useful to obtain SCC characterized by suitable
properties.

FA cold-bonded aggregates have good characteristic as self-curing agent. In fact, the water 461 absorbed by the aggregates during their pelletization can be gradually released to the cement 462 463 particles in the concrete allowing further hydration process and concrete characteristics improvement. Coupled with LECA, FA-LWA leads to SCC production with very high 464 compressive strength, split tensile strength and flexural strength under self-curing conditions. 465 According to the study, the reported results were achieved with replacement levels of 5% of 466 467 LECA and 30% of FA-LWA (Gopi, Revathi, & Kanagaraj, 2015). SCC characterized by good mechanical properties can be also obtained under water-curing conditions, with replacement 468 levels of 5% of LECA and 40% of FA-LWA (Gopi et al., 2015). Tests investigating the 469 470 characteristics of SCCs made by combining waste LWA (MSWI BA, PSA, coal FA, and washing aggregate sludge) with or without addition of materials for aggregates property 471 472 enhancement (i.e. nano-silica and polypropylene fiber) (Tang & Brouwers, 2018) showed a decrease of SCC flexural and compressive strength for all the concrete mixtures compared to 473 the control conditions. This result is more noticeable for the concrete obtained using the lowest 474 475 replacement level (30%). It was observed that the slump flow diameter decreased only for SCC made by combined waste LWA+polypropylene fiber, and an overall V-funnel time increased 476 compared to the control condition for all the tested concretes having the highest aggregate 477 replacement level (60%) (Tang & Brouwers, 2018). 478

479

#### 480 **5.3 Cold-bonded pellets for less investigated applications**

481 The use of pelletized aggregates for self-compacting mortars (SCM) is not frequently reported482 in the scientific literature. Nonetheless, the use of FA-LWA as replacement material in SCM

shows promising applicability. The increase of the FA-LWA replacement level leads to the 483 484 improvement of SCM properties such as workability and flowability, although it negatively affects the SCMs strength and porosity (Güneyisi, Gesolu, Altan, & Öz, 2015). Other effects 485 of increasing FA-LWA replacement level values on SCM, are reported by Güneyisi, Gesoglu, 486 Ghanim, Ipek, & Taha (2016). These authors indicate an increase of the SCM V-funnel flow 487 time, associated to a decrease of the slump flow diameter, with increasing percentage of added 488 489 FA-LWA. Both effects are significantly affected by the water-to-binder ratio (w/b) with the lowest variations observed for the highest tested ratio (0.44). At the same time, the highest SMC 490 compressive strength is obtained increasing the FA-LWA replacement level, while keeping the 491 492 lowest w/b ratio (0.33) (Güneyisi, Gesoglu, Ghanim, et al., 2016).

493 Negative effects on mortars compressive strength are observed on increasing the replacement 494 level of cold-bonded bottom-ash LWA. Obtained values are lower compared to the compressive 495 strength of mortars produced with raw bottom-ash or expanded shale aggregates, using the same 496 replacement percentage (Kim, Ha, & Lee, 2016). At the same time, the use of cold-bonded 497 bottom-ash aggregates allows higher reductions of the mortar autogenous-shrinkage, due to the 498 higher water absorption of the cold-bonded aggregate samples.

Few studies have investigated the applicability of cold-bonded pellets as adsorbent, for metal 499 500 removal from aqueous solutions. Tests on arsenic ions sorption using cold-bonded FA indicate that the process can achieve good removal efficiencies (Polowczyk et al., 2007). Process 501 kinetics follow a Lagergren pseudo-first-order model which has a similar form as the Boyd film 502 diffusion model. According to Polowczyk et al. (2007), this latter aspect makes it unclear if the 503 process is controlled by a pseudo-first order reaction or diffusion. Suitable adsorption capacity 504 of cold-bonded FA is reported also for copper and cadmium (Papandreou, Stournaras, & Panias, 505 506 2007). The process appears to be reversible in acidic condition (Papandreou et al., 2007), strengthening the significance of the potential application of cold-bonded pellets to replace 507

natural aggregates in cement matrixes. This clearly highlights the opportunity to reducecontaminants leaching phenomena, and ensure environmental sustainability.

510

# 511 6. Conclusions

The literature overview reported in the present work highlights the significant applicability of 512 the cold-bonding process as a sustainable solution for the disposal of several waste types. In 513 514 this perspective, the cold-bonding process displays i) interesting waste treatment results in the production of stabilized aggregates; and ii) wide applicability of the produced cold-bonded 515 516 aggregates in the construction sectors. Therefore, the process answers to the modern world 517 requirements of more environmentally sustainable practices for waste disposal, and can be fully ascribed in the framework of new technologies inspired by the principle of the circular 518 519 economy. The environmental-friendly and cost-saving characteristics of the cold-bonded 520 process compared to sintering represent a further advantage which emphasizes the scientific and practical interest. However, according to the literature, the methodology aimed at producing 521 cold-bonded aggregates characterized by proper characteristics usable for concretes can be 522 affected by different factors. The latter are mainly represented by i) the different characteristics 523 of the waste used for aggregates production; ii) the component proportions adopted for the 524 aggregate mix-design; iii) the selection of the operating parameters; iv) the aggregates 525 526 replacement level in the concrete mixture; and v) the environmental factors affecting the 527 aggregate characteristics. Providing a wide overview of different experimental studies on the 528 cold-bonding process, the present work allows the main process advantages/disadvantages to be identified. The detailed information reported can be useful to further support practical 529 530 decision-making focused on the selection of suitable operating conditions for a high efficient cold-bonded process producing aggregates with usable properties. 531

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#### 538 Table 1. Effects of relevant mix-designs on the cold-bonding pellets characteristics.

Pellet mix-design	Main result	Reference
Waste material: ore mix (from 88 to 90%) Binder: clinker (used alone or in different ratio with granulated blast furnace slag), granulated blast furnace slag (used in different ration with clinker) Addition: gypsum (used at 3% in 1:1=granulated blast furnace slag:clinker binder at 12%)	<ul> <li>Higher crushing strength observed for steam cured pellets compared the normal and accelerated cured ones</li> <li>Higher porosity after 28 d of normal curing conditions</li> <li>Highest crushing strength after 28 d of normal curing for mixture with ore mix+clinker (88%:12%)</li> </ul>	Dutta et al. (1992)
Waste material: pyrrhotite tailings mixed with binder percentage values of 2, 4, 6, 8, and 10% Binder: cement+gypsum $\beta$ -hemihydrate (used in percentage variable from 100 to 0%)	<ul> <li>Increasing fracture load (N pellet<sup>-1</sup>) with increasing pellet diameter</li> <li>Highest fracture load (over the whole curing period ranging from 0 to 28 d) for pallets with 100% of cement and binder dosage of 10%</li> <li>Optimal binder mixture observed for pellets with binder cement+gypsum β-hemihydrate of 40%:60%</li> </ul>	Amaratunga (1995)
Waste material: gold mill tailings mixed with binder percentage values of 2, 4, 6, 8, and 10% Binder: cement+gypsum $\beta$ -hemihydrate (used in percentage variable from 100 to 0%)	<ul> <li>Increasing fracture load (N pellet<sup>-1</sup>) with increasing pellet diameter for all the curing period values (1, 3, 7,14, and 28 d)</li> <li>Pellet optimal conditions with binder dosage of 4% and binder cement+gypsum β-hemihydrate of 80%:20% for pellets diameters equal to 12.7 and 19 mm</li> <li>Fulfilling fracture load strength (237 N pellet<sup>-1</sup>) and lowest Portland cement consumption percentage (3.2%) after 3 d of curing with binder dosage of 4% and binder cement+gypsum β-hemihydrate of 80%:20% for pellets diameter equal to 19 mm</li> </ul>	Amaratunga & Hmidi (1997)
Waste material: blue dust (90-96%) Binder: cement (4-10%) Addition: water (5-9% on dry raw mixture basis) and partial replacement of binders with rice husk ash silica or quartz silica (0.4-0.8%) for some mixture	<ul> <li>Higher early crushing strength values (at 1 d) observed by partially replacing cement with silica fines</li> <li>Overall higher crushing strength values for accelerated cured pellets compared to normal cured ones</li> </ul>	Dutta et al. (1997)
Waste material: iron ore (from 95 to 85%) mixed with coal (from 5 to 15%) or activated char (10%) or petroleum coke (10%) Binder: oil slush (5%), sodium silicate (5%), bentonite (6%), molasses (9%), slake lime (7%), and dextrin (from 1 to 5%)	<ul> <li>Higher similar values of dry strength for mixture with binder mixtures+iron ore:coal (90%:10%)+dextrin (30 kg pellet<sup>-1</sup> for 4% of dextrin and 31 kg pellet<sup>-1</sup> for 5% of dextrin)</li> <li>Lower similar decrepitation values</li> <li>Highest reduced pellets strength observed for sample with 4% of dextrin as binder, granulometry of 40%, and internal reductant percentage of 10% (using non-coking coal as reductant)</li> </ul>	Agrawal et al. (2000)
Waste material: fly-ash (100 or 92%) Binder: cement (8%) or lime (8%)	<ul> <li>Highest crushing values for pellets whit cement at 7 and 14 curing days</li> <li>Highest crushing strength at 28 curing day for pellets whit lime</li> </ul>	Baykal & Döven (2000)

Waste material: electric arc furnace dust with specific surface area of $4.32\pm0.21$ m <sup>2</sup> g <sup>-1</sup> (82.7%) and $0.59\pm0.04$ m <sup>2</sup> g <sup>-1</sup> (80.9%) Binder: cement (used at 3 and 5%), coal (used at 19.1 and 17.3%) Addition: CaCO <sub>3</sub> (used at 12%)	<ul> <li>Cold compressive strength improvement with addition of 3 and 5 wt% of cement</li> <li>No decrepitation occurrence after test at 700 °C for dried pellets with 3 and 5 wt% of cement</li> <li>Zn removal enhancement (1124 °C) from pellets by adding 5 wt% of cement or 12 wt% of CaCO<sub>3</sub></li> </ul>	Mantovani & Takano (2000)
Waste material: coarse mill scale (40%), blast furnace sludge (20%), blast furnace dust (15%), basic oxygen furnace sludge (15%), basic oxygen furnace dust (5%), and oily mill scale (5%) Binder: blast furnace slag (6%) and clinker (4%) Addition: coke (from 5 to 20%) or graphite (from 4.1 to 16.5%)	<ul> <li>Best reduction degree with coke as reducing agent compared to the graphite</li> <li>Reduction degree increase with increasing C<sub>fix</sub>/ Fe<sub>total</sub> ratio</li> <li>Increase of the reduction period with decreasing temperatures</li> </ul>	Çamci et al. (2002)
Waste material: electric arc furnace dust with specific surface area of $3.62\pm0.41$ m <sup>2</sup> g <sup>-1</sup> (83.3%) and $4.32\pm0.21$ m <sup>2</sup> g <sup>-1</sup> (82.7%) Binder: cement (used at 5%), coal (used at 16.7 and 17.3%) Addition: CaCO <sub>3</sub> (used at 12%) or KCl (used at 12%)	<ul> <li>Significant compressive strength decrease after heating treatment for mixtures with electric arc furnace dust+coal (83.3%:16.7%) and electric arc furnace dust+coal+cement (83.3%:16.7%:5%) at pellets size of 14 mm</li> <li>No compressive strength decrease after heating treatment for mixture with electric arc furnace dust+coal+cement (83.3%:16.7%:5%) at pellets size of 7 mm</li> <li>Best Zn removal for mixture with electric arc furnace dust+coal+KCl (82.7%:17.3%:12%) at 1123.85 °C</li> <li>No significant differences with various additives at about 1199.85 °C or higher temperatures</li> </ul>	Mantovani et al. (2002)
Waste material: fly-ash (from 90 to 80%) Binder: cement (from 10 to 20%)	<ul> <li>Specific gravity increase (from 1.65 to 1.76) and water absorption decrease (from 34.4 to 20.8%) with increasing cement percentage from 10 to 20%</li> <li>Increase of bulk unit weight (from 857 to 972 kg m<sup>-3</sup>) and particle strength (from 6.01 to 8.57 MPa) with increasing cement percentage from 10 to 20%</li> </ul>	Chi et al. (2003)
Waste material: basic oxygen furnace coarse sludge (35%), basic oxygen furnace fine sludge (20%), blast furnace flu dust (20%), briquette fines (10%), and filter dust (5%) Binder: cement (10%)	<ul> <li>Fe<sub>2</sub>O<sub>3</sub> reduction to Fe<sub>3</sub>O<sub>4</sub> between 500 and 600 °C</li> <li>Fe<sub>3</sub>O<sub>4</sub> reduction to FeO between 640 and 850 °C</li> <li>FeO reduction to Fe between 850 and 1200 °C</li> <li>Carbonate decomposition occurrence at about 700 °C forming Ca<sub>2</sub>Fe<sub>2</sub>O<sub>5</sub> and other products</li> </ul>	Robinson (2005)
Waste material: iron ore (91.5%) Binder: alumina cement (7%), bentonite (0.6%)+0.9% silica fume+0.14 super plasticiser	<ul> <li>Average point strength of pellets with alumina cement equal to 2300 N</li> <li>Higher crushing strength values for pellets with alumina cement compared to pellets made by Portland cement</li> </ul>	Aota et al. (2006)

Addition: silica fume (0.9%), super plasticiser (0.14%)

Waste material: fly-ash (100% or 90-70%) Binder: cement (from 10 to 30%)	<ul> <li>Increasing shear strength with increasing cement percentage addition</li> <li>Specific gravity increase and water absorption decrease of the aggregates with increasing cement content</li> </ul>	Arslan & Baykal (2006)
Waste material: fly-ash type A, fly-ash type B, fly-ash type A with water glass surface treatment Binder: cement	<ul> <li>Highest crushing strength for pellets with B type fly-ash</li> <li>Highest water absorption values for type A fly-ash</li> <li>Lowest water absorption values for pellets with A type surface treated fly-ash</li> </ul>	Gesoğlu et al. (2007)
Waste material: fly-ash class F Binder: bentonite (from 4 to 14%) or kaolinite (from 4 to 30%)	<ul> <li>Pelletization efficiency increase from 48 to 98% with increasing content of bentonite</li> <li>Pelletization efficiency increase from 29 to 98% with increasing content of kaolinite</li> </ul>	Manikandan & Ramamurthy (2007)
Waste material: quarry fines Binder: biomass ash, cement bypass, kiln dusts, MSWI bottom ash and fly-ash, paper ashes, pulverized fuel ash, sewage sludge ash, and wood ash (used as CO2- reactive binders with addition from 10 to 50%)	• Similar properties for all the carbonated LWAs compared to commercially available LWAs	Gunning et al. (2009)
Waste material: fly-ash class F (fineness of 257 m <sup>2</sup> kg <sup>-1</sup> ) Binder: high swelling bentonite (from 6 to 14%) or medium swelling bentonite (from 9 to 21%)	<ul> <li>Pelletization efficiency equal to 98% with 14% of high swelling bentonite</li> <li>Pelletization efficiency equal to 97% with 21% of medium swelling bentonite</li> </ul>	Manikandan & Ramamurthy (2009)
Waste material: bottom ash Binder: cement or lime Addition: Ca(OH) <sub>2</sub> (as pelletization admixture) or Na <sub>2</sub> SO <sub>4</sub> (as chemical activator)	<ul> <li>Optimal mixture values (expressed as % by weight of BA) equal to 31.27% (moisture content), 1.59% (Ca(OH)<sub>2</sub>), 8.99% (binder), 3.19% (Na<sub>2</sub>SO<sub>4</sub>) form cement</li> <li>Optimal mixture values (expressed as % by weight of BA) equal to 31.19% (moisture content), 1.47% (Ca(OH)<sub>2</sub>), 8.99% (binder), 3.19% (Na<sub>2</sub>SO<sub>4</sub>) for lime</li> <li>Values equal to 92.8% of pelletization efficiency, 928.6 kg m<sup>-3</sup> of bulk density, 2.41 t of ten percentile fines, 35.1% of porosity, 19.1% of water absorption with optimal mixture of cement</li> <li>Values equal to 90.7% of pelletization efficiency, 920.8 kg m<sup>-3</sup> of bulk density, 2.18 t of ten percentile fines, 36.8% of porosity, 19.3% of water absorption with optimal mixture of lime</li> </ul>	Geetha & Ramamurthy (2010a)
Waste material: bottom ash Binder: cement or lime (as cementitious binders from 0 to 14%) and kaolin, metakaolin, clay with plasticity index of 78 and 108, medium swelling bentonite, or high swelling bentonite (as clay binders from 0 to 30%) Addition: Ca(OH) <sub>2</sub>	<ul> <li>Maximum pelletization efficiency (97-98%) without Ca(OH)<sub>2</sub> addition with high swelling bentonite (14%), medium swelling bentonite (25%), other clay binders (30%), cement and lime (14%)</li> <li>Pelletization efficiency improvement 97-98% with Ca(OH)<sub>2</sub>, 10 % with high swelling bentonite, cement and lime, 18% with medium swelling bentonite, 20% with kaolin, metakaolin and clay with plasticity index of 78, 25% with clay with plasticity index of 108</li> </ul>	Geetha & Ramamurthy (2010b)

Waste material: iron ore (from 58.70 to 69.28%), coal from mines (32.49 to23.12%) and from steel plant (from 28.99 to 20.33%) Binder: 10% of slaked lime and 5% of dextrose on iron ore weight basis (used from 8.81 to 10.39%)	<ul> <li>Highest compressive strength equal to 330±10.31 N pellet<sup>-1</sup> for mixture with iron ore+mines coal+binder (66.85%:23.12%:10.03%) and 362±11.72 N pellet<sup>-1</sup> for mixture with iron ore+steel plant coal+binder (69.28%:20.33%:10.39%)</li> <li>Minimum shatter index equal to 0.336% for mixture with iron ore+mines coal+binder and 0.245% for mixture with iron ore+steel plant coal+binder</li> </ul>	Sah & Dutta (2010)
Waste material: fly-ash (added in binary and ternary mixture from 90 to 30%) Binder: cement (added in binary and ternary mixture from 10 to 30%) Addition: granulated blast furnace slag (added in ternary mixture from 15 to 45%) or rice husk ash (added in ternary mixture from 15 to 45%)	<ul> <li>Better results in terms of aggregate compressive strength with addition of granulated blast furnace slag compared to rice husk ash addition</li> <li>Water absorption increase and aggregate unit weight decrease with increasing addition of rice husk ash</li> <li>Opposite tendency by increasing the addition of granulated blast furnace slag</li> </ul>	Bui, Hwang, Chen, & Hsieh (2012)
Waste material: fly-ash (alone or added in binary and ternary mixture from 75 to 50%) Alkaline activator: 10M NaOH+sodium silicate Addition: granulated blast furnace slag (added in binary or ternary mixture from 13 to 50%) and rice husk ash (added to binary or ternary mixture from 12 to 50%)	<ul> <li>Higher crushing strength for fly-ash+granulated blast furnace slag aggregates (about 15 MPa)</li> <li>Highest absorption capacity (20.5%) for fly-ash+rice husk ash (50%:50%) aggregates</li> <li>Lowest value absorption capacity (7.8%) for fly+granulated blast furnace slag (50%:50%) aggregates</li> </ul>	Bui, Hwang, Chen, Lin, et al. (2012)
Waste material: hematite (from 78.12 to 83.17%), coke (from 18.16 to 13.87%), and limestone (from 3.71 to 3.31%) Binder: molasses (from 20 to 50%)	<ul> <li>Higher compressive strength and lower porosity values with 50% of molasses for each Fetot/Cfix ratio</li> <li>More similar compressive strength and porosity values for 50% and 40% of molasses in mixture with hematite:coke:limestone=83.17%:13.87%:3.31% (Fetot/Cfix ratio=3.5)</li> </ul>	Cevik et al. (2013)
Waste material: weathered fly-ash (added in binary, ternary and quaternary mixture from 90 to 20%) Binder: lime (added in binary, ternary and quaternary mixture from 10 to 30%) Addition: wastewater treatment sludge (added in ternary and quaternary mixture from 50 to 60%) and desulfurization device sludge (added in ternary and quaternary mixture from 30 to 10%)	<ul> <li>Highest strength values for fly-ash+lime (60%:40%) and fly-ash+lime+desulfurization device sludge (40%:30%:30) aggregates</li> <li>Lowest strength values for fly-ash+lime+wastewater treatment sludge (30%:20%:50 and 24%:16%:60)</li> <li>Lower density values for ternary mixture with 30% desulfurization device sludge and both quaternary mixture aggregates</li> </ul>	Ferone et al. (2013)
Waste material: fly-ash class F (used alone or binary mixture at 80 or 70%) Binder: cement (20%), furnace slag (30%), metakaolin (30%) or bentonite (20%) Addition: NaOH in all the mixtures	<ul> <li>Highest crushing strength for mixture with fly-ash+furnace slag (70%:30%)) at all sizes (values from 22.81 to 12.80 MPa with decreasing pellets size)</li> <li>Highest efficiency (99.07%) for mixture with furnace slag aggregates</li> <li>Lowest efficiency (66.95%) for mix with on fly-ash</li> </ul>	Gomathi & Sivakumar (2014)

Waste material: bituminous pond ash or lignite pond ash

Binder: cement (for both wastes) or lime (for bituminous pond ash)

Addition: Na<sub>2</sub>SO<sub>4</sub> (for bituminous pond ash), Ca(OH)<sub>2</sub> (for both wastes), and hardening admixture (i.e. carbonic aluminate salt made by 80% of cement, 9% of Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, 6% of Na<sub>2</sub>CO<sub>3</sub>, and 5% of CaO) for lignite pond ash

> Maximum pelletization efficiency of lignite pond ash-cement mixture equal to 99% without Ca(OH)2 and 20% of binder

•

•

Bulk density and ten percent fines value increase with increasing dosage of • binder+Na<sub>2</sub>SO<sub>4</sub> and Ca(OH)<sub>2</sub> addition for bituminous pond ash

Maximum pelletization efficiency of bituminous pond ash-cement mixture equal

Maximum pelletization efficiency of bituminous pond ash-cement mixture equal

Maximum pelletization efficiency of bituminous pond ash-lime mixture equal to

Maximum pelletization efficiency of bituminous pond ash-lime mixture equal to

Maximum pelletization efficiency of lignite pond ash-cement mixture equal to 82

to 78% with Ca(OH)<sub>2</sub> and 20% of binder

79 with Ca(OH)2 and 20% of binder

with Ca(OH)2 and 20% of binder

to 98% without Ca(OH)2 and 20% of binder

96% without Ca(OH)2 and 20% of binder

- Water absorption and porosity decrease with increasing dosage of binder+ Na<sub>2</sub>SO<sub>4</sub> and Ca(OH)<sub>2</sub> addition for bituminous pond ash
- Bulk density and ten percent fines value increase with increasing dosage of hardening admixture and Ca(OH)<sub>2</sub> for lignite pond ash
- Water absorption and porosity decrease with increasing dosage of hardening admixture and Ca(OH)<sub>2</sub> for lignite pond ash
- Significantly high crushing strength values, low water absorption capacity and Colangelo et al. Los Angeles coefficient values for double-step pelletization mixtures with stoker (2015)furnace fly-ash+cement (70%:30%), stoker furnace fly-ash+lime+coal fly-ash (60%:15%:25%), and stoker furnace fly-ash+lime+coal fly-ash (50%:30%:20%) •
- Low heavy metals leaching for all the granules except for the granules produced with only lime as binder
- Higher crushing strength of sintered aggregates of 10 mm (20.62 MPa) and 12 Gomathi & mm diameter (18.34 MPa) compared to cold-bonded ones at 10 mm (10.22 MPa) Sivakumar and 12 mm (14.51 MPa) (2015)
- Overall decreasing crushing and particle crushing strengths with increasing percentage of hydrogen peroxide for mixtures with and without cement addition;
- Higher crushing and particle crushing strengths for aggregates with surface • treatment compared to aggregates without surface treatment
- Overall increasing water absorption with increasing percentage of H<sub>2</sub>O<sub>2</sub>

Waste material: rotary furnace fly-ash (used in binary and ternary mixture from 70 to 50%), stoker furnace fly-ash (used in binary and ternary mixture from 70 to 50%) Binder: cement (used in binary mixture at 30%), lime (used in binary and ternary

mixture at 30 and 15%), coal fly-ash (used in ternary mixture at 25 and 20%) Double-step pelletization: 1:1=cement:coal fly-ash binder mixture in amount equal to 40% of the granules weight

Waste material: fly-ash class F (80%) Binder: bentonite (20%) Addition: water (25% of binder weight) mixed with 10M NaOH

Waste material: fly-ash (used in binary and ternary mixture from 80 and 70%) Binder: cement (used in ternary mixture at 10%), ground blast furnace slag (used in binary and ternary mixture at 20%)

Addition: H<sub>2</sub>O<sub>2</sub> (used in binary and ternary mixture from 3.5 to 8.75%) and surface treatment with alkaline solution 2.5M SiO<sub>2</sub>/Na<sub>2</sub>O (used in binary and ternary mixture at 5-7% aggregates by weight)

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Hwang & Tran

(2015)

Vasugi Ramamurthy (2014)

&

Waste material: iron ore fines Binder: bentonite (from 0.4 to 0.8 wt %)	•	Increasing dry compressive strength of green pellets with increasing bentonite percentage and increasing values of Blaine fineness Increasing cold crushing strength values with increasing hardening temperatures and increasing Blaine fineness	Pal, Ghorai, Agarwal, et al. (2015)
Waste material: iron ore mixed with various percentage of carbon (from coal) Binder: molasses	• • •	Higher decrease of cold compressive strength by increasing coal percentage from 0 to 20 wt% in pellets with 10 wt% of calcined lime and no molasses compared to pellets with 10 wt% of calcined lime and 3 wt% of molasses Higher cold compressive strength for pellets with 3 wt% of molasses also with coal percentage value higher than 20 wt% Increasing abrasion loss by increasing coal percentage from 0 to 20 wt% in pellets with 10 wt% of calcined lime and no molasses Almost costant abrasion loss for pellets with 10 wt% of calcined lime and 3 wt% of molasses also with coal percentage higher than 20 wt% of calcined lime and no molasses also with 20 wt% of calcined lime and no molasses also with 20 wt% of calcined lime and 20 wt% of molasses also with coal percentage higher than 20 wt%	Pal, Ghorai, & Das (2015)
Waste material: siliceous ore tailings Binder: Ca(OH) <sub>2</sub> (from 6 to 14%)	•	Highest compressive strength observed within 15 min for 14% of $Ca(OH)_2$ Highest pellet porosity (25%) observed for 12% of $Ca(OH)_2$	McDonald et al. (2016)
Waste material: iron ore fines Binder: carboxymethyl-cellulose or calcium-lignosulfonate (used at 1 or 2%), dextrin or dextrin+bentonite (used at 3 or 4%) Addition: coke (used at 15 and 20%)	•	Overall dry strength increase with increasing binder percentage (for all the binders) and decreasing coke percentage Highest dry strength value by adding 4% of dextrin+bentonite Optimal binder represented by the carboxymethyl-cellulose addition with dry strengths in excess of 300 N pellet <sup>-1</sup> , metallization value up to 95.5% (pellets reduction at 1100 °C for 20 min), and decrepitation indices lower than 0.1%	Nikai & Garbers- Craig (2016)
Waste material: peat wood fly-ash (used alone or in binary mixture at 80 and 60%) Binder: blast furnace slag (used in binary mixture at 20 and 40%), coal fly-ash (used in binary mixture at 20 and 40%), and metakaolin (used in binary mixture at 20 and 40%) Addition: H <sub>2</sub> O or K <sub>2</sub> O <sub>3</sub> Si as liquid binders	•	Strongest granules with alkali activators and blast furnace slag Lowest strength for the mixture with peat wood fly-ash+H <sub>2</sub> O	Yliniemi, Nugteren, et al. (2016)
Waste material: fly-ash from electrostatic precipitator and silos of fluidized bed combustion treatment Addition: sodium silicate as alkali activator	•	Best aggregate strength with highest amount of selectively soluble $SiO_2$ and $Al_2O_3$ Leaching increase after alkali activation especially for anionic heavy metal species	Yliniemi, Pesonen, et al. (2016)
Waste material: fuel-biofuel fly-ash (used alone or in binary mixture at 80 and 60%) Binder: blast furnace slag (used in binary mixture at 20 and 40%) and metakaolin	•	Highest crushing strength for mixture with fly-ash+blast furnace slag (60%:40%) for samples cured in plastic bags for 28 d or immersed in water for 3 d	Yliniemi, Tiainen, et al. (2016)

(used in binary mixture at 20 and 40%)

Waste material: car fluff (coal fly-ash:car fluff ratio equal to 0.83) Binder: cement+coal fly-ash (water to binder ratio equal to 0.41, 0.36, and 0.31)

Waste material: fly-ash (used in multiple additions mixture at 100 and 50% on  $\bullet$  total fly-ash+paper sludge ash)

Binder: cement (used in multiple additions mixture from 5 to 15% on total solids) Addition: bottom ash fines (used in multiple additions mixture from 10 to 75% on total solids), paper sludge ash (used multiple additions mixture at 50% on total flyash+paper sludge ash), washing aggregate sludge (used in multiple additions mixture at 5% on total solids), granulated blast furnace slag (used in multiple additions mixture at 67% on cement), and polypropylene fibre (used in multiple additions mixture from 1 to 4.5% on volume)

Waste material: bottom ash fines (from 20 to 80%), washing aggregate sludge (5%), coal fly-ash (from 5 to 65%), and paper sludge ash (from 5 to 65%) Binder: cement (10%)

Waste material: fluidized bed combustion fly-ash, mine tailing Addition: sodium silicate to form geopolymer aggregates

• Highest crushing strength (5.2±0.4 MPa) for lower water to binder ratio

Lowest water absorption  $(10.3\pm0.7\%)$  for lower water to binder ratio

- Heavy metals immobilization degree higher than 90% for all the samples
  - Best crushing strength values (after 28 d curing) for mixture with cement+flyash+paper sludge ash+bottom ash fines+washing aggregate sludge 1 (15%:50%:50%:40-75%:5% of the corresponding weight basis) and mixture with cement+fly-ash+paper sludge ash+bottom ash fines+washing aggregate sludge+polypropylene fibre (10%:50%:50%:40-75%:5%:2.5% of the corresponding weight and volume basis)
- Lowest crushing strength values for mixture with cement+granulated blast furnace slag+fly-ash+paper sludge ash+bottom ash fines+washing aggregate sludge (6%:67%:50%:50%:40-75%:5% of the corresponding weight basis) and mixture with cement+fly-ash+bottoma ash fines+washing aggregate sludge (10%:100%:40-75%:5% of the corresponding weight basis)
- Leaching of Cu, Mo, and chloride influenced by the amount of bottom ash fines
- Mo leaching influenced by the amount of fly-ash
- Chloride leaching influenced by the amount of paper sludge ash
  - High crushing resistance of the pellets for bottom ash fines addition Tang et al. (2017)
- Lower cement requirement for pellets production with comparable crushing strength by using bottom ash fines compared only powders
- Sulphate and antimony leaching decrease from pellets
- Copper leaching increase from pellets
- Higher crushing force required for the fly-ash geopolymer aggregates compared Yliniemi et al. (2017)
- Lower crushing force required for the mine tailing geopolymer aggregates compared to lightweight expanded aggregates

Tang & Brouwers (2017)

Colangelo et al.

(2017)

Waste material: fluxed hematite Binder: modified humic acid or bentonite (in various percentages)	<ul> <li>Wet and dry compression strength increase with increasing dose of modified humic acid</li> <li>Overall compression strength increase of pellets with modified humic acid at increasing time and temperature of pre-heating and roasting treatments</li> <li>Lower compression strength for pre-heated (980 °C, 12 min) and roasted (1250 °C, 10 min) pellets with modified humic (0.6 wt%) compared to pellets with bentonite (0.66 wt%)</li> <li>Higher mass loss rate of roasted pellets with modified humic acid compared to roasted pellets with bentonite at 980 °C</li> <li>Lower mass loss rate of roasted pellets with modified humic acid compared to roasted pellets with bentonite at 1100 °C</li> <li>Comparable mass loss rate values for roasted pellets with modified humic acid compared to roasted pellets with bentonite at 1250 °C</li> </ul>	Zhou et al. (2017)
Waste material: fly-ash class C (used alone or in binary mixture from 95 to 85%) Binder: cement (from 5 to 15%) and Ca(OH) <sub>2</sub> (at 5 or 10%)	<ul> <li>Highest water absorption values of 16.9% for mixture with cement+fly-ash (15%:85%) and 18.46% for mixture with Ca(OH)<sub>2</sub>+fly-ash (10%:90%)</li> <li>Increasing crushing strength up to 10% of cement addition</li> <li>Lowest crushing strength value with 15% of cement (299 N)</li> <li>Crushing strength equal to 470.9 N mixture with Ca(OH)<sub>2</sub>+fly-ash (5%:95%) and 285.8 N for mixture with Ca(OH)<sub>2</sub>+fly-ash (10%:90%)</li> </ul>	Narattha & Chaipanich (2018)
Waste material: fly-ash (from 95 to 80%) Binder: cement (from 5 to 20%) Addition: various replacement level of FA with expandend perlite particles	<ul> <li>Highest aggregates specific strength factor with 20% of cement at 30 rpm and 35°</li> <li>Aggregate properties improvement with a replacement level of expanded perlite particles up to 40%</li> </ul>	Tajra et al. (2018)
Waste material: fly-ash or slag Mixtures: cement+sand, cement+fly-ash+sand, and cement+slag+sand Addition: varying percentage of phase change material for each mixture group (0, 6, 12.5, 25, and 50% for cement+sand mixture while 0, 6, and 12.5% for mixtures with fly-ash or slag)	<ul> <li>Increasing water absorption and permeable porosity with increasing phase change material percentage addition for all the investigated mixtures</li> <li>Higher crush strength for fly-ash mixture compared to slag mixture with 0, 6, and 12% of phase change material addition</li> <li>Overall higher crush strength for cement+sand mixtures with comparable phase change material percentages</li> </ul>	Tuncel & Pekmezci (2018)

Operational parameter	Main result	Reference	
	<ul> <li>Main significant influence on the pelletization efficiency of 414 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash</li> <li>Reduced influence on pelleteziation efficiency of 257 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash</li> </ul>	Manikandan & Ramamurthy (2007)	
Inclination angle	<ul> <li>Increasing pelletization efficiency with increasing inclination angle for pellet sizes of 20-12.5 mm and 12.5-10 mm</li> <li>Increasing pelletization efficiency with decreasing inclination angle for pellet sizes of 10-4.75 mm</li> </ul>	Manikandan & Ramamurthy (2009)	
-	• More negligible effect of inclination angle compared to rotation speed and binder content on the pelletization efficiency	Tajra et al. (2018)	
	<ul> <li>Main significant influence on the pelletization efficiency of 414 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash</li> <li>Reduced influence on pelleteziation efficiency of 257 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash</li> <li>Higher effect of rotation speed compared to the inclination angle</li> </ul>	Manikandan & Ramamurthy (2007)	
Rotation speed	<ul> <li>Increasing pelletization efficiency with increasing rotation speed for pellet sizes of 20-12.5 mm</li> <li>Reduced influence on pelletization efficiency for pellets size of 12.5-10 mm</li> <li>Increasing pelletization efficiency with decreasing rotation speed for pellet sizes of 10-4.75 mm</li> </ul>	Manikandan & Ramamurthy (2009)	
	<ul> <li>Pellets strength increase with increasing rotation speed up to a certain value (45 rpm)</li> <li>Lower efficiency for values higher than 45 rpm due to centrifugal force prevalence on gravitational one</li> </ul>	Colangelo & Cioffi (2013)	
	<ul> <li>Higher effect on pellets crushing strength and pelletization efficiency compared to the inclination angle</li> <li>Lower effect on pellets crushing strength and pelletization efficiency compared to the binder content</li> </ul>	Tajra et al. (2018)	
Process duration	<ul> <li>Negligible effect on pelletization efficiency of 414 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash</li> <li>More significant effect for 257 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash with both bentonite or kaolinite content compared to moisture content</li> </ul>	Manikandan & Ramamurthy (2007)	
	• Lower influence on pelletization efficiency for pellet sizes of 20-12.5 mm and 12.5-10 mm than 10-4.75 mm	Manikandan & Ramamurthy (2009)	
	• Lower effect compared other parameters	Manikandan & Ramamurthy (2007)	
Binder content	• Higher effect on pelletization efficiency for pellet sizes of 10-4.75 mm than 20-12.5 mm and 12.5-10 mm	Manikandan & Ramamurthy (2009)	
Diffeet content	• Most influencing parameter on pellets crushing strength and pelletization efficiency compared to the inclination angle and rotation speed	Tajra et al. (2018)	

#### 544 Table 2. Effects of different operational parameters on cold-bonding pelletization efficiency.

	Moisture content	<ul> <li>Negligible effect on pelletization efficiency of 414 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash</li> <li>Higher effect for 257 m<sup>2</sup> kg<sup>-1</sup> fineness fly-ash with bentonite or kaolinite content</li> </ul>	Manikandan & Ramamurthy (2007)
		Increasing pelletization efficiency with increasing moisture content	Manikandan & Ramamurthy (2009)
	Curing method	<ul> <li>Improvement of ten percent fines value and reduced water absorption through normal water curing</li> <li>No significant pellet properties improvement with autoclaving and steam curing compared to the normal water curing</li> </ul>	Manikandan & Ramamurthy (2008)
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546			

#### 548 Table 3. Main applications of cold-bonded pellets and related affecting operating conditions.

Main application	Operating condition	Main result	Reference
	Aggregate replacement level	<ul> <li>Suitable lightweight concrete characteristics for replacement level ranging between 30-60%</li> <li>Overall properties worsening for high values of the replacement level</li> </ul>	Gesoglu et al. (2004)
	Curing method	• Lightweight concrete properties improvement through hot water and steam curing	Gomathi & Sivakumar (2015)
Lightweight concrete		• Bond strength enhancement (65 to 78%) with steel fiber addition (volume ratio ranging from 0.35 to 1.50%)	Güneyisi, Gesoğlu, & Ipek (2013)
	Mix-design	• Compressive strength enhancement with silica fume addition (10%)	Güneyisi, Gesoğlu, Pürsünlü, et al. (2013)
		• Compressive strength enhancement (15-60%) with double-step pelletization for various lightweight aggregates mixtures	Colangelo et al. (2015)
Self-compacting	A garegate	• Suitable self-compacting concrete properties for GBBS-LWA replacement level up to 100%	Gesoĝlu et al. (2012)
concrete	replacement level	• Self-compacting concrete worsening with increasing FA-LWA replacement level from 10 to 50%	Gesoglu et al. (2015)

Adsorbent material	Operating parameters	<ul> <li>Adsorption increase with increasing solution pH (100% Cu and Cd adsorption for pH above 9)</li> <li>Adsorption decrease with stirring velocity decrease from 140 to 60 rpm (Cu adsorption decrease from 55 to less than 10%)</li> <li>Adsorption decrease with stirring velocity significant increase (above 140 rpm)</li> <li>Further heavy metals stabilization with adsorbent LWA in concrete at replacement level of 50 and 75%</li> <li>Decreasing concrete compressive strength with replacement level increase</li> </ul>	Papandreou et al. (2007)
Self-compacting mortar	Aggregate replacement level	<ul> <li>Self-compacting mortar compressive strength worsening with increasing FA-LWA replacement level (25, 50, 75, and 100%)</li> <li>More noticeable replacement level negative effect on self-compacting mortar compressive strength for increasing water to binder ratio (0.33, 0.37, 0.40, and 0.44)</li> <li>No self-compacting mortar compressive strength decrease with raw bottom ash and expanded shale aggregates replacement level of 25 and 50%</li> <li>Self-compacting mortar compressive strength decrease by increasing bottom ash-LWA replacement level (at 25 and 50%)</li> </ul>	Güneyisi, Gesoglu, Ghanim, et al. (2016) Kim et al. (2016)
	Mix-design	<ul> <li>Suitable self-compacting concrete properties for 15% LWA replacement level and 5-10% silica fume addition</li> <li>Optimal H<sub>2</sub>O<sub>2</sub> percentage at 7% with resulting LWA suitable for replacement in self-compacting concrete</li> <li>Suitable self-compacting concrete properties with 40% FA-LWA and 5% LECA replacement levels under water-curing</li> <li>Compressive and flexural strengths decrease with 30 and 60% LWA replacement level also for LWA with 2.5% of polypropylene fiber or 0.8% of nano-silica</li> </ul>	Güneyisi et al. (2012) Hwang & Tran (2015) Gopi et al. (2015) Tang & Brouwers (2018)



553 Fig. 1. Schematic representation of the pelletizer a) front section and b) side section; Legend: rotating plate (1), rotation speed controller (2), scrapers (3), nebulizer (4), pump (5), 554 water tank (6), endless screw (7).



557 Fig. 2. Exemplifying graphical representation of qualitative variation trends for aggregate (dots and lines) and concrete (histograms) mechanical performance at different values of 558 the operating parameters. Parameter values increase from left to right side and mechanical performance improvement from down to up side.

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