## POLITECNICO DI TORINO Repository ISTITUZIONALE

Recycled coarse aggregates from pelletized unused concrete for a more sustainable concrete production

Original

Recycled coarse aggregates from pelletized unused concrete for a more sustainable concrete production / Bassani, M.; Diaz Garcia, J. C.; Flavia, Meloni; Volpatti, G.; Zampini, D.. - In: JOURNAL OF CLEANER PRODUCTION. - ISSN 1879-1786. - STAMPA. - 219:(2019), pp. 424-432. [10.1016/j.jclepro.2019.01.338]

Availability: This version is available at: 11583/2725472 since: 2019-02-16T16:43:57Z

Publisher: Elsevier

Published DOI:10.1016/j.jclepro.2019.01.338

Terms of use:

This article is made available under terms and conditions as specified in the corresponding bibliographic description in the repository

Publisher copyright Elsevier preprint/submitted version

Preprint (submitted version) of an article published in JOURNAL OF CLEANER PRODUCTION © 2019, http://doi.org/10.1016/j.jclepro.2019.01.338

(Article begins on next page)

# RECYCLED COARSE AGGREGATES FROM PELLETIZED UNUSED CONCRETE FOR A MORE SUSTAINABLE CONCRETE PRODUCTION

#### ABSTRACT

A significant amount of the concrete produced worldwide is returned unused to production plants and has the potential to cause serious damage to natural soils and waters.

This study proposes the use of a new pelletization process to convert fresh unused concrete into an artificial aggregate, which can be reintroduced into the concrete production cycle. Specifically, the investigation focuses on the properties of two pelletized recycled aggregates (PRAs) derived from two sources of an unsettled cementitious mixture. This research is based on the hypothesis that properties of the returned concrete may affect the characteristics of PRAs.

PRAs were evaluated by particle size distribution, particle density, water absorption, resistance to fragmentation, and chemical and mineralogical analysis. Test results confirm that the concrete influences some properties of PRAs, and that PRAs exhibit different properties to those of conventional aggregate. However, it can be reused in place of natural coarse aggregate in the production of new concrete.

#### **KEYWORDS:**

Unsettled concrete, recycled concrete, pelletization process, recycled aggregate, sustainable concrete production.

#### **1. INTRODUCTION**

Aggregate is one of the basic components of concrete, occupying about 70-80% of its volume (De Brito and Saikia, 2013). It constitutes the structural skeleton of cementitious composites and its properties influence the behaviour of fresh and hardened mixtures. Natural aggregates, which are generally used in its production, are extracted from quarries of igneous, sedimentary, and metamorphic rock deposits.

The demand for aggregates is rising on a daily basis because concrete is the most widely used material in the world for civil constructions (Aïtcin, 2000; Crow, 2008). This extensive use has led to a significant depletion of natural resources, so for sustainability reasons there is an increasing need to reuse wastes to limit the exploitation of natural raw materials as well as to reduce the impact on the environment. Moreover, the use of construction wastes as an alternative source for aggregates is becoming popular due to increasing landfill charges (Katz, 2003), and as a consequence of the growing awareness that the supply of raw materials is not endless. This is why the Directive 2008/98/CE (European Commission, 2008) has imposed an increase in the use of construction and demolition waste (CDW) to a minimum of 70% by weight by 2020.

Rao et al. (2007) stated that there are barriers to recycling since it may not be the most economically attractive solution, and because there is low confidence in recycled materials. However, research has widely demonstrated that crushed concrete is an excellent source of material for the production of aggregates to be reused in concrete manufacturing (Evangelista and De Brito, 2007). The growing need to use increased quantities of sustainable materials is leading to the identification of new sources and technologies for the supply of aggregates.

The employment of fresh unused concrete is one of these. It comes back to the plants in quantities ranging from 2% to 10% of the estimated 348 million m<sup>3</sup> of ready mixed concrete produced annually in the US (Obla et al., 2007), and of the more than 2 billion tons produced per year worldwide (Crow, 2008). Although normal values are around 0.4% to 0.5% (World Business Council for Sustainable Development Report, 2009), the amount of waste concrete increases to between 5% and 9% of total production during peak periods. Unused concrete results from over-ordered concrete, the frequency of which depends on ordering and delivery practices, and from the leftover concrete in a truck mixer after each delivery (Kazaz et al.,

2016). Nowadays, most of the returned concrete is diverted to landfill sites, while the rest is used in site paving, casting/breaking/re-using, and washing out (Kim and Goulias, 2014).

Landfill operations are causing several climate-change related environmental issues due to the production of gases, water pollution, and limited availability of suitable sites. Moreover, fresh concrete is alkaline so it needs special treatment before being discharged. Site paving of the production plant area is not the best option from a practical point of view because there is a limit to the spaces to be paved (Obla et al., 2007). Casting/breaking/re-using means that fresh concrete is casted into formworks and, after a period of curing, mechanically crushed into parts of different dimensions. While finer particles are difficult to manage and reuse, larger ones are suitable for use in the formation of granular bases and subbases for road pavements. Finally, washing out consists of using water to separate fine and coarse aggregates, which can then be used again in concrete production (Sealey et al., 2001).

The methods currently used to recycle fresh concrete are characterized by their complexity, higher costs, increased working time, and the space required for cumbersome machines, while they also raise a number of environmental issues. In recent years, the pelletization process has increasingly been used in the production of aggregates from by-products and wastes (Tang et al., 2017). For instance, lightweight aggregates can be produced by the agglomeration technique which converts fly ash into a pelletizing disk either by agitation, granulation or compaction (Manikandan and Ramamurthy, 2007; Sivakumar et al., 2012) together with a proportionate quantity of water. Additional procedures during the mixing phase (Harikrishnan et al., 2006) can increase the efficiency of new aggregate formation. The results of tests performed on fly ash aggregate show values comparable with natural gravel, hence it can be regarded as a substitute material in new concrete production (Priyadharshini et al., 2011). In general, pelletization consists of the agglomeration method in which pellets are formed from finer grains thanks to the use of pelletizing agents and devices, the most usual example of the latter being a disk pelletizer (Baykal and Döven, 2000).

The paper presents the results of a laboratory investigation into the production of recycled aggregate from the pelletization of unsettled cementitious mixtures. The innovative feature is the procedure by which pelletization is carried out. The proposed method includes the following steps: (a) addition of the pelletizing agent to an unsettled cementitious mixture, (b) mixing of the material at a constant speed (in a mixer) to produce pellets, (c) discharging of

the pellets, and finally (d) drying of the pellets (Zampini et al., 2016). In real scale applications, the pelletized agent is simply poured into the truck mixer in a quantity proportional to the amount of unsettled concrete present in the rotating mixing drum. After an appropriate mixing time, the pellets are discharged and then left to air. With this procedure, there is no need for further operations which would increase logistic effort and costs. From an industrial point of view, this is a key advantage with respect to other recycling procedures for unused concrete.

The effectiveness of this method was assessed for two different concretes: a conventional concrete (CC), and high performance self-compacting concrete (HPSCC). They were characterized to evaluate size distribution, particle density, water absorption, performance under chemical and mineralogical analysis, and resistance to fragmentation. Test results highlight that the properties of the concrete are partially transferred to the aggregate. Although recycled aggregate presents different properties to those of natural aggregate, pelletized aggregates can certainly be reused in place of natural coarse aggregate for a more sustainable and cleaner concrete production process.

#### 2. MATERIALS AND METHODS

The recycled aggregates were produced from unsettled cementitious mixtures: a conventional concrete (CC) and a high performance self-compacting concrete (HPSCC). An assessment of these concretes was performed to establish if the same process could be used to obtain recycled aggregates from different concretes, and to determine if the mix-type affected aggregate properties.

Both concretes contain different quantities of Portland cement (i.e., CEM I 52.5R), which contains a percentage of clinker above 95%. In the preparation of both concrete mixtures, only one cement was used so as to avert any side effects resulting from the inclusion of other binder components, and any misinterpretation of the results. This notwithstanding, the proposed process has been demonstrated to work successfully with any kind of concrete irrespective of its composition; in other words, it works with any cement type, any aggregate type, any admixture type, and with any consistency (Zampini et al., 2016).

Both CC and HPSCC have the same percentage of natural aggregates (fraction 0/4 round, 4/8 crushed and 8/11 crushed). The properties of the raw materials are presented in the following sections.

#### **2.1. RAW MATERIALS**

#### 2.1.1 Cement

The particle size distribution for CEM I 52.5R cement was obtained with the ISO 13320-1 test method. Figure 1 illustrates the position of the grading curve with respect to the limits suggested in Mamlouk and Zaniewski (2011), thus highlighting the fineness of the binder employed in this investigation. Table 1 reports the cement setting time, density, Blaine, and compressive strength results for cement at 2, 7 and 28 days of curing. They were obtained by following EN 196-3, EN 196-6, ASTM C604, ASTM D7481, and EN 196-1 respectively. The Blaine fineness confirms that the cement employed is of a fine-grade, since it is close to the higher limits of the 3000-4500 cm<sup>2</sup>/g typical value range.

Mechanical and physical requirements specified by EN 197-1 are met because the compressive strengths at 2 and 28 days are greater than 30 MPa and 52.5 MPa respectively. A chemical and mineralogical analysis of cement was performed using the X-ray fluorescence (XRF) method adhering to the ISO 29581-2 standard, the loss on ignition (LOI) test using the ASTM C114, and finally the X-ray diffraction (XRD) method took the EN 1744-1 standard into account.

The resulting values reported in Table 2 confirm that the cement used for the investigation meets all the chemical requirements imposed by EN 197-1. Indeed, LOI is less than 5.0%, sulphate content is less than 4.0% and chloride content is null. Table 2 indicates the main components of Portland Cement clinker which are CaO, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub>; it also includes the requirements established by the EN 197-1 standard for SO<sub>3</sub>, Cl<sup>-</sup> and LOI, which are all satisfied. The quantity of belite highlights that the binder used was a common cement whose clinker was cooked at a temperature of 1450 °C. Using lower temperatures than the conventional one for cooking clinker results in different quantities of belite (Tantawy et al., 2014).



**Figure 1**. Particle size distribution for CEM I 52.5 R used in the investigation, with limits reported in Mamlouk and Zaniewski (2011).

Table 1. Physica	I properties of CEM I 52.5 R.
------------------	-------------------------------

Test	standard	unit	value	EN 197-1 Requirements
Initial setting time (Vicat)	EN 196-3	min	160.0	≥ 45
Final setting time (Vicat)	EN 196-3	min	240.0	
Specific surface, Blaine	EN 196-6	cm²/g	4326.0	
Density	ASTM C604	g/cm <sup>3</sup>	3.14	
Bulk density	ASTM D7481	g/cm <sup>3</sup>	1.00	
Compressive strength 2 days (w/b=0.5)	EN 196-1	MPa	38.1	≥ 30.0
Compressive strength 7 days (w/b=0.5)	EN 196-1	MPa	46.6	
Compressive strength 28 days (w/b=0.5)	EN 196-1	MPa	62.5	≥ 52.5

**Table 2**. Chemical analysis by means of X-ray fluorescence, mineralogical analysis by means of X-ray diffraction, and loss of ignition (LOI) test results of CEM I 52.5 R.

Mineral	Mass	Mineral			
Name	(%)	Name	Chemical Formula	iviass (%)	
SiO <sub>2</sub>	19.17	Alite	Ca₃SiO₅	70.23	
Al <sub>2</sub> O <sub>3</sub>	4.79	Belite	Ca <sub>2</sub> SiO <sub>4</sub>	3.67	
Fe <sub>2</sub> O <sub>3</sub>	2.69	C₃A cubic	Ca <sub>3</sub> Al <sub>2</sub> O <sub>6</sub>	3.13	
CaO	64.24	C₃A ortho	Ca <sub>3</sub> Al <sub>2</sub> O <sub>7</sub>	2.62	
MgO	1.80	C4AF	Ca <sub>2</sub> (Al,Fe <sup>3+</sup> ) <sub>2</sub> O <sub>5</sub>	9.93	
SO₃	2.62	Periclase	MgO	0.39	
Na <sub>2</sub> O	0.09	Free lime	CaO	0.00	
K <sub>2</sub> O	1.04	Portlandite	Ca(OH)₂	0.03	
TiO <sub>2</sub>	0.32	Quartz/sand	SiO <sub>2</sub>	0.36	
P <sub>2</sub> O <sub>5</sub>	0.16	Limestone	CaCO <sub>3</sub>	4.42	
Mn <sub>2</sub> O <sub>3</sub>	0.04	Dolomite	CaMg(CO <sub>3</sub> ) <sub>2</sub>	1.07	
Cl⁻	0.00	Gypsum	CaSO4 (H20)2	1.58	
		Calcium sulphate hemihydrate	CaSO4 (H20)0.5	1.93	
		Calcium sulphate	CaSO <sub>4</sub>	0.65	
LOI @ 950°C	2.82				

#### 2.1.2 Aggregates

The physical properties of natural aggregates (NA) were evaluated in accordance with EN 933-1 and 1097-2. Indeed, Figure 2 shows the particle size distribution for all aggregates present in the composition of CC and HPSCC mixtures, while Table 3 reports all the physical, geometrical, mechanical, and durability properties for the three classes of aggregate employed. The results listed in the table serve to underline the suitability of the NA employed for the production of both conventional and high- performance concrete mixtures.

The EN 12620 standard also requires that aggregates used in the production of concrete meet certain chemical requirements, focusing attention on the content of chlorides, sulphides and other constituents, such as the presence of organic material that may modify the setting and hardening properties of the concrete. The test results from chemical analysis by X-ray fluorescence, loss on ignition (LOI), and mineralogical analysis as per the X-ray diffraction are reported in Table 4 and Table 5. The results explain the composition of the natural rock mass from which NA were extracted.



**Figure 2**. Particle size distribution for the three aggregate classes used in the preparation of CC and HPSCC concretes.

Parameters	Standard	0/4 round	4/8 crushed	8/11 crushed
Water absorption (%)	EN 1097-6	0.9	1	1
Particle Density [kg/m <sup>3</sup> ]	EN 1097-6	2650	2655	2655
Bulk density [kg/m <sup>3</sup> ]	EN 1097-3	-	1231.2	1197.5
Shell content, SC (%)	EN 933-7	-	0	0
Shell content category	EN 12620	-	SC10	SC10
Flakiness Index, FI (%)	EN 933-3	-	12.50	10.89
Flakiness Index category	EN 12620	-	FI <sub>15</sub>	FI <sub>15</sub>
Shape Index, SI (%)	EN 933-4	-	23.40	15.80
Shape Index category	EN 12620	-	SI <sub>40</sub>	SI <sub>20</sub>
Particles retained at 1.6 mm sieve [g]	EN 1097-2	-	3855.1	3760.1
Los Angeles coefficient, LA (%)	EN 1097-2	-	23	25
LA category	EN 12620	-	LA <sub>25</sub>	LA <sub>25</sub>
micro-Deval coefficient, MDE (%)	EN 1097-1	-	15.20	14.97
micro-Deval category	EN 12620	-	M <sub>DE</sub> 20	MDE15
Impact test coefficient, SZ (%)	EN 1097-2	-	17.87	18.34
Impact test category	EN 12620	-	SZ <sub>18</sub>	SZ <sub>22</sub>
Freeze and thaw in water, F (%)	EN 1367-1	-	0.92	0.90
Freeze and thaw in water category	EN 12620	-	F <sub>1</sub>	$F_1$
Freeze and thaw in MgSO4 solution, MS (%)	EN 1367-2	-	18.21	19.74
Freeze and thaw in MgSO4 category	EN 12620	-	MS <sub>25</sub>	MS <sub>25</sub>
Freeze and thaw in NaCl solution (%)	EN 1367-6	-	2.89	2.67

**Table 3**. Physical, geometrical, and thermal resistance for the 0/4, 4/8, and 8/11 aggregates.

Table 4. Chemical analysis of 8/11 crushed aggregate by means of X-ray fluorescence and los	S
of ignition (LOI) test results.	

Mineral Name	Mass (%)	Requirements
SiO <sub>2</sub>	57.57	
Al <sub>2</sub> O <sub>3</sub>	2.73	
Fe <sub>2</sub> O <sub>3</sub>	1.03	
CaO	20.27	
MgO	0.28	
SO <sub>3</sub>	0.00	≤ 0.8 % (AS <sub>0,8</sub> )
Na <sub>2</sub> O	0.50	
K <sub>2</sub> O	0.60	
TiO <sub>2</sub>	0.19	
P <sub>2</sub> O <sub>5</sub>	0.06	
Mn <sub>2</sub> O <sub>3</sub>	0.03	
LOI @ 950°C	16.7	

|--|

Mineral Name	Chemical Formula	Mass (%)
Calcite	CaCO₃	29.1
Quartz	SiO <sub>2</sub>	59.3
Albite High	NaAlSi <sub>3</sub> O <sub>8</sub>	6.4
Albite Low	NaAlSi <sub>3</sub> O <sub>8</sub>	2.3
Portlandite	Ca(OH) <sub>2</sub>	0.1
Biotite 6A	K(Mg,Fe)3(OH,F)2(Si3AlO10)	2.7

#### 2.1.3 Superplasticizer

A proprietary Polycarboxylate ether-based (PCE) superplasticizer (CX ISOFLOW 825), characterized by an active solid content of 30%, was used in the production of HPSCC only.

#### **2.2. CONCRETE DESIGN, PRODUCTION AND PROPERTIES**

HPSCC and CC concretes were produced in laboratory according to the job mix formulas shown in Table 6. Two concretes with the same aggregate distribution ratios but different w/b ratios and different paste volumes, were prepared for the investigation. The w/b ratios that characterize the mix design of the two concretes represent the upper and lower limits of concrete classes. Indeed, Table F1 from the EN 206 standard imposes a maximum w/b of 0.65 for XC1 concretes and 0.45 for XA3 concretes. Consequently, at the design stage, the w/b equal to 0.65 was adopted for CC to ensure a classical structural concrete, while a value lower than the limit (w/b=0.3) was used for HPSCC to facilitate analysis of a state of the art concrete for special applications.

The optimal aggregate volume distribution was defined by taking the A&A theory of Andreasen and Andersen (1930) into account, according to which the percentage of (P) passing through a sieve with an opening d [mm], is given by:

$$P(d) = \frac{d^q - D_{min}^q}{D_{max}^q - D_{min}^q} \tag{1}$$

where  $D_{max}$  is the maximum diameter of the aggregates [mm],  $D_{min}$  is the minimum diameter of the aggregates [mm], and q is the dimensionless parameter that varies between 0 and 1.  $D_{max}$  was taken equal to 12.5 mm and  $D_{min}$  equal to 0.063 mm, as per the particle size distribution of the three aggregate fractions presented in Figure 2. The optimum packing with the A&A model was reached with q = 0.37 (Mueller et al. 2011).

	5	00 0		
Component	Unit	HPSCC	CC	
CEM I 52.5 R	kg/m <sup>3</sup>	600	300	
Water binder ratio (w/b)	-	0.3	0.65	
Superplasticizer	% of cement mass	1.5	-	
Aggregate 0/4 round	% volume of total aggregates	45	45	
Aggregate 4/8 crushed	% volume of total aggregates	20	20	
Aggregate 8/11 crushed	% volume of total aggregates	35	35	

**Table 6**. Concrete mix designs used in the production of pelletized aggregate.

Additional support for this value of q comes from other works on the fractal analysis on soils (Bittelli et al., 1999), and on the fractal analysis of concrete (Carpinteri et al., 1999). The papers mentioned provide results for fractal dimension (D) which are very similar to the theoretical one (D = 2.73) calculated with the so-called Menger's Sponge (Carpinteri et al., 1999; Turk et al., 2010). The q used in the A&A model is the inverse of the fractal dimension D for a Menger's sponge-like material:

$$q = \frac{1}{D} = \frac{1}{2.73} = 0.366 \tag{2}$$

The aggregate linear combining coefficients in Table 6, used for the concrete mix design preparation, were computed using the ordinary least square method at each sieve size. Figure 3 shows the graph obtained from the actual linear combination of the particle size distribution for each aggregate with the coefficients mentioned in Table 7 versus the theoretical design curve based on the A&A model, calculated with the coefficients D<sub>max</sub> and q mentioned above.



**Figure 3**. Comparison between the grading curve for aggregates employed in the formation of both HPSCC and CC, and the theoretical curve derived from the A&A model (Andreasen and Andersen, 1930).

The requirements for both fresh and hardened concretes (defined at the design stage) are shown together with their properties as measured in laboratory tests in Table 7. The slump test was evaluated in accordance with EN 206-1, the slump flow as per EN 206-9, the air content as per EN 12350-7 and finally the compressive strength using cubical samples of 150 mm size in accordance with EN 12390. The density values reported in Table 7 were calculated on samples prior to the measurement of compressive strength, by dividing the mass of each by its volume. As expected, the two materials showed very different fresh and hardened properties thus meeting the research aims and covering a large spectrum of concrete that can be used in the production of pelletized aggregates.

Condition			Type of Returned Concrete	
Condition	Design Requirements		HPSCC	CC
	Slump Class		-	S4-S5
	Slump Flow Class		SF1-SF2	-
Fresh	Slump [cm]		-	17
	Slump Flow [cm]		68.0	-
	Air Content (%)		1.0	4.1
	Compressive Strength Class		C60/75	C25/30
		1 day	54.9	10.9
	Compressive Strength [MPa]	7 day	71.8	27.6
Hardened		28 day	84.0	31.3
		1 day	2419.8	2309.9
	Density [kg/m³]	7 day	2418.5	2296.7
		28 day	2417.8	2334.1

 Table 7. Physical and mechanical properties of HPSCC and CC concretes.

#### 2.3 PELLETIZATION PROCESS

In the pelletization process, a pelletizing agent was added to the still-fresh concrete mixture. The procedure, which was carried out in accordance with patents PCT/EP2015/062689 and PCT/EP2016/062868 (Zampini et al., 2016), used the commercial pelletizing agent called CX ISOCYCLE 9130 (Claim 3 of PCT/EP2016/062868). In the experiments conducted in support of the two patents, a time delay of up to 2 hours between the mixing stage and the starting of the pelletizing process was deemed acceptable since no differences in the physical, geometrical, and mechanical properties of the PRA were observed.

Fresh concrete was produced by putting the natural aggregates (NA), cement, water and, in the case of HPSCC, the superplasticizer, in the bowl mix characterized by a speed of 23 rpm. A mixing time of 10 minutes was chosen to ensure homogenization of all the components (concrete production). In this experiment, concrete production preceded the pelletization process.

The concrete and the pelletizing agent, in a dosage level of 1.5 kg/m<sup>3</sup> determined following a number of preliminary tests conducted in the course of the development of the patents PCT/EP2015/062689 and PCT/EP2016/062868 (Zampini et al., 2016), were poured into an electric bowl mixer with a gross capacity of 170 dm<sup>3</sup> (Figure 4A). The same 23 rpm mix preparation speed was adopted for a duration of 4 minutes to facilitate the dispersion of the pelletizing agent in the concrete (Figure 4B). On completion of this procedure, the pellets were discharged into a pile and left to dry at a temperature of 20°C for a curing period of 24 hours with a relative air humidity value of 55% (Figure 4C). After that, the hardened pellet is ready to be used as recycled aggregate for different applications (Figure 4D). The production method was identical for the two concretes.



**Figure 4.** Sequence of images of pelletized recycled aggregate production: (A) inclusion of the pelletizing agent; (B) mixing at 23 rpm, 4 minutes; (C) curing at 20°C, 55% RH, 24 hours; (D) final material.

#### 2.4 TESTING PROGRAM

Aggregate produced by this process was then analysed to derive particle size distribution as per the EN 933-1 standard. Water absorption test, particle density on a saturated and surface-dried basis ( $\rho_{ssd}$ ) apparent particle density ( $\rho_a$ ), and particle density on an oven-dried basis ( $\rho_{rd}$ ) values were evaluated recurring to EN 1097-6. To characterize the aggregate in terms of density and water absorption the sample was divided into two fractions: 0.063-4 mm, and 4-31.5 mm. The testing procedure was basically the same for both fractions, with the only difference being the sample mass which was differentiated as a function of the maximum size of the aggregates, and the evaluation of the Saturated Surface Dry (SSD) mass. Indeed, for the finest fraction, the SSD was evaluated with the cone method which established that on removal of the cone, the aggregate almost collapses leaving just a peak visible. For the coarse aggregate the SSD condition is defined when all the permeable voids are full of water and the surface is dry.

As with the assessment of NA properties (Table 3), the geometrical, physical, chemical, mechanical, and durability requirements of PRA were checked to ensure compliance with to EN 12620 on aggregate classification for concrete production.

#### 3. RESULTS AND DISCUSSION

Figure 5 shows that both concretes perform in a similar way, yielding a pelletized material that can be classified as coarse aggregate, seeing as the percentage of material passing at a 4 mm mesh sieve is around 10%. PRA derived from HPSCC show a lower curve than that for the PRA from CC, which means that the first PRA is coarser than the second one.

The pellet creation process was the same in both cases. The only thing that changed was the mix design of the two concretes. It should be noted that, referring to a m<sup>3</sup> of mix, HPSCC has a higher paste volume than CC: 380.08 l/m<sup>3</sup> for HPSCC and 290.54 l/m<sup>3</sup> for CC. So, for HPSCC, the quantity of paste that enwraps the natural aggregate during the pelletization process is greater than that for CC. Similarly, the aggregate volume in CC is higher with respect to HPSCC, so the surface to cover is greater as well.



**Figure 5**. Particle size distribution of the PRA derived from CC and HPSCC concretes and comparison with the original grading distribution of aggregates used in CC and HPSCC production.

For the purpose of making a detailed comparison of the particle size distribution values for the two aggregates, three diameters corresponding to 10, 50 and 90% of the material passing have been considered here (i.e.,  $D_{10}$ ,  $D_{50}$ , and  $D_{90}$  respectively). Furthermore, to understand if the aggregate can be considered monogranular the analysis is based on the dimensionless parameter  $D_{90}/D_{10}$ . Granular materials may be considered monogranular when  $2 < D_{90}/D_{10} < 10$ . A monogranular particle size distribution means that it includes a small quantity of fractions, assuming a very vertical profile. In this case the definition of the fraction to be replaced in the composition of a concrete mix is identified by  $D_{50}$  parameter. Table 8 exhibits results for pelletized recycled aggregates coming from CC and HPSCC: both aggregates can be classed as monogranular, so fractions of natural aggregate can be replaced by PRA in the production of a new concrete mix.

**Table 8**. Size parameters of the pelletized recycled aggregate (PRA) obtained from CC and HPSCC concretes.

Origin of aggregate	D <sub>10</sub>	D <sub>50</sub>	D <sub>90</sub>	D <sub>90</sub> /D <sub>10</sub>
PRA - HPSCC	5.60	10.63	23.15	4.14
PRA – CC	3.22	8.96	13.48	4.19

Table 9 shows the density and water absorption test results. It is worth noting that the densities of the two fractions differ from the density values of natural aggregates, expressed in Table 3. Indeed, pelletized recycled aggregates never reach the density of aggregates directly extracted from the rock mass. Moreover, water absorption values are always greater than the typical values for natural aggregates. More detailed examination reveals that the highest values are achieved by the coarser fraction. Water absorption is an indirect parameter for the porous rate of aggregates. This means that the composition of PRA is characterized by more voids than the natural one, in both fractions analysed. On the other hand, it should be pointed out that the standard test, based on the SSD state concept, assumes the use of natural material, so it could provide unexpected results in the case of artificial aggregates. Consonant with the above comments, the bulk density value reported in Table 10 is also slightly lower than those exhibited by the original NA (Table 3).

In contrast, significant changes in the geometric properties of PRAs are evident when compared to those of the original NA (Table 3). These changes are a result of the cement paste adhering to grains and, thereby, producing almost perfectly rounded particles, as clearly illustrated in Figure 4. The results in Table 10 indicate that PRAs fall into the first class for both flakiness (FI<sub>15</sub>) and shape (SI<sub>15</sub>) indexes for all size fractions.

The mechanical properties as per the Los Angeles and Impact tests suggest that PRAs are more sensitive to degradation actions than the NA from which they derive. The comparison between LA categories (Table 10 vs. Table 3) indicates that the LA<sub>25</sub> category of original aggregates has been replaced with categories ranging from LA<sub>30</sub> to LA<sub>40</sub> in the case of PRAs. The surface of PRA is made of hardened paste which is less resistant to erosion and degradation than the core which consists of the original aggregate. When struck, small particles of paste separate from the tougher core and increase the fine produced during the tests. This also explains the results on Impact tests with SZ classes that change from SZ<sub>18</sub>-SZ<sub>22</sub> (Table 3) to SZ<sub>22</sub> – SZ<sub>26</sub> (Table 10).

The different degradation process reproduced with the micro-Deval test leads to better results than for the two previous testing methods. The results of PRA-HPSCC indicate that the two  $M_{DE}$  classes of original aggregates ( $M_{DE}20$  for 4/8 and  $M_{DE}15$  for 8/11 size fractions) are essentially maintained ( $M_{DE}20$ ), while only PRA-CC exhibits a greater sensitivity to abrasion ( $M_{DE}25$ ). These results suggest that PRAs are less sensitive to abrasion than to fragmentation.

Concrete type	PR	A - HPSCC	PRA	- CC
Fractions	0.063 - 4 mm	4 - 31.6 mm	0.063 - 4 mm	4 - 31.6 mm
ρ <sub>ssd</sub> [kg/m <sup>3</sup> ]	2262	2250	2141	2364
ρ₄ [kg/m³]	2346	2445	2195	2554
ρ <sub>rd</sub> [kg/m³]	2199	2115	2095	2241
Water absorption (%)	2.8	6.4	2.2	5.5
Notes: $o_{set}$ = saturated surface dry particle density: $o_a$ = apparent particle density: $o_{rd}$ = oven-dry particle density				

**Table 9**. Density and water absorption test results for the pelletized recycled aggregate (PRA) obtained from CC and HPSCC concretes.

The differences in toughness and porosity levels between the surface and the core of PRAs help to explain the slight difference observed in the case of the durability test under thermal actions independently of test conditions (in water as well as in MgSO<sub>4</sub> and NaCl solutions).

Table 10 also indicates that the LA test results for PRA from HPSCC are better than those for PRA from CC for the same fraction. This is due to the fact that the paste for HPSCC is characterized by a lower w/b than that of CC, so the PRA coming from HPSCC will be harder to break. Moreover, the LA value for smaller fractions is less than that for coarser ones, for the PRA from both CC and HPSCC.

In conclusion, when comparing the mechanical, physical and durability properties of NA and PRA, it is possible to state that the slightly higher values exhibited by PRA can be attributed to its non-homogeneous structure consisting of a hardened paste surrounding a core of natural aggregate; under the degrading action of mechanical and durability tests the paste separates from the core thus resulting in a slightly higher quantity of fine particles when compared to the original NA. Moreover, this phenomenon can be linked to the size effect on strength behaviour of a material that changes as a function of geometry (Carpinteri et al., 1999).

However, it is important to state that values are always within the current limits accepted in concrete production. This means that PRA can be used in new concrete in accordance with EN 206.

Concrete type	Standard	PRA - HPSCC		PRA - CC	
Fractions		10 - 12.5 - 14	4 - 6.3 - 8	10 - 12.5 - 14	4 - 6.3 - 8
Bulk density [kg/m <sup>3</sup> ]	EN 1097-3	1150.2	1183.4	1212.7	1248.6
Shell content, SC (%)	EN 933-7	0	0	0	0
Shell content category	EN 12620	SC10	SC10	SC10	SC10
Flakiness Index, FI (%)	EN 933-3	0.00	0.00	0.00	0.00
Flakiness Index category	EN 12620	FI <sub>15</sub>	FI15	FI <sub>15</sub>	FI15
Shape Index, SI (%)	EN 933-4	0.00	0.00	0.00	0.00
Shape Index category	EN 12620	SI15	SI15	SI15	$SI_{15}$
Particles retained at 1.6 mm [g]	EN 1097-2	3332.9	3729.2	3051.4	3343.1
Los Angeles coefficient, LA (%)	EN 1097-2	33.34	25.42	38.97	33.14
Los Angeles category	EN 12620	LA35	LA <sub>30</sub>	LA <sub>40</sub>	LA35
Microdeval coefficient, MDE (%)	EN 1097-1	17.52	15.36	21.32	20.17
Microdeval category	EN 12620	Mde20	M <sub>DE</sub> 20	MDE25	Mde25
Impact test coefficient, SZ (%)	EN 1097-2	20.64	19.78	24.37	21.63
Impact test category	EN12620	SZ <sub>22</sub>	<b>SZ</b> 22	SZ <sub>26</sub>	SZ22
Freeze and thaw in water, F (%)	EN 1367-1	1.10	0.95	2.31	1.7
Freeze and thaw in water category	EN 12620	F <sub>2</sub>	F1	F4	F <sub>2</sub>
Freeze and thaw in MgSO <sub>4</sub> solution, MS (%)	EN 1367-2	24.74	21.29	28.71	26.18%
Freeze and thaw in MgSO4 category	EN 12620	MS25	MS25	MS <sub>35</sub>	MS35
Freeze and thaw in NaCl solution (%)	EN 1367-6	3.24	3.87	5.67	6.43

**Table 10**. Physical, geometrical, mechanical and durability test results for the pelletized recycled aggregate (PRA) obtained from CC and HPSCC concretes.

Table 11 and Table 12 show the results from an XRD and XRF analysis of both aggregates. From a chemical analysis it was possible to evaluate the LOI which indicates the loss in mass of a combustion residue after heating it to 950°C. At this temperature, all the water inside the sample evaporates; indeed, for CC, in which the w/b is higher, a higher LOI value was observed. From a mineralogical analysis, the presence of the main original minerals forming the natural aggregates (Calcite, Quartz) may be clearly observed. Albite and biotite, already present in small percentages (of mass) in the natural aggregates, have both experienced an increase in volume thanks to the cement hydration reaction. All other newly detected minerals are simply the by-products of the cement hydration reaction.

Mineral Name	Mass	(%)	Dequirements	
	PRA - HPSCC	PRA - CC	Requirements	
SiO <sub>2</sub>	42.19	42.20		
Al <sub>2</sub> O <sub>3</sub>	9.93	4.28		
Fe <sub>2</sub> O <sub>3</sub>	1.47	1.20		
CaO	31.30	28.16		
MgO	1.00	0.68		
SO <sub>3</sub>	0.69	0.30	≤ 0.8 % (AS <sub>0,8</sub> )	
Na2O	0.61	0.87		
K <sub>2</sub> O	0.96	1.04		
TiO <sub>2</sub>	0.16	0.14		
P <sub>2</sub> O <sub>5</sub>	0.09	0.07		
Mn <sub>2</sub> O <sub>3</sub>	0.04	0.05		
LOI @ 950°C	17.50	20.80		

**Table 11**. Chemical analysis of PRA-CC and PRA-HPSCC aggregate by means of X-ray fluorescence and loss of ignition (LOI) test results.

**Table 12**. Mineralogical analysis of PRA-HPSCC and PRA-CC aggregate by means of X-ray diffraction.

Mineral News	Chamical Formula	Mass	Mass (%)		
wineral Name	Chemical Formula	PRA - HPSCC	PRA - CC		
Calcite	CaCO <sub>3</sub>	23.9	33.8		
Quartz	SiO <sub>2</sub>	34.1	40.8		
Albite High	NaAlSi₃O <sub>8</sub>	10.3	7.7		
Albite Low	NaAlSi <sub>3</sub> O <sub>8</sub>	5.1	4.1		
Portlandite	Ca(OH) <sub>2</sub>	2.4	2.4		
Biotite 6A	K(Mg,Fe)3(OH,F)2(Si3AlO10)	4.4	5.8		
C <sub>2</sub> S_mumme	Ca <sub>2</sub> SiO <sub>4</sub>	5.3	1.9		
C₃S monoclinic (NISHI)	Ca <sub>3</sub> SiO <sub>5</sub>	9.0	0.2		
Brownmillerite	Ca2(Al,Fe)2O5	4.8	2.2		
Chamosite	$Fe_2+_3Mg_{1.5}AIFe_3+_{0.5}Si_3AIO_{12}(OH)_6$	0.8	1.0		

### 4. CONCLUSIONS

The present work provided an assessment of the pelletization process for producing pelletized recycled aggregate (PRA) from unsettled cementitious mixtures. By adding 1.5 kg/m<sup>3</sup> of a specific pelletizing agent to fresh concrete aggregates, the pelletization process can be completed in just 4 minutes, and the PRA can be used after a 24 h curing period of drying. This one-step process can certainly be replicated in the field albeit allowing for a possible variation in mixing times and pelletizing agent dosage levels which may depend on the particular machine used.

In this study, aggregates were formed from two different concretes to analyse the degree to which the starting material affects the properties of the final aggregate produced. The experimental work has led to the following conclusions:

- pelletization as per the methodology presented here can be successfully applied to a range of concretes from conventional ones (CC) to high performance self-compacting (HPSCC) ones;
- PRA particles are round and possess completely different shape and flakiness index values compared to the original NA;
- PRA produced from CC and HPSCC have similar, although slightly lower, physical, mechanical and durability properties (i.e., they are less tough) in comparison to the original natural aggregates (NA);
- nevertheless, both the recycled aggregates (PRA-CC and PRA-HPSCC) comply with the regulations governing the use of aggregates in concrete as per the European standard (EN 12620);
- while the properties of the PRAs obtained from both CC and HPSCC are quite similar, the values for HPSCC are slightly superior than those for CC in terms of mechanical, thermal, and weathering properties.

Although the pelletization process used to produce the PRA in this investigation was replicated in the laboratory only, it can be easily applied in the field using concrete mixer trucks. To adapt the procedure for application in the field, it is important to be aware of the technical characteristics of the machines. The most influential parameters are inclination and speed of the spiral blade. The laboratory investigation was conducted by keeping the slope fixed if the mix was equal to that of a typical concrete mixer truck (around 20°), so no correction had to be applied to this parameter. In contrast, for the duration of the pelletization process in the field, it is necessary to introduce a conversion factor. Thus, considering that the lab mixer frequency of rotation is equal to 23 rev/min and that of the concrete mixer truck is around 14-16 rev/min, a conversion factor of 23/15=1.53 is needed to adjust the procedure for field applications. Hence, the mixing time in the field should be increased by around 50% (i.e., 6 min) with respect to the 4 min adopted in the lab.

The results in Table 10 would suggest that PRAs are sufficiently tough and durable for use in new concrete mixtures for conventional applications. On the basis of the quality classification test carried out in this investigation, there is no a priori contraindication to the use of PRA in high performance concrete formulations, albeit operational decisions should dictate whether it is used together with NA or as a substitute.

As a result, new investigation steps should include the introduction of PRA, as a partial substitute for natural aggregate, into a new concrete matrix. Laboratory tests on these concretes should be developed to determine whether PRA can modify the fresh, hardened and durability properties of the final concrete.

#### ACKNOWLEDGEMENT

This research work was made possible by a cooperation between PoliTO and Cemex Research Group AG in the field of student exchange for internship. Thanks to this agreement, Flavia Meloni spent six months at the research headquarters in Biel (Switzerland).

#### REFERENCES

- Aïtcin, P.C., 2000. Cements of yesterday and today: concrete of tomorrow. Cement Concrete Res., 30(9), 1349-1359.
- American Society for Testing Materials (ASTM), 2015. Standard Test Methods for Chemical Analysis of Hydraulic Cement. ASTM C114 – 15, ASTM International, West Conshohocken, PA, US, DOI: 10.1520/C0114-15.
- American Society for Testing Materials (ASTM), 2012. Standard Test Method for True Specific Gravity of Refractory Materials by Gas-Comparison Pycnometer. ASTM C604 – 12, ASTM International, West Conshohocken, PA, US
- American Society for Testing Materials (ASTM), 2009. Standard Test Methods for Determining
   Loose and Tapped Bulk Densities of Powders using a Graduated Cylinder. ASTM D7481
   09, ASTM International, West Conshohocken, PA, US
- Andreasen, A.H.M., Andersen, J., 1930. Über die Beziehungen zwischen Kornabstufungen und Zwischenraum in Produkten aus losen Körnern (mit einigen Experimenten). *Kolloid*, 50(3), 217–228 (In German).

- Baykal, G., Döven, A.G., 2000. Utilization of fly ash by pelletization process; theory, application areas and research results. Resour. Conserv. Recy., 30(1), 59-77.
- Bittelli, M., Campbell, G. S., Flury, M., 1999. Characterization of particle-size distribution in soils with a fragmentation model. Soil Sci. Soc. Am. J., 63(4), 782-788.
- Carpinteri, A., Chiaia, B., Invernizzi, S., 1999. Three-dimensional fractal analysis of concrete fracture at the meso-level. Theor. Appl. Fract. Mec., 31(3), 163-172.
- Crow, J.M., 2008. The concrete conundrum. Chem. World-UK, 5(3), 62-66.
- De Brito, J., Saikia, N., 2013. *Industrial Waste Aggregates. Recycled Aggregate in Concrete* (pp. 23-80). Springer London.
- European Commission, 2008. Directive 2008/98/EC of the European Parliament and of the council of 19 November 2008 on waste and repealing certain directives. 312(11-2b), Brussels, Belgium.
- European Committee for Standardization (CEN), 2000. Testing Hardened Concrete. EN 12390, Brussels, Belgium.
- European Committee for Standardization (CEN), 2005. Methods of Testing Cement Part 3: Determination of Setting Times and Soundness. EN 196-3, Brussels, Belgium.
- European Committee for Standardization (CEN), 2005. Concrete Part 1: Specification, Performance, Production and Conformity. EN 206-1, Brussels, Belgium.
- European Committee for Standardization (CEN), 2009. Tests for Chemical Properties of Aggregates Part 1: Chemical Analysis. EN 1744-1, Brussels, Belgium.
- European Committee for Standardization (CEN), 2009. Testing Fresh Concrete Part 7: Air Content Pressure Methods. EN 12350-7, Brussels, Belgium.
- European Committee for Standardization (CEN), 2010. Methods of Testing Cement Part 6: Determination of Fineness. EN 196-6, Brussels, Belgium.
- European Committee for Standardization (CEN), 2010. Concrete Part 9: Additional Rules for Selfcompacting Concrete (SCC). EN 206-9, Brussels, Belgium.
- European Committee for Standardization (CEN), 2011. Cement Part 1: Composition, Specifications and Conformity Criteria for Common Cements. EN 197-1, Brussels, Belgium.

- European Committee for Standardization (CEN), 2013. Tests for Geometrical Properties of Aggregates Part 1: Determination of Particle Size Distribution Sieving Method. EN 933-1, Brussels, Belgium.
- European Committee for Standardization (CEN), 2008. Aggregates for concrete. EN 12620, Brussels, Belgium.
- Evangelista, L., De Brito, J., 2007. Mechanical behaviour of concrete made with fine recycled concrete aggregates. Cement Concrete Comp., 29(5), 397-401.
- Harikrishnan, K. I., Ramamurthy, K., 2006. Influence of pelletization process on the properties of fly ash aggregates. Waste Manage., 26(8), 846-852.
- International Organization for Standardization (ISO), 2009. Particle size analysis Laser diffraction methods Part 1. ISO 13320-1.
- International Organization for Standardization (ISO), 2010. *Cement Test methods Part 2: Chemical analysis by X-ray fluorescence*. ISO 29581-2.
- Katz, A., 2003. Properties of concrete made with recycled aggregate from partially hydrated old concrete. Cement Concrete Res., 33(5), 703-711.
- Kazaz, A., Ulubeyli, S., 2016. Current Methods for the Utilization of the Fresh Concrete Waste Returned to Batching Plants. Procedia Engineer., *161*, 42-46.
- Kim, H., Goulias, D.G., 2014. Shrinkage Behavior of Sustainable Concrete with Crushed Returned Concrete Aggregate. J. Mater. Civil Eng., 27(7), 04014204.
- Mamlouk, M. S., Zaniewski, J.P., 2011. *Materials for civil and construction engineers*. Pearson Higher Ed.
- Manikandan, R., Ramamurthy, K., 2007. Influence of fineness of fly ash on the aggregate pelletization process. Cement Concrete Comp., 29(6), 456-464.
- Mueller, F. V., Wallevik, O.H., 2011. Particle packing by gyratory intensive compaction as tool to optimize the aggregate gradation of low binder SCC, Eco-SCC. Proceedings of Our World in Concrete and Structures (36th OWICs), Singapore, 97-100.
- Obla, K., Kim, H., Lobo, C.L., 2007. Crushed Returned Concrete as Aggregates for New Concrete. National Ready Mixed Concrete Association, College Park, MD, US.
- Priyadharshini, P., Ganesh, M. G., Santhi, A.S., 2011. Experimental study on cold bonded fly ash aggregates. *International journal of Civil and Structural engineering*, 2(2), 493.

- Rao, A., Jha, K. N., Misra, S., 2007. Use of aggregates from recycled construction and demolition waste in concrete. Resour. Conserv. Recy., 50(1), 71-81.
- Sealey, B. J., Phillips, P. S., Hill, G.J., 2001. Waste management issues for the UK ready-mixed concrete industry. Resour. Conserv. Recy., 32(3), 321-331.
- Sivakumar, A., Gomathi, P., 2012. Pelletized fly ash lightweight aggregate concrete: a promising material. J. Civ. Eng. Constr. Technol., 3(2), 42-48.
- Tang, P., Florea, M. V. A., Brouwers, H.J.H., 2017. Employing cold bonded pelletization to produce lightweight aggregates from incineration fine bottom ash. J. Clean. Prod., 165, 1371-1384.
- Tantawy, M.A., Shatat, M.R., El-Roudi, A.M., Taher, M.A., Abd-El-Hamed, M., 2014. Low temperature synthesis of belite cement based on silica fume and lime. Int. Sch. Res. Notices, 2014, ID 873215.
- Türk, C., Carbone, A., Chiaia, B.M., 2010. Fractal heterogeneous media. Phys. Rev. E, 81(2), 026706.
- World Business Council for Sustainable Development, 2009. The cement sustainability initiative recycling concrete. (Accessed March 2017, http/www.wbcsdcement.org/pdf/CSI- RecyclingConcrete-FullReport.pdf).
- Zampini, D., Guerini, A., Volpatti, G., 2016. Method to produce aggregates from unsettled cementitious mixtures. Cemex Research Group Ag, Patent no. WO2016198384 A1.