NOVEL MICRO-GRANULATES FOR PORCELAIN STONEWARE TILES: PRELIMINARY DATA ON POWDER RHEOLOGY AND COMPACTION

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1. ABSTRACT

Using spray-dried bodies for feeding tile production plants is nowadays widely consolidated. The industrial production is moving towards increasingly larger sizes, especially for porcelain stoneware tiles and slabs. This trend implies a demand of high-quality ceramic powders, performing appropriately all along the production process, as achievable by spray-dried bodies. However, spray-drying requires a significant consumption of both water and energy. An interesting alternative appears to be the dry granulation process, because of remarkable water and energy savings. In the past decades, it has always been penalized by worse rheology and compaction behavior, with respect to spray-dried powders, which was reflected in the microstructure and characteristics of the fired products. Recently, a novel micro-granulation hybrid process has been developed for porcelain stoneware bodies. It is based on wetting-agglomeration-drying operations that are able to provide granules with technological characteristics adjustable on request and behavior apparently close to that of spray-dried powders. The aim of our work is the evaluation of these novel micro-granulates in the industrial production of porcelain stoneware tiles and large slabs. A comparison is made with conventional technologies used for dry-granulated and spray-dried powders. For this purpose, dry micro-granulates manufactured on the Migratech 4.0 pilot line were investigated for their characteristics (grain size, shape and moisture distribution), flowability (static and dynamic angles of repose, poured and tapped density, mass flow rate, Hausner ratio) and compaction behavior (bulk density of green and dried bodies for increasing applied load, green and dry modulus of rupture, microstructure by optical and scanning electron microscopy, pore size distribution by mercury intrusion porosimetry). The new dry micro-granulates exhibit improved rheological and compaction performances, approaching those of spray-dried bodies. Some differences in compaction behavior are discussed in relation to the distinct microstructures and textural properties of powders obtained by spray-drying and dry micro-granulation, respectively.

2. INTRODUCTION

Water and energy saving in the production of porcelain stoneware tiles is drawing increasing worldwide interest, due to a major attention globally given to the environmental impact of industrial production [1]. In tile manufacturing, the direction followed is the reduction of both energy and water consumption, while minimizing wastes, and a major option would be avoiding the energy intensive and water-wasting spray-drying step [2].

However, tile production is nowadays trending to increased sizes, including slabs up to 8 square meters, which require high quality feeding powders. Proper characteristics are undoubtedly provided by spray-dried powders, whereas dry granulates suffered in the past from worse rheological properties and compaction behavior. Nevertheless, the dry route is an intrinsically water-saving process, offering energy sparing, hence a subject of research and development over the last decades [2-4]

Granulation technology	Type	Milling	Granulation steps	Data source
Spray-drying	WET	WET	single: simultaneous drying and agglomeration	[5-6]
Hybrid granulator (*)	HYBRID	DRY-WET	triple: mixing/agglomeration + crushing + drying	This work, and [7]
Droplet powder granulator	HYBRID	DRY-WET	double: simultaneous drying and agglomeration + mixing	[8-9]
Sinusoidal granulator (*)	DRY	DRY	double: agglomeration + drying	This work, and [3]
Intensive mixer granulator	DRY	DRY	single: agglomeration	[10-14]
Vertical axis turbine	DRY	DRY	single: moistening	[3]

(*) integrated in the Migratech 4.0 technology.

Table 1. *Granulation technologies used in ceramic tile manufacturing and materials under examination.*

Different granulation technologies are utilized in tile-making, beyond the overwhelming wet route with spray-drying, which span from a fully dry route to hybrid solutions (i.e., wet + dry). The dry processes encompass traditional moistening (by vertical axis turbine) and the sinusoidal granulator (and fluid bed drier) – which are the most popular in the ceramic tile industry – as well as the intensive mixer granulator (Table 1). The hybrid route, including the use of droplet powder granulator (Fig. 1), has been recently improved through a set of plant and technological solutions (Migratech 4.0©, LB Officine Meccaniche patent: PCT/IB2017/055856). The novel, adjustable, triple step micro-granulation process that makes use of both wet milled slip and dry milled powders (Table 1) allows a range of technological solutions and powders to be obtained, namely hybrid and dry granulates.

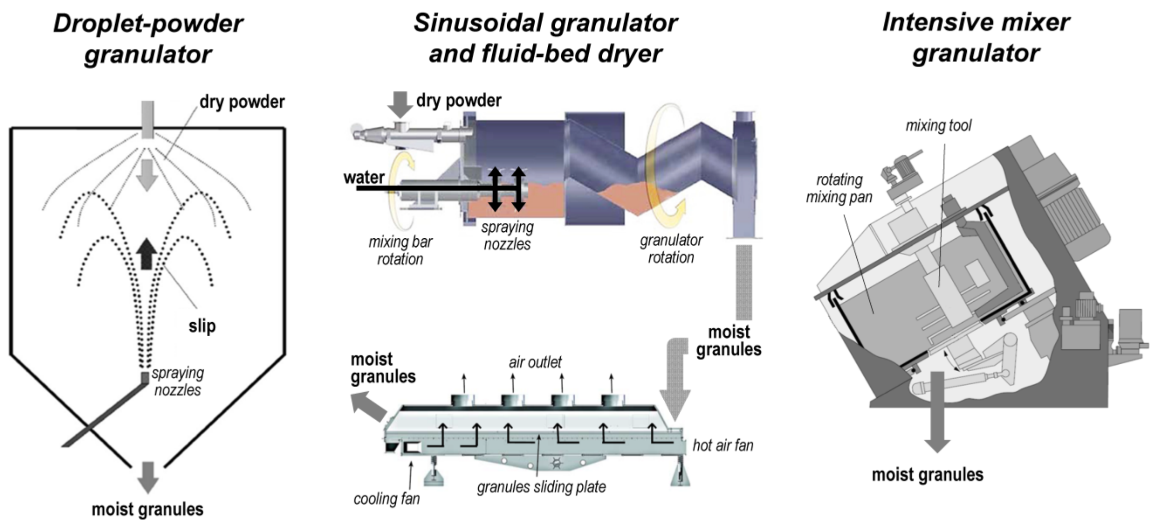

Figure 1. *Alternative powder granulation systems to spray-drying.*

In the literature on powders used in tile-making, a detailed characterization is available for spray-dried bodies [5-6] and dry granulates obtained by an intensive mixer granulator [10-14]. Just few data were published for a droplet powder granulator [3-4] or sinusoidal granulator and vertical axis turbine [3]. A snapshot of the micro-granulates manufactured by the Migratech 4.0 technology is given by Cavani and Battaglioli [7].

To the best of our knowledge, only few papers compared dry granulates with spray-dried powders [9, 12, 14].

The goal of the present work is the preliminary evaluation of characteristics, rheological properties and compaction behavior of powders prepared by the innovative micro-granulation process Migratech 4.0, both hybrid and dry routes, compared with a large set of granulates, industrially made with conventional technologies. In particular, our emphasis will address the novel hybrid route.

3. EXPERIMENTAL

Materials

Industrially manufactured granulates were taken into account (porcelain stoneware and stoneware bodies). Both Migratech 4.0 micro-granulation processes were sampled: one by the hybrid route and five by a sinusoidal granulator with fluid-bed drying. Data for powders obtained by spray-drying, droplet powder granulation and intensive mixer granulation are from the literature (Table 1). Samples were characterized as received.

Methods

The powder characteristics determined are: agglomerate size distribution by dry sieving (meshes: 2, 1, 0.63, 0.5, 0.4, 0.315, 0.2, 0.1 mm); granule morphology (optical microscopy on sieve fractions); moisture distribution as a function of agglomerate size (weight difference of wet and dry fractions for each sieve).

The rheological behavior and flowability were appraised by measuring static and dynamic angles of repose, mass flow rate through a Ford cup, and poured and tapped density (Hausner ratio HR). Full details on the experimental procedures are in Soldati et al. [5].

The compaction behavior was investigated by pressing with increasing applied load (up to 60 MPa) and determining, on the resulting green compacts, bulk density (geometrical method) and microstructure (optical microscopy). Full details on the experimental procedures are in Soldati et al. [6].

4. RESULTS AND DISCUSSION

Powder characteristics

As far as the particle size and moisture distributions are concerned, the data collected for sinusoidal granulates and the new hybrid granulate are reported in Table 2 and depicted, in comparison with the range of spray-dried powders, in Figure 2.

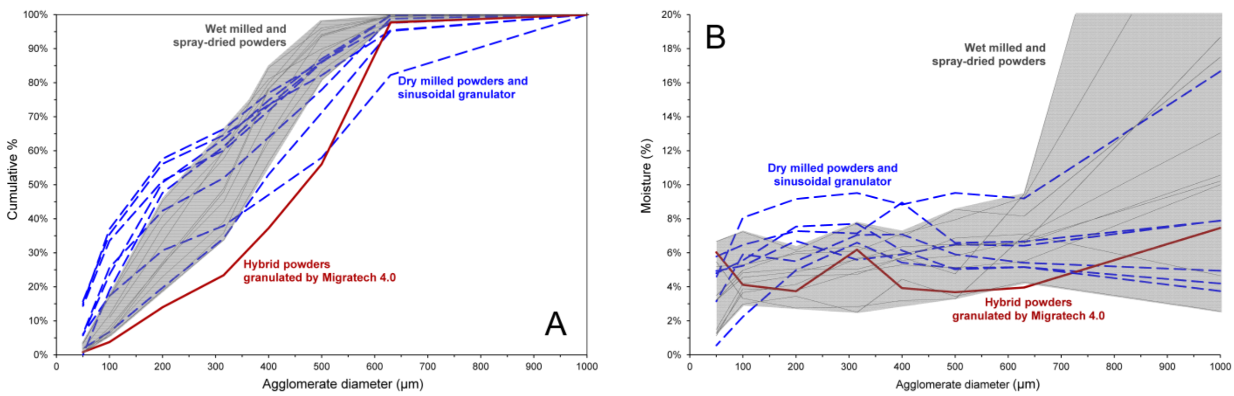

Figure 2. *Agglomerate size distribution (A) and moisture distribution (B).*

Two sets of behaviors can be immediately noticed, with clear relations to spray-dried or dry granulated samples. The spray-dried bodies are characterized by lower contents of the finest fractions, and by steeper trends while reaching the bigger granule sizes [5]. In contrast, the dry granulated powders often show more gradual growths, thus more homogenous percentage contribution of the different sizes to the global composition.

The sample chosen as reference for the micro-granulation hybrid process presents an interesting versatility of the technology: this sample has reached a trend as steep as for spray-dried bodies, but a much lower fine fraction content. Thanks to adjustable mixing time, starting moisture content, Migratech 4.0 technology allows production of granulates with a wide range of properties, such as aggregate size and moisture distribution similar to those of spray-dried powders. Moreover, the hybrid granulate shows a relatively regular moisture content, comparable with dry granulated powders.

Granulation technology >		HYBRID	HYBRID	DRY	DRY	WET
Property of agglomerates	Unit	Hybrid granulator (*)	Droplet poder granulator	Sinusoidal granulator l (*)	Intensive mixer granulator	Spray-drier
Median diameter	μm	475	240-270	160-430	450-470	225-375
Powder moisture	% wt.	4.3	5.5-6.0	5.1-7.5	5.0	3.4-7.2
Static angle of repose	°	33.7	NA	30-39	NA	30-35
Dynamic angle of repose	°	45.9	NA	53-69	NA	40-45
Mass flow rate	$g \cdot cm^{-2} \cdot s^{-1}$	13.3	NA	11-13	NA	13-15
Poured density	$g \cdot cm^{-3}$	1.03	1.08-1.13	0.93-1.05	1.12-1.17	0.92-1.05
Tap density	$g \cdot cm^{-3}$	1.22	1.28-1.32	1.12-1.33	1.35-1.42	1.03-1.12
Hausner ratio	1	1.19	1.13-1.21	1.16-1.29	1.15-1.27	1.07-1.17

(*) integrated in the Migratech 4.0 technology.

Table 2. *Powder characteristics and rheological properties of different types of granulates (porcelain stoneware and stoneware bodies).*

The shape distribution of the powder obtained by the novel hybrid technology is compared, in Figure 3, with a spray-dried body and a dry granulate manufactured by a sinusoidal granulator. Spray-drying ideally provides spheroidal granules with an inner cavity [5, 8, 14]. However, while the size increase, a plurality of irregular aggregates appears and became, in percentage, more relevant and finally exclusive [5]. In contrast, dry granulates do not exhibit any significant change in shape in function of size. Nevertheless, they have a more irregular shape and a rough and bumpy surface, which influence their rheology behavior. The hybrid granulate pictures show a rounded shape and a smoother surface in comparison to powders prepared by dry technology, which can account for some of the property improvements.

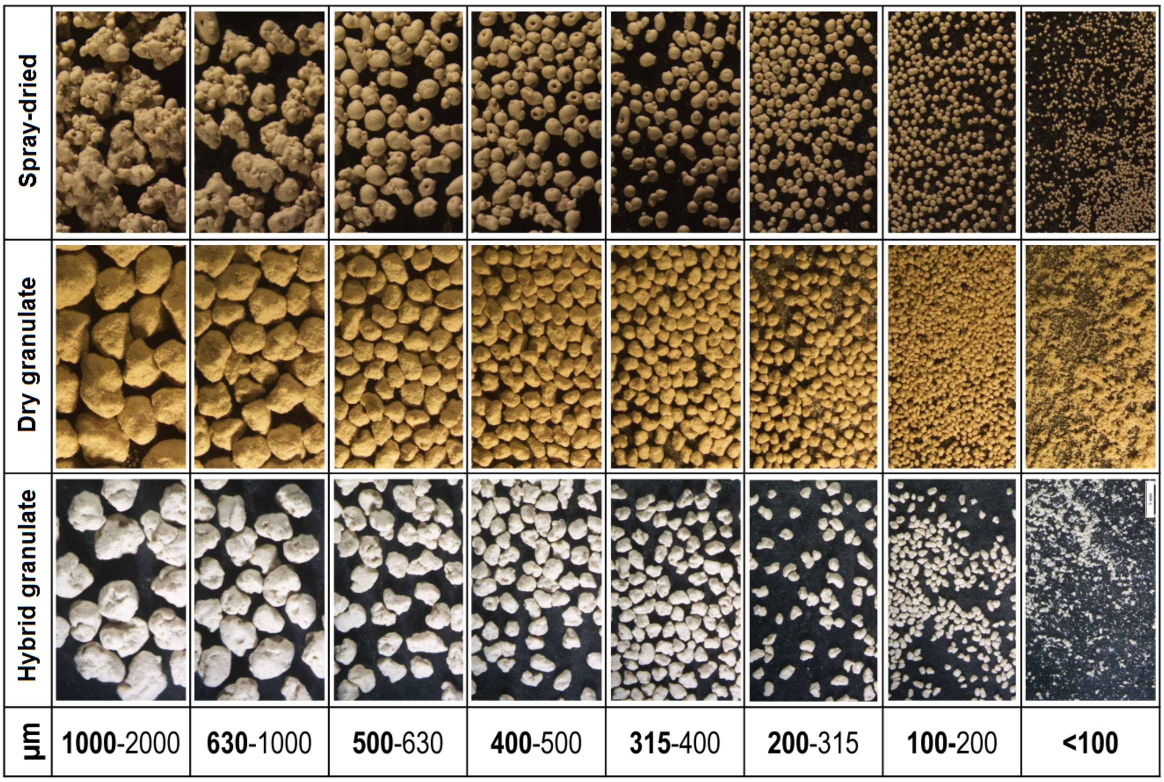

Figure 3. *Agglomerate shape distribution.*

Powder rheology

Preliminary data on the rheological properties of novel hybrid granulates – such as static and dynamic angles of repose, mass flow rates, poured and tap density, and Hausner ratio – are compared with conventional dry granulates and spray-dried powders (Table 2). The results obtained show interesting similarities with spray-dried bodies, as evidenced in Figure 4.

While the ranges of static angle of repose (SAR) are close one to each other, data about the dynamic angle of repose (DAR) show important differences. Interestingly, the hybrid granulates approach values typical for spray-dried powders. A similar observation can be made about the mass flow rate measured through a Ford cup: the typical flowability of spray-dried powders cannot be achieved by dry granulates. However, the hybrid sample has a flow rate included in the range of spray-dried bodies. In contrast, both poured and tap densities for hybrid and dry granulates are always higher – or in the highest part of the range – with respect to powders collected from spray-driers. Unlike dry granulation, the novel hybrid technology appears to be able to produce granulates with Hausner ratio as low as that achievable by spray-drying, thus involving a free-flowing powder (intended for HR<1.25).

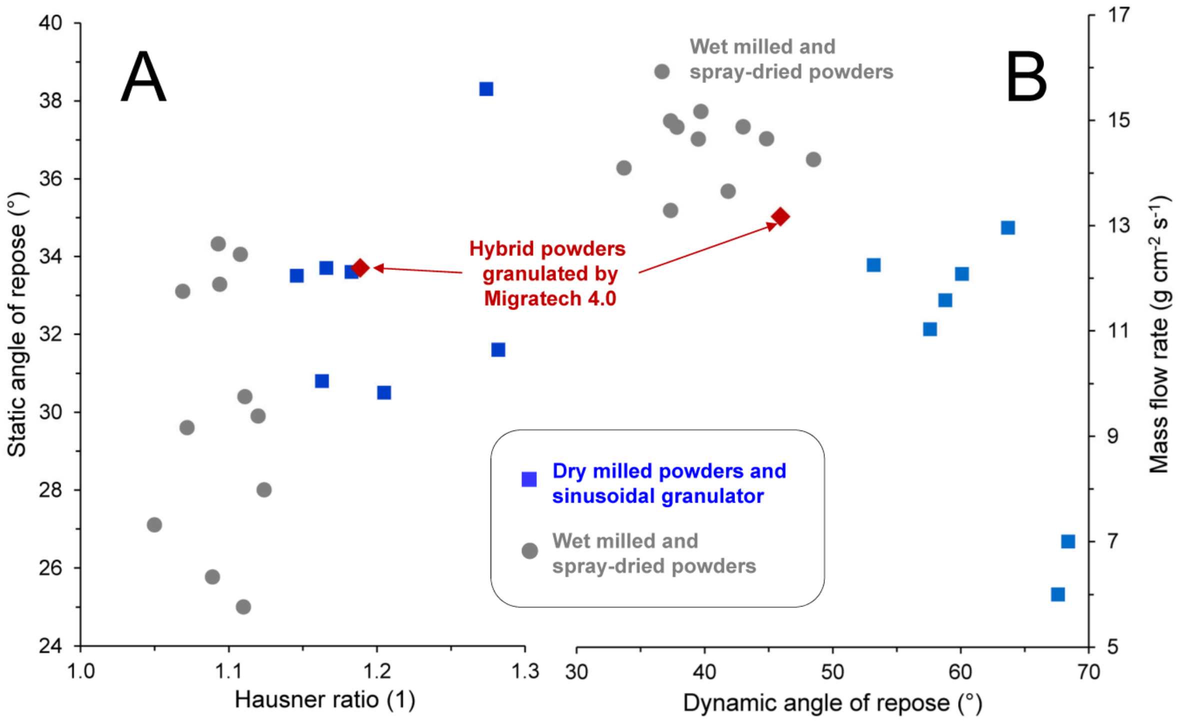

Figure 4. *Comparison of powder flowability of wet, dry or hybrid granulates: static angle of repose vs. Hausner ratio (A); mass flow rate vs. dynamic angle of repose (B).*

Analogous observations can be made on analyzing the relations between SAR and HR, as well as mass flow rate as a function of DAR. In the graph of SAR versus HR, the spray-dried bodies fall on the left side, with HR values below 1.13, while the sample obtained by hybrid technology falls amongst dry granulates, although not so far from the spray-dried bodies area (Fig. 4A). Still, by contrasting mass flow rate versus DAR, the same hybrid granulate plots very close to, if not among, data of spray-dried bodies (Fig. 4B).

Powder compaction

The surface microstructure of green compacts, as pressed at increasing applied loads, was verified by optical microscopy for both hybrid granulates and spray-dried bodies. This comparison is reported in Figure 5: it clearly shows how fast the hybrid granules lose their individuality, even at very low pressure. In fact, the granule contour is hardly appreciable already for a load of ~5 MPa (50 kg/cm^2). At variance, the contour of spray-dried granules can be still perceived at ~15 MPa (150 kg/cm^2). This is a clue of a lower stiffness of hybrid granules, confirming the higher yield strength and fragility that was observed in spray-dried bodies [6, 10-12, 15].

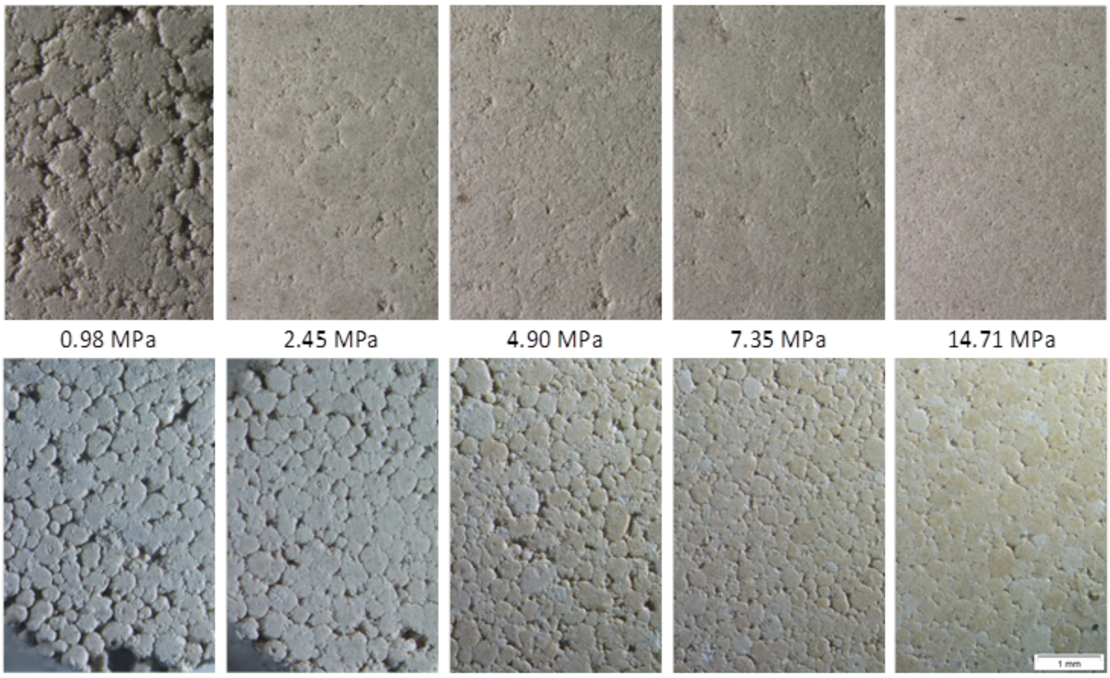

Figure 5. *Microstructure of hybrid micro-granulates (top) and spray-dried powders (bottom) compacted at increasing applied load. Scale bar = 1 mm.*

As a matter of fact, the bulk density of green tiles is always higher when using dry granulates in comparison with spray-dried powders [9, 12-14]. This fact is confirmed in Figure 6, where the bulk density curves of dry granulates and spray-dried bodies are reported versus the applied pressure. This tendency can be partly explained by the collapse of the inner cavity of spray-dried granules, even though it can never be completely closed while pressing [6]. However, it surely derives also from the different bulk density of the starting powders and their poured density in the mold, thus making any hypothesis complex on the mechanisms responsible for the different microstructures and compaction curves.

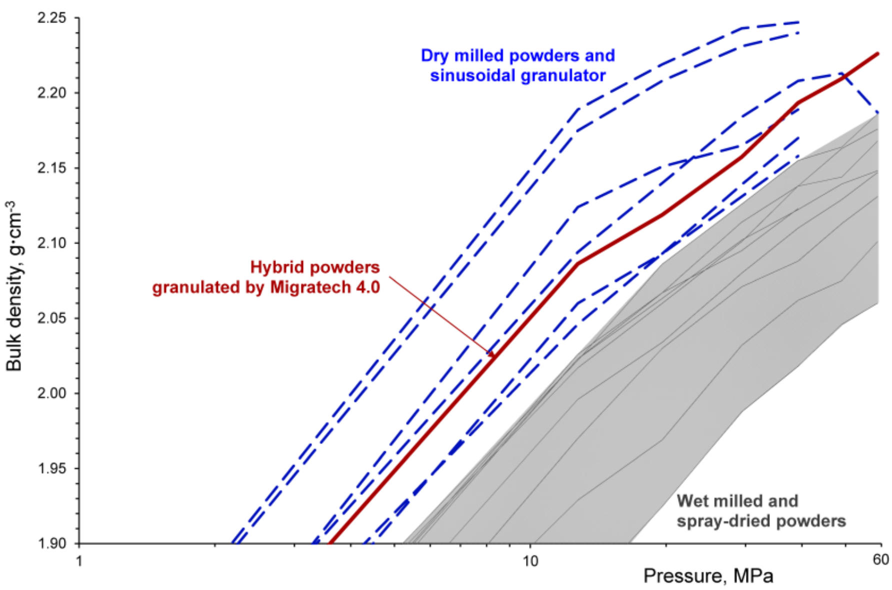

Figure 6. *Compaction curve of the hybrid micro-granulates compared with spray-dried and dry granulated powders.*

5. CONCLUSIONS

A novel microgranulation process that operates through an innovative hybrid adjustable (wet + dry) route – within the Migratech 4.0 technological framework – allows obtainment of powders suitable for the production of porcelain stoneware tiles. The target constituted by the improvement of technological characteristics of powders has been achieved, as hybrid granulates can approach the rheological and compaction properties of spray-dried powders. These positive results – although based on preliminary data – confirm what was already suggested in previous studies [9, 10]: the hybrid route is the way to overcome dry granulation constraints and undoubtedly represents a step forward in terms of powder properties.

In addition, this new technology exhibits a certain versatility, through a triple step process with settable technological parameters, which apparently allows quite a wide range of powder properties to be achieved. This aspect, and the chance of fine tuning of properties by adjustment oriented to production needs, will be the object of further investigation.

6. REFERENCES

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