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AN OVERVIEW OF SUSTAINABLE CONCRETES WITH MAXIMIZED AGGREGATE CONTENT: NATURAL LIMESTONE VERSUS STEEL-MAKING SLAGS

García-Cortés, Verónica¹; Garcia, David²; Revilla-Cuesta, Victor³; Romera, Jesús-María⁴; San-José, José-Tomás⁵*

 Basque Research and Technology Alliance (BRTA), TECNALIA. <u>veronica.garcia@tecnalia.com</u>, <u>https://www.tecnalia.com</u>
 Basque Research and Technology Alliance (BRTA), TECNALIA. <u>david.garcia@tecnalia.com</u>, <u>https://www.tecnalia.com</u>

3: University of Burgos, Department of civil Engineering.

vrevilla@ubu.es, https://investigacion.ubu.es/grupos/1820/detalle

4: University of the Basque Country, Department of Mechanical Engineering.

jesusmaria.romera@ehu.es, https://www.ehu.eus/es/web/ingenieria-mecanica/home

5: University of the Basque Country, Department of Mining, Metallurgical and Materials Science. josetomas.sanjose@ehu.es, https://www.ehu.eus/en/web/im3/

ABSTRACT

The conversion of various industrial by-products from Spanish factories into co-products used in partial substitution of cement and concrete aggregate has been extensively studied since the 1990s. Building on that research effort, the present investigation is focused on improving the packing density of concrete aggregates, with special emphasis on two central objectives: firstly, the reduction of cement and natural aggregate content within concrete; secondly, the validation of their substitution by Electric Arc Furnace Slag (black-slag) aggregate. To do so, several experimental campaigns were conducted, in which 4 compaction procedures were applied under dry conditions to: 4 sieved fractions of natural limestone and 3 sieved fractions of black-slag aggregates. The physical properties of the 7 sieved fractions had previously been characterized and compared with theoretical models, in order to validate their dosing in the experimental tests: Fuller curve, Funk and Dinger curve, Compressible Packing Model, and the 3-Parameter Packing model. The aggregate-packing densities were experimentally and theoretically studied with dry methods. Our findings showed that, unlike natural aggregates, other methods based on aggregate shape are preferable for black-slag mixtures, due to the specific textures and their abrupt particle contours. The conclusions from the investigations were that both the Compressible Packing Model and the 3-Parameter Packing models produced valuable packing-density predictions for the binary mixes.

KEYWORDS: Compressible Packing Model (CPM); 3-Parameter Packing Density Model (3-PM); Electric Arc Furnace Slag (ES); Natural (limestone) Aggregate (NA); concrete design.

1. INTRODUCTION

Concrete is among the materials that humankind has processed in ever greater quantities and yet its negative environmental effects have only recently raised great alarm: CO₂ emissions, high (non-renewable) resource consumption, and raw-material extraction. Since the 1990s, the properties of various industrial by-products from Spanish factories have been studied for their partial substitution of cement and concrete aggregates. Building on that research effort, the present investigation is focused on improving the Packing Density (PD) of concrete aggregates, with emphasis on two central objectives. Firstly, the reduction of cement (the more compact the aggregate structure, the

less the need for cement paste) and natural aggregate (NA, 70% by mass of concrete) content within concrete. Secondly, the validation of Electric Arc Furnace Slag (ES, black-slag recovered from the primary steel-making process) aggregate used in substitution of concrete aggregate (as an example, 0.4 Mt/year alone is produced in the Basque country), in order to reduce these wastes through their reuse in the manufacture of sustainable concretes [1-2].

One of the earliest pioneers [3] recognized that concrete strength is affected by aggregate gradation. Among others, there is the research work of Funk and Dinger on a set of ideal grading curves [4]. On the one hand, discrete methods were applied to concrete mixes that incorporated NA and recycled concrete aggregates [5]; however, their applicability to ES aggregates and PD prediction has still to be demonstrated. On the other hand, the Compressive Packing Model (CPM), of demonstrated accuracy at predicting PD [7], was extensively used [6] and, the 3-parameter packing model (3-PM) that first included the wedging effect over 8 years ago, has presented new theories based, respectively, on the wall effect and the loosening effect [8].

Aggregate PD was experimentally and theoretically studied (with dry methodologies), after which the NA and ES aggregates were compared with 4 theoretical models, in order to validate the experimental results for their dosing: Fuller curve, Funk and Dinger curve, CPM, and 3-PM. Our findings showed that, unlike NA aggregates, other methods based on aggregate contours should be preferred for ES mixtures, due to the specific shape and textural variability of these co-products. In conclusion, the investigations showed that both the CPM and the 3-PM models produced valuable packing density predictions for binary mixes, although the superior performance of the 3-PM model was evident in relation to both the ternary and the quaternary mixes.

2. MATERIALS AND METHODS

2.1. Materials: aggregates, cement and water

Two aggregate types from Northern Spain (Basque Country) were mixed (see figure 1). Firstly, natural aggregate (NA) from limestone quarries in 4 fraction sizes: 11/22, 4/11, 0/4, and 0/2 millimeters, respectively. Secondly, Electric Arc Furnace Slag (ES) aggregate in 3 fraction sizes: 11/22, 4/11 and 0/5, after crushing and spontaneously weathered outdoors for three months, until their volumetric stabilization (a process that creates surfaces with an angular morphology).



Figure 1: Aggregates under study.

Some physical properties from NA and ES aggregates are presented in Table 1. The average-size particle aggregate fraction is a key point in the discrete packing model. However, there is no a clear agreement on its calculation. Some authors [9] calculated the main aggregate diameter according to specific distributions, others [6] defined it as a geometric mean, and yet others established that mean diameter represented 50% of all retained particles, etc. In the present research, the mean size of each fraction was calculated from the particle-size distribution [10].

Aggregates	Saturated surface dry gravity (kg/dm ³)	Specific dry gravity (kg/dm ³)	Bulk density (kg/dm ³)	Absorption (%)	Fineness modulus	Mean size aggregate fraction (mm)
NA (0/2)	2.7	2.7	1.6	1.8	2.8	1.0
NA (0/4)	2.7	2.7	1.6	0.9	3.0	1.2
NA (4/11)	2.6	2.6	1.4	1.1	6.2	7.6
NA (11/22)	2.7	2.7	1.3	0.4	7.7	18.1
ES (0/5)	3.6	3.5	2.1	1.8	4.0	2.3
ES (4/11)	3.5	3.4	1.8	2.2	6.2	7.4
ES (11/22)	3.57	3.4	1.7	1.7	7.5	16.8

Table 1: Properties of aggregates.

The NA consisted of a commercial natural limestone (calcite fraction >95%) and, as detailed by the authors [1], some limestone fine fraction (0/2) was added to improve concrete workability by avoiding segregation and to compensate for the spontaneous presence of fewer fine particles within the smaller size fraction (0-5) of ES (siderurgical aggregates).

ES aggregates are a by-product of steel processing where, in the primary steel-making process, ferrous scrap is smelted in electric arc furnaces. These black slags are gravelling products (stony materials) with a higher density than NA, and an interesting mechanical performance [1-2], but their superficial roughness, in general, hinders adequate in-fresh workability. The main chemical components of the ES were: Fe₂O₃ (22.3%), CaO (33.0%), SiO₂ (20.2%), Al₂O₃ (12.2), MnO (5.0%), MgO (3.0%), Cr₂O₃ (2.0%), TiO₂ (0.8%), P₂O₅ (0.5%), SO₃ (0.4%), and others (0.6%); XRD analysis revealed Wüstite, Ghelenite, and Kirsteinite.

In the present research, a CEM II/A-M (V-L) 42.5R containing Portland clinker (80%) was mixed with active additions: fly ash (9%), limestone (9%) and others (2%). This cement had a specific density of 2.99 kg/dm³ and a specific Blaine surface of 4130 cm²/g.

The mix water source from the urban mains supply of the city of Derio (Spain) contained no compounds with adverse effects on hydraulic mixes. Besides, no admixtures were added, in order to avoid undesirable effects of the aggregate proportion on mix properties (the admixtures increased the complexity of particle packing).

2.2. Testing the aggregates compactness

The maximum packing densities of NA and ES aggregate mixes were tested under dry methods (packing density measure), which is highly sensitive to the compacting energy. This compacting energy was measured with 4 methods: loose (L), compacted by tamping rod (C), compacted on a vibration table at a frequency of 26 Hz under 10 kPa compression (C26) and, finally, compacted on a vibration table at 33 Hz and 10 kPa (C33), respectively. L and C methods follow UNE-EN 1097-3 (1999) and C26, while C33 follows the [6] research indicated in Figure 2: a pre-set amount of 5.3 kg NA plus 7 kg of ES poured into a cylindrical container and then pressurized at 10 kPa and vibrated for 3 min.



Figure 2: C26 and C33 compaction device described in the procedure [2].

2.3. Particle packing models

The experimental results of dosing NA and ES aggregates were compared with the results of 4 theoretical models:

- -Fuller curve (optimized curve). This method requires a particle-size distribution and its output is the aggregate proportion according to the best fit with the curve.
- -Funk and Dinger curve (optimized curve) [4]. This method requires a particle-size distribution and its output is the aggregate proportion according to the best fit with the curve.
- -CPM (discrete model). This method requires a characteristic diameter of each granular fraction, a particle-size distribution of each granular size and a compaction index. Its output is a prediction of the packing density at different proportions.
- -3-PM (discrete model). This method requires a characteristic diameter and a particle-size distribution of each granular fraction. Its output is the prediction of packing density proportions.

OPTIMIZED CURVES: FULLER (1) AND FUNK & DINGER (2) METHODS

Both curve-equations were applied, to establish an optimal particle-size proportion of NL and ES aggregate in the concrete mixes.

$$P(d) = (d/d_{max})^q \tag{1}$$

$$P(d) = ((d - d_{min})/(d_{max} - d_{min}))^q$$

$$\tag{2}$$

These simple and practical procedures require three inputs: the maximum/minimum (d_{max}/d_{min}) particle size and the particle-size distribution (P). The optimal packing density can be determined by varying the "q" factor. A "q" value of 0.5 is the most widely used for the Fuller curve and 0.37 for the Funk and Dinger curves (other researchers [11] recommended q=0.25÷0.3 depending on concrete types). The optimal NA/ES aggregate rate fractions were obtained by minimizing the residual sum of squares.

COMPRESSIBLE PACKING MODEL (CPM)

The CPM method was applied under 3 scenarios to calculate the virtual packing density (β) of each fraction:

- $-\beta_m$. Each aggregate fraction should be mono-size, neglecting the interaction effects between particle sizes.
- $-\beta$. The second scenario considers the interaction between different particles within the same aggregate fraction. In the end, this procedure was excluded, because the virtual packing density was lower than the real one in some fractions of NA (0/4, 4/11 and 0/2) and ES (0/5 and 4/11).
- $-\gamma$. The virtual packing density was determined on the basis of the second scenario, except that β was considered as the maximum virtual density (γ), defined as the minimum packing density of an aggregate class when the latter is the dominant one in a poly-sized mix.

Once the β factor had been determined in each aggregate fraction, the model was applied to binary mixes. Initially, this method was applied to coarse and medium sizes, grading the volumetric fractions from 0.0 to 1.0 (in 0.1-point increments). It was subsequently applied to the mixing of all NA and ES fractions (in addition to the NA fine fractions).

3-PARAMETER PARTICLE PACKING MODEL (3-PM)

The 3-PM model was applied to binary mixes of coarse/medium aggregate sizes of NA plus ES, following previous research [8]. Afterwards, informed of the developments at the University of Hong Kong under Dr. Wong, based on the multi-component particle-mix model, applied to ternary and quaternary mixtures, it was concluded that the loosening effect, the wall effect and the wedging effect were dependent on the aggregate size-ratio and, additionally, that the former were dependent on both the aggregate morphology (whether rounded or not) and the compaction energy.

3. RESULTS

3.1. Concrete mix design and properties

The 12 concrete mixes presented in Table 2 were prepared. The ES fractions were mixed as needed with 0/2 mm NA, to adjust the siderurgical aggregate grading curves. These mixtures respond to the optimum combinations of aggregates based on experimental packing density results and the 4 theoretical models previously presented.

Materials and menories	Experimental			Funk & Dinger				CPM and 3P				
Materials and properties		NA2	ES1	ES2	NA3	NA4	ES3	ES4	NA5	NA6	ES5	ES6
NA-0/4 (kg/m ³)	905	844			996	978			804	751		
NA-4/11 (kg/m ³)	436	407			579	542			594	555		
NA-11/22 (kg/m ³)	664	619			403	373			603	563		
NA-0/2 (kg/m ³)			402	375			335	376			905	844
ES-0/5 (kg/m ³)			1065	994			919	828			586	547
ES-4/11 (kg/m ³)			630	588			574	518			433	404
ES-11/22 (kg/m ³)			416	389			681	594			430	401
CEMII/A-M(V-L)42.5R (kg/m ³)	260	317	260	317	270	306	267	326	260	317	260	317
q-factor	- 0.31 0.29 0.35 0.33 -											
W/C	0.55 (panish	Structural Concrete Instruction EHE-08)							
Solids (%)	84.4	82	83.4	81.0	82.7	82.2	83.7	81.4	85.0	82.8	82.8	81.0
Slump (mm)	15	80	0	150	0	40	15	160	15	150	15	40
Bulk density (kg/dm ³)	2.4	2.4	2.9	2.9	2.4	2.4	2.9	2.9	2.4	2.4	2.7	2.7
Packing density aggregates (%)	71.6	71.5	72	72.1	75.4	75.8	74.9	74.3	71.4	71.6	74	74
P. density aggregates+CEM (%)	75.2	75.8	74.1	75.0	76.0	76.1	76.5	76.3	75.5	75.9	75.4	74.8

Table 2: Concrete mixes and their properties.

Some of the mixes were cast with a reduced % volume paste (23-24%) and others were dosed with a typical content of $27\div29\%$. Additionally, several q-parameters (more than initially mentioned) were tested until a $260\div300$ kg/m³ cement content. These mixes had been designed regarding be comparable with these three mixing aggregates methods.

According to the ACI 211.1 code, the absolute volume method (2% of air content when the maximum aggregate size is over 20 mm, in a total volume of 1 m³) should be applied to the mixes.

3.2. Factors that affect the packing density of aggregate fractions

The packing densities of both aggregate types (natural and siderurgical) differed, despite their similar grain size distributions and despite having followed a similar compaction process, probably because of the differences in their surface texture (roughness of ES vs plain NA) and shape (depending on whether fractions were crushed or crushed/screened).

The packing density of both the NL and the ES aggregates under study decreased as their mean particle size increased, contrary to the findings of other studies (practically mono-size, while wider fractions were tested in this study), due very probably to a higher agglomeration effect between the finer particles (inter-particular forces), which causes these finer particles to fill the voids left by the larger particles and the agglomeration effect is less prevalent. In addition, the authors observed how both aggregate types were sensitive to the compaction method and, specifically, the fine fractions, due to the above-mentioned agglomeration effect.

3.3. Assessment of particle packing models: experimental vs predicted

EXPERIMENTAL PACKING DENSITY OF MIXING AGGREGATES

Two different approaches were analyzed close to the maximum packing value scenario. Firstly, a dosage with the highest packing density computed with a 2^{nd} degree-polynomial curve in the packing model: 40%(11/22)+60%(4/11) of NA aggregates and 20%(11/22)+80%(4/11) of ES. Secondly, an optimum aggregate packaging proportion for concrete mixes was designed, because coarse aggregates provide the concrete strength and reduced aggregate surface areas will also reduce the water requirement. In this second approach, the desired workability was obtained with a combination of: 60%(11/22)+40%(4/11) of natural aggregates and 40%(11/22)+60%(4/11) of ES aggregates. The packing density of ES aggregates was maximized when the 4/11 fraction was dominant and, as expected, the higher the packing density, the higher the compaction energy.

It should be highlighted that the aggregate proportions with the maximum packing value were independent of the applied compaction method. The fine fractions had a compaction sensitivity greater than all others. Additionally, a maximum packing density will require siderurgical mixes with fewer fillers (ES 0/5 plus NA 0/2) when the coarse aggregate contents are in lower proportions.

OPTIMUM MIX DESIGN BASED ON IDEAL CURVES

The comparison between Funk/Dinger and Fuller methods showed that the former required higher amounts of fines and that when the Fuller method was applied, the crushed aggregates were in preferable proportions. Besides, the Fuller method provided preferable NA proportioning of 50%(11/22)+50%(4/11), while, in the Funk/Dinger procedure, the preferred proportions were 40%(11/22)+60%(4/11).

In the present research, the medium aggregate fractions (40%(11/22)+60%(4/11)) dominated the experimental ES combinations with maximum packing densities. Although, when fitted with the ideal curves, the prediction was 60%(11/22)+40%(4/11). In conclusion, the ideal curves for the siderurgical (crushed) aggregates were not in agreement with their experimental counterparts.

ASSESSMENT OF DISCRETE MODELS FOR AGGREGATE PACKING DENSITY

The predictions of the CPM and 3-PM models and their applicability were verified and compared with the experimental packing densities throughout the 4-compaction procedures (L, C, C26 and C33): binary, ternary and quaternary mixtures, respectively. Firstly, we will present the results of the NA-based mixtures.

It was concluded that, in both the binary and the ternary mixtures, the CPM overestimated the packing density and its prediction of the most compacted combinations reached maximum values of aggregate ratios close to 50%(11/22)+50%(4/11). In contrast, the 3-PM model fitted the experimental results of the ternary mixtures better, especially in two compacting methods: C and C26. The 3-PM model appeared to be suitable for predicting the packing density of these natural aggregates, regardless of the compaction method, taking into account the mean diameter in each aggregate fraction.

The results of the siderurgical aggregate mixes are now presented. In ES binary mixtures, the CPM model (coarse and fine sizes dominant) appears valid. On the other hand, the predictions of this model overestimated the packing density when there was no dominant aggregate fraction. However, the 3-PM fit for the intermediate mixes was better, especially for the two compacting methods: C26 and C33.

In the ternary ES mixtures, the CPM model fitted the experimental values and the 3-PM model clearly showed a better fit when C and C26 compacting methods were applied. Besides, CPM overestimated the packing density in the quaternary mixtures (added NA 0/2 fines), although it was underestimated in

the 3-PM model. These behaviors could be based on how the 3-PM model accounts for the welding effect between particles or because of the reduction of the packing density when the fine particle layers cannot be formed (gaps between coarse particles).

Model accuracy for ES materials based on the predicted CPM (binary mix) packing densities deviated by around 6%. Furthermore, this error increased when ternary and quaternary mixtures were assessed (overestimation). On the contrary, the 3-PM model fitted a better agreement, showing itself to be a suitable method for predicting the packing density of ES materials.

As an additional study, the CPM and 3-PM models were analyzed to validate aggregate proportioning at the highest packing density (see Table 3). The table below presents 3 results: a wide range of aggregate combinations were close to the maximum packing density when mixing three or more granular fractions, and close enough therefore to the maximum experimental values.

Compacting	Model	Natural aggregates (NA)				Siderurgical aggregates (ES)					
		0/4	4/11	11/22	PD	NA-0/2	0/5	4/11	11/22	PD	
L	CPM	0.4	0.3	0.3	0.68	0.5	0.2	0.2	0.2	0.75	
	3P				0.69	0.4	0.3	0.2	0.1	0.66	
С	CPM	0.5	0.3	03	0.76	0.5	0.2	0.2	0.2	0.84	
	3P				0.75	0.6	0.2	0.1	0.0	0.72	
C26	CPM	0.4	0.2	0.4	0.83	0.5	0.2	0.2	0.2	0.91	
	3P	0.5	0.3	0.3	0.80	0.4	0.5	0.1	0.0	0.76	
C33	CPM	0.4	0.2	0.4	0.84	0.5	0.2	0.2	0.2	0.92	
	3P	0.5	0.3	0.3	0.81	0.4	0.4	0.2	0.0	0.78	

Table 3: Optimal granular (in volume) proportioning and maximum Packing Densities (PD).

As can be seen in Table 3, there are differences in aggregate proportioning depending on the compacting procedures. The 3-PM model predictions for the ES mixtures implied that the denser the mix the lower the content of coarse aggregates (11/22), which suggests that the model is not useful for concrete applications. Moreover, taking into account the same criteria in the experimental campaign, to identify the optimum packed aggregate proportioning for concrete mixes, the optimal aggregate proportioning will be: 0.17(11/22):0.25(4/11):0.28(0/5):0.30(0/2-NA), compacted with the C33 method; proportions that were very close to the predictions.

4. CONCLUSIONS

Numerous experimental test campaigns have been conducted, in which 4 compaction procedures (dry conditions) have been applied to four sieved fractions of natural limestone and three others from siderurgical aggregates. Different physical and chemical properties of these aggregates have been characterized. Different aggregate proportioning studies have been compared and the experimental results validated with four theoretical models (Fuller, Funk and Dinger, CPM and 3-PM).

Regardless of the compaction method, the 3-PM model showed higher accuracy for the ternary and the quaternary mixtures when determining the PD of individual fractions. Our conclusion is that, unlike natural aggregates, other methods based on aggregate shape should be preferred for these ES aggregates mixtures, due to the variability of these co-products, their specific textures and contours.

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