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Development of an Innovative Technique for Demolition Concrete Up-cycling: a Response to Mitigate GHG's Emissions and Natural Resources Depletion

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ABSTRACT

Concrete is the second most used material worldwide and the first used man-made material. Its production entails large emissions of GHG due to lime decarbonation during the clinkering process, as well as large consumption of non-renewable minerals and natural aggregates. Currently legislative pressure aiming at preserving natural resources is growing tighter on natural aggregates extraction, which motivates research for alternative raw materials in building concrete manufacturing. Among these materials, aggregates from waste concrete are the most promising as they already fulfill some of the physical and mechanical requirements for the use in concrete. One key point that has to be considered is the cement paste content at the aggregate surface, since it adversely affects the adhesion between the aggregates and their surrounding cement matrix in concrete, thereby resulting in concrete with low mechanical properties (). Current practices in concrete recycling consist in largely non-selective crushing and screening processes, which do not allow a straight separation between the clean aggregates and the cement paste.

The present study describes a high-voltage pulsed-power process of concrete fragmentation which enables to recover clean aggregates and hydrated cement paste (HCP). This HCP conveys valuable elements – Ca, Si, Fe, Al – that would potentiallyreduce energy and natural resources consumption as well as CO_2 emissions of the clinker making process, if it were introduced in the raw mix of the clinker kiln. Overall environmental benefits of this up-cycling chain depend on the mineralogical composition of the cement paste, which in turn depends on the operational conditions of the concrete fragmentation process. To date, only scarce data are available to assess this recycling scheme. In this framework, our study tackle the subject by putting forward an experimental investigation of the pulsed-power process, coupled with a calculation methodology to assess the environmental benefits associated with the recycling of the obtained hydrated cement paste in clinker manufacturing. The proposed methodology leads the way to a comprehensive environmental assessment of concrete recycling schemes, based on a forthcoming LCA study that will make use of the results presented.

Keywords: concrete, cement paste, aggregates, clinker, CO_2, recycling, fragmentation, pulsed power, environmental assessment

INTRODUCTION

Despite being the second most used material worldwide and the first used man-made material, concrete when recycled does not get a good press. Treated by means of low selective technics, the use of crushed concrete remains restricted to low added-value uses (most often as road base materials) since the produced aggregates are covered with a residual cement paste which impairs a good adhesion during a subsequent concrete reprocessing (Domingo-Cabo*et al* (2009), Gómez-Soberón (2002a, 2002b)).At the present time, none of current technology allows recovering this cement paste, even though its unique composition and reactivity make it an ideal candidate for a clinker raw mix additive among all possible cementitious materials.Thus an additional benefit of crushing out clean aggregateswould be tolower natural resources depletion for Ca, Si, Fe and Al minerals bydirectly feeding clinker raw mix with a a high quality cement paste.

So asto estimate the potential of Recycled Cement Paste use in the raw mix, a calculation methodology has been developed tolink the raw mix composition with the CO_2 emissions and the energy consumption of the clinker process. The mineralogical composition of the RCP, its water content and its mean carbonation rate are taken into account in the calculation procedure. The raw mix composition is constrained by three specific parameters traditionally used by clinker manufacturer: the Lea and Parker lime saturation ratio, the silica ratio and the Kühlalumino-ferric module. The cement paste considered is produced by an innovative process for concrete recycling. This process includes an embrittlement step that uses the energy of high voltage pulses in order to induce a selective fragmentation of concrete waste blocks immersed in water. In addition to being a highly energy efficient technic, pulsed-power fragmentation is highly selective. This technology permits to recovermore hydrated and partly non-carbonated cement paste than a usual impact crushing for new clinker elaboration.

MATHEMATICAL MODELLING

General assumptions

Clinkerisation refers to the high-temperature reactionstaking place in a cement kiln, to dissociate CO_2 from $CaCO_3$ and combine the resulting CaO with silica, alumina and ferrous oxides to form clinker. The resulting powder is mostly composed of following compounds: $3CaO.SiO_2$ (C3S), $2CaO.SiO_2$ (C2S), $3CaO.Al_2O_3$ (C3A), $4CaO.Al_2O_3.Fe_2O_3$ (C4AF), with final lime content below 1 wt per cent.

In what follows we consider a dry clinker processing route in which raw materials are grinded and intimately blended before being pre-heated and sent to the kiln to form the clinker. We develop a mass- and enthalpy-balance model to relate the clinkercomposition to the raw mill composition, and determine the variations in the energy consumption and the CO_2 emissions when part of raw mill is replaced by a hydrated cement paste.

Massbalance

In the model, raw materials consist of limestone ($CaCO_3$, clay (kaolinite: $2SiO_2.Al_2O_3.2H_2O$), iron oxide ore ($Fe_2O_3.H_2O$) and Recycled Concrete (RC), composed of Hydrated Cement Paste (HCP) and sand (S).

Following Olivier (1969), the HCP is assumed to be composed of residual $Ca(OH)_2$ and the 4 following mineralogical phases:

- C3S2H3 : $3CaO.2SiO_2.3H_2O$
- C3AH6 : $3CaO.Al_2O_3.6H_2O$
- C3FH6 : $3CaO.Fe_2O_3.6H_2O$
- C3ACScH12 : $3CaO.Al_2O_3.CaSO_4.12H_2O$

The mean contents of anhydrous compounds in the clinker – CaO, Al_2O_3, FeO and SiO_2 – are estimated by mass conservations of the initial reactants.

Enthalpybalance

The enthalpy-balance model aims at estimating the variation in the energy consumed by the precalciner and the kiln when alternative raw materials are used. It assumes that the temperatures and the residence times in the pre-calciner and the kiln are not altered by the change in the raw mill composition. This implies that the various thermal losses around the equipments or associated with clinker and the transport of hot gases remainunchanged. In this configuration, the variation in the process specific energy (E_S) is only contributed by the variation in the enthalpy of the reaction leading to clinker formation with recycled concrete, $\Delta_r H^{ck}$:

$$E_S = \left(E_{S,ref} - \Delta_r H^{ck}_{ref}\right) + \Delta_r H^{ck} \qquad \text{(Eq.1)}$$

The term in bracket accounts for the thermic losses of the process without recycling, andis thus used as a reference value. $\Delta_r H^{ck}_{ref}$ is the reference enthalpy for clinker formation using hydrated cement paste. In industrial plants of Portland cement dry processing, a mean value for $E_{s,ref}$is 750 kcal per kg of clinker (Elkjaer, 1977).

The calculation of $\Delta_r H^{ck}_{ref}$ and $\Delta_r H^{ck}$ are performed using the enthalpy of formation of the raw materials, the mineralogical phases of the clinker and the gases arising from the clinker formation - H_2O and CO_2 (Table 1), according to the formula:

$$\Delta_r H^{ck} = \sum_{i,j} \left(n_i.H^0_{F,i} - m_j.H^0_{F,j}\right)$$

m_j (resp. m_i) is the mass of reactants j (resp. of products i, clinker and gases). $H^0_{F,i}$ stands for the enthalpy of formation of species i per unit mass.

All calculations are performed at 298 K, since to date no data were found concerning the sequence of reactions accounting for the thermal decomposition of the hydrated cement paste into reactive materials inside the kiln.

CO_2emissions

CO_2emissions resulting from the coal combustion in the kiln can be calculated (Eq. 3) and summed with the emissions from the dissociation of calcite (Eq. 4).

$$m_{CO_2}^{coal} = \frac{y_{coal}.m^{ck}}{M_{coal}}.y_C^{coal}.M_{CO_2} \qquad \text{(Eq.3)}$$

$$m_{CO_2}^{raw} = \left(y_{CaCO_3}^{raw}.\frac{M_{CO_2}}{M_{CaCO_3}} + y_{RC}^{raw}.y_{CaCO_3}^{sand}.\left(1 - y_{HCP}^{RC}\right).\frac{M_{CO_2}}{M_{CaCO_3}} \right).m^{ck} \qquad \text{(Eq.4)}$$

y_{coal} stands for the weight proportion of combustible burnt to achieve the formation of 1 kg of clinker, y_C^{coal} is the carbon molar contents in the combustible, $y_{CaCO_3}^{raw}$ y_{RC}^{raw} are the weight proportions of $CaCO_3$, recycled concrete, clay, and iron ore in raw materials and $y_{CaCO_3}^{sand}$ is the limestone content of sand. y_{coal} is related to the specific energy E_S (Eq.1) according to:

$$y_{coal} = \frac{E_S}{LHV} \qquad \text{(Eq.5)}$$

withLHV: Lower Heating Value of the combustible.

Cement paste substitution in raw mix for clinker production

Several criteria can be usedto control the quality of the clinker made from recycled concrete, and to precise the range of values allowed for the proportions of recycled concrete in the crude materials.

On a mineralogical point of view, a clinker formulation dedicated to a standard Portland cement must meet the following requirements, expressed in weight percent: 45.0 < C3S < 79.7, 5.7 < C2S < 29.8, 1.1 < C3A < 14.9, 2.0 < C4AF < 16.5 (Taylor, 1997). These values correspond to the main oxides composition in clinker: 20.0 < SiO_2< 24.3 %, 3.7 < Al_2O_3< 7.1, 1.7 < Fe_2O_3< 5.7 and 61.0 < CaO < 68.0 %. These ranges of acceptable values are interrelated according to various characteristic ratios stemming from the mechanistic description of clinker formation in kiln. Main ones are the Lea and Parker formula for lime saturation ratio (LaP), the Kühlalumino-ferric(MK) and silica ratio (SR). Associated ranges of acceptability are:

$$LaP = \frac{y_{CaO}^{ck}}{2.8y_{SiO_2}^{ck} + 1.1y_{Al_2O_3}^{ck} + 0.70y_{Fe_2O_3}^{ck}} \Rightarrow 0.90 < LaP < 0.95$$

$$SR = \frac{y_{SiO_2}^{ck}}{y_{Al_2O_3}^{ck} + y_{Fe_2O_3}^{ck}} \Rightarrow 2.0 < SR < 3.0 \qquad \text{(Eq.6)}$$

$$MK = \frac{y_{Al_2O_3}^{ck}}{y_{Fe_2O_3}^{ck}} \Rightarrow 1.50 < MK < 2.50$$

Lime saturation ratio represents the ratio of total lime content to the lime content which entirely combines with SiO_2, Al_2O_3 and Fe_2O_3. A high SR meansahigh silica content at the expense of flux agents, resulting in an impaired clinker burnability. Alumino-ferric ratio gives an indication of the formation of the C3A phase: below a value of 1.5, only alumina combines with iron oxide and only C4AF is formed. Lastly, the total of the four major oxides is considered to show very little variation, due to the fact that most cement plants use raw material from reliable sources that limit the variability of the minor and the major constituents. Therefore, the following sum is taken as a constant:

$$t = y^{ck}_{CaO} + y^{ck}_{SiO_2} + y^{ck}_{Al_2O_3} + y^{ck}_{Fe_2O_3} \qquad \text{(Eq.7)}$$

The mean value for t found after a review of six Portland cement plants by Moore (1982)is 0.96.

Considering the dependence of CaO, SiO_2, Al_2O_3 and Fe_2O_3 content on the proportions of the crude materials and on the part of HCP in the recycled concrete, fulfilling these previous inequalities brings a range of acceptable values for each main constituent in the crude materials. In what follows, the proportions of crude materials ($CaCO_3$, clay, iron ore and recycled concrete) are expressed in term of the previous characteristic ratios, given (Eq.6) and the mass conservation of CaO, SiO_2, Al_2O_3 and Fe_2O_3along the reaction path between the crude materials and the clinker. Considering the content of these latter species in the recycled concrete as parameter values, this approach leads to 4 equations with 4 parameters and 3 variables, whose range of variation are known. Setting the parameters values, the determination of theacceptable range of variation for the crude materials proportions consist in calculating the absolute minima and maxima of each of the 4 equations. A Sequential Quadratic Programing algorithm is implemented in MATLABand is used to solve the equation system.

EXPERIMENTAL

Experiments are performed on concrete wasteusing two different technologies: an impact crusher, which is widely used for concrete recycling, and an innovative technique based on pulsed power fragmentation. The objectiveis to evaluate the feasibility of the pulsed-power route to induce a selective fragmentation of the concrete.

The initial material is composed of 18wt per cent of hydrated cement paste and 82wt per cent of a mixture of silica sand and aggregates. This latter mixture is made up of 40wt per cent of -2 mm sand and 60wt per cent of 2-20 mm aggregates.

The impact crushing is performed on a Hazemagimpactor with a rotation speed of 290 rpm.The pulsed power fragmentation is performed on a laboratory device designed by Selfrag Company (Switzerland). The material is loaded into a 0.2 dm^3 bowl filled with tap water and whose bottom consists of a counter electrode. A second electrode is fixed on the upper part of the bowl, and a high-voltage current is discharged onto concrete blocks. The electrical breakdown of solids takes place when the voltage of the pulse exceeds the electrical strength of the mineral matrix. High voltageaffected concrete blocks are splitted along grains interfaces characterized by different permittivity and/or electrical conductivity. Operating parameters are the

working electrode gap, the number of pulses, the pulse frequency and the voltage.Each experiment consists of several batches (450 g each) to treat at least 10 kg of material.Two configurations are tested: *conf.1* (working electrode gap: 30 mm, number of pulses: 5, voltage: 110 kV) and *conf.2* (working electrode gap: 30 mm, number of pulses: 15, voltage: 125 kV).The amounts of fine particles produced at the outlet of both fragmentation devices are obtained by manual sieving (-2 mm). The cement paste content (assumed to be similar to the insoluble content) is obtained after an acid digestion of the recovered fine particles, performed by contacting the solid with an18.5 wt percent hydrochloric acid solution in a ratioliquid:solid 3:1during 4 h.

Results

Table 2 summarizes the preliminary results. The chemical characterization is currently under way and will be given on the oral presentation.

Table 2

According to these results, the cement paste content shows similar values throughout the tests, i.e with a crushing or a pulsed power configuration (52 – 49 – 48 %wtrespectively). The amounts of fines produced with fragmentation are at least equivalent to the one produced with the usual crushing process, around 3 wt per cent. This value corresponds to the direct outlet of the concrete crushing/fragmentation processes and would beincreased by adding an attrition step on both processes to recover more fines.In the case of selective electrofragmentation, the embrittlementpreferentially occurs at aggregates surface and not within. Thus, less aggregates particles are likely to be produced during the attrition stage; therefore, the quality of the fines fraction recovered could be improved. Tests are currently under progress to support this assumption. With more stringent conditions of fragmentation (*conf.2*) the fines amount reaches 10 wt per cent. The energy consumptions are almost the same and remains low, around 1 kWh/t of materials.

MODEL IMPLEMENTATION

Two cases are considered, with and without recycled concrete.In both cases, the solid combustible is a dry bituminous coal with a lower heating value of 8500 kcal/kg. The air required for the combustion is supposed to be dry.The raw mixesare assumed to be dry when they enter the preheater of the clinker production process. In the case of no recycled concrete use, raw materials are the pure minerals considered previously: limestone ($CaCO_3$), kaolinite ($2.SiO_2.Al_2O_3.2H_2O$), silica (SiO_2) and goethite ($Fe_2O_3.H_2O$). In the case of recycling, same raw materials are used (except for silica) and the Recycled Concrete is made up of y_{HCP}^{RC} percent of Hydrated Cement Paste and $(100 - y_{HCP}^{RC})$ percent of a silica Sand composed of 99 wt per cent of SiO_2, and 1 wt per cent of Fe_2O_3.Given the experimental results, the model is implemented considering a fine fraction made of $y_{HCP}^{RC} = 50$ wt per cent of a hydrated cement pasteand 50 wt per cent of silica sand. To remedy the temporary lack of information concerning thecontents of chemicals in the cement paste, calculations are made with literature data (Olivier, 1969). Table 3 gives the corresponding composition.

Table 3

Given the contents in CaO, SiO_2, Al_2O_3 and Fe_2O_3 in recycled concrete, calculations give the following range of variations for limestone(y_{CaCO3}^{raw}), clay (y_{clay}^{raw}), iron ore (y_{I}^{raw}) and RC (y_{RC}^{raw}) proportions in the raw mix(expressed in weight parts calculated on the basis of 100 weight parts of clinker produced):

$$104.9 < y_{CaCO3}^{raw} < 109.2\,,\ 9.9 < y_{clay}^{raw} < 18.8\,,\ 1.9 < y_{I}^{raw} < 4.5\,,\ \text{and}\ 22.3 < y_{RC}^{raw} < 33.0$$

From these values, and a given case of clinker processing without recycled cement paste, Table 4 describes the results of mix proportions calculations obtained in two cases : *(i)* when modules with recycled cement paste are equal to modules without recycling, and *(ii)* when the biggest reduction in CO_2 emissions is targeted (this corresponds to the minimum value allowed for y_{CaCO3}^{raw}).

Table 4

According to the results, the specific energy consumption is reduced by 3per cent when recycled concrete is used and clinker with equal module is formed. Total calcite content in a recycling targeting the lowest possible CO_2 emissions corresponds to a calcite introduction in the raw mix of 107.9 weight parts,or similarly a decreaseof14per cent in calcite consumption, along with small reductions in iron ore and kaolinite minerals. Decrease in natural resources requirements brings about a reduction in CO_2 emissions, by a factor of 10 per cent.

Recycled concrete from demolition waste amounted to 15 Mtpa in France in 2008 (CGDD 2010), which represents:

- a maximum of 12 Mtpa recoverable aggregates, covering 15 wt per cent of the yearly natural aggregates consumption for the production of building concrete.
- a maximum 3 Mtpa of hydrated cement pastewhich recycling in the clinker raw mix would also ensure 15 wt per cent of national cement consumption, along with a reduction of $2.4.10^5$ tons in CO_2 emissions each year given the previous theoretical calculations at one cement kiln scale.

CONCLUSIONS

On the basis of theoretical calculations, the present study proves that reusing the cement paste contained in concrete waste as a raw constituent for clinker processing,lowers the CO_2 emissions as well as the mineral resources consumption. The exact potential substitution in the raw mix will be refined with the forthcoming

chemical characterization of the cement pastes obtained at the outlet of a pulsed power fragmentation process and associated downstream processes. To evaluate the environmental benefits in using pulsed-power in place of impact crushing for concrete waste treatment, a comprehensive comparison will be drawn and will include the various consumptions and emissions of both processes, withoutsidesteppingthe reuse of the cleaned aggregates generated along with the cement paste in the pulsed power process for new concrete production. This forthcoming evaluation will performed according to a LCA methodology, that will make use of the theoretical and experimental results given in the present study.

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TABLE CAPTIONS

Table 1 – Enthalpies of formation for reactants and products involved in clinker processing

Table 2 – Amounts of fines produced by a usual impact crushing and the pulsed-power fragmentation process

Table 3 – Mineralogical and chemical compositions of the recycled concrete used in the model implementation

Table 4 – Estimation of minerals amounts, emissions of CO2 and enthalpy of clinker formation in the case of no recycling in the crude materials, and recycling of a mixture hydrated cement paste/sand : 50/50 wt per cent. ΔH_F^{clk} : enthalpy of clinker formation. E_S : specific energy.

TABLE 1

Enthalpies of formation for reactants and products involved in clinker processing.[1]: CEMDATA07 data base, see Lothenbach*et al* (2008), Matschei*et al* (2007) and Möschner*et al* (2008). [2]: Barin*et al* (1977). [3]: Barin*et al* (1993). [4]: Barin*et al* (1989). [5]: Frenkel*et al* (1994). [6]: Landolt*et al* (2002).

Material	$H^0_{F,i}$, 298 K **(kJ.kg^{-1})**
C3S[1]	- 12 855
C2S[1]	- 13 419
C3A[1]	- 13 189
C4AF[1]	- 10 453
C3AH6[1]	- 14 656
C3FH6[1]	- 10 642
C3ACScH12[1]	- 14 067
C3S2H3[2]	- 13 970
$Fe_2O_3.H_2O$[2]	- 662
$CaCO_3$[3]	- 12 054
$Ca(OH)_2$[3]	- 13 310
$2SiO_2.Al_2O_3.2H_2O$[3]	- 15 952
Fe_2O_3[3]	- 5139
SiO_2[4]	- 15 166
CO_2[5]	- 8935
H_2O[6]	- 13 422

Table 2

Amounts of fines produced and associated content of cement paste by an usual impact crushing and the pulsed-power fragmentation process

	Energy consumption	Amounts of fines	Content of cement paste
	kWh/t	% weight	% weight
Impact crushing	1	3	52
Conf.1 - pulsed-power fragmentation (gap: 30 mm, 5 pulses, 110 kV)	0,5	3	49
Conf.2 - pulsed-power fragmentation (gap: 30 mm, 15 pulses, 125 kV)	1,5	10	48

Table 3

Mineralogical and chemical compositions of the recycled concrete used in the model implementation

Mineralogical Composition		Chemical Composition	
$CaCO_3$	0	CaO	25.59
SiO_2	49.50	SiO_2	58.23
$Fe_2O_3.H_2O$	0.50	Al_2O_3	2.19
C3S2H3	24.89	Fe_2O_3	1.69
C3AH6	4.35	MgO	0.60
C3FH6	3.25	Na_2O+K_2O	0.30
C3ACScH12	6.17	CO_2	6.60
$Ca(OH)_2$	10.51	H_2O	10.32

Table 4

Estimation of minerals amounts, emissions of CO_2 and enthalpy of clinker formation in the case of no recycling in the raw materials, and recycling of a mixture hydrated cement paste/sand : 50/50wt per cent. ΔH_F^{clk} :enthalpy of clinker formation. E_S : specific energy.

Raw Materials	**wtper cent relative to clinker**			**Clinker**	**wt per cent relative to clinker**		
	No recyling	**Recyling** *Equal modules*	**Recyling** *Lowest CO_2*		**No recyling**	**Recyling** *Equal modules*	**Recyling** *Lowest CO_2*
$CaCO_3$	121.7	107.9	104.9	**C3S**	57.9	56.8	54.5
$2.SiO_2.Al_2O_3.2H_2O$	16.5	14.8	10.3	**C2S**	21.2	21.2	27.8
SiO_2	14.9	(-)	(-)	**C3A**	12.4	12.4	7.4
$Fe_2O_3.H_2O$	3.1	2.5	2.9	**C4AF**	8.3	8.3	9.6
Recycled cement paste	(-)	26.5	33.0				
CO_2 emissions	**wt per cent relative to clinker**			**Modules for clinker characterization**			
	No recyling	**Recyling** *Equal modules*	**Recyling** *Lowest CO_2*		**No recyling**	**Recyling** *Equal modules*	**Recyling** *Lowest CO_2*
Raw materials	53.5	47.5	46.2	**LaP**	0.94	0.94	0.90
Coal combustion	26.5	26.7	25.8	**MK**	2.36	2.36	1.50
Total	80.0	74.2	72.0	**SR**	2.44	2.44	3.00
Enthalpy							
	No recyling	**Recyling** *Equal modules*	**Recyling** *Lowest CO_2*				
ΔH_F^{clk} for 1 kg clinker	1623 kJ	1575 kJ	1479 kJ				
Specific Energy E_S for 1 kg clinker	2945 kJ	2897 kJ	2801 kJ				