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A new cleaner process to prepare pressing-powder

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An alternative cleaner process of pressing-powder preparation, based on filter-pressing and a novel granulation method, is presented to substitute the existing spray-drying process. In the new process, about two-thirds of wet-milled slurry is filter-pressed, dried and milled into dry fine powder. The other one-third of the slurry and the as-obtained dry fine powder are spray-mixed in a tower, where the slurry droplets adsorb the dry powder to form granules which are then rolled and dried into a useable pressing-powder for tile pressing.
The key stages, filter-pressing and granulation (consisting of spray-mixing and rolling treatment), are specially studied. The pressing-powder properties and pressing/firing behavior, and, energy/water consumption and pollution emission data are presented, and a comparison between the new process and the existing spray-drying process is made. This new process has been found to be feasible and provides a pressing-powder with suitable properties, together with lower energy/water consumption and pollution emission (particulate matter and CO_2).

Keywords: processing; powder; microstructure; porcelain tiles

Un nuevo proceso más sostenible para preparar polvo de prensas

En ese trabajo se presenta un proceso alternativo y más ecológico para la preparación de polvo de prensas por molienda vía húmeda de las materias primas. En este proceso la eliminación del agua de la suspensión obtenida en la etapa de molienda, en vez de realizarse por secado por atomización, se lleva a cabo en dos etapas, en una primera etapa dos tercios de esta suspensión se filtro-prensan, posteriormente se secan y molturan hasta obtener un polvo seco micronizado. Este material seco se introduce por la parte superior de una torre granuladora, en la que se pulveriza el tercio de la suspensión restante por la parte inferior, de forma que las gotas adsorben sobre su superficie las partículas secas formando gránulos, que posteriormente se compactan por rodamiento (“rolling”), y finalmente se secan hasta la humedad requerida para el prensado. En este trabajo se estudian con detalle las dos etapas clave del nuevo proceso: el filtro prensado y la granulación (mezclado por pulverización y “rolling”). Además, se describen las propiedades del polvo de prensas obtenido, el comportamiento en crudo y en la fase de cocción. Los resultados indican que el nuevo proceso desarrollado, aunque es más complejo técnicamente, es viable y permite obtener un polvo de prensas con características adecuadas para obtener baldosas de gres porcelánico, aunque requiere mayores temperaturas de cocción. El análisis del proceso indica que conlleva menores emisiones contaminantes (partículas y CO_2) que el proceso de atomización tradicional, así como consumos inferiores de agua y de energía.

Palabras clave: procesado; polvo de prensas; microestructura; gres porcelánico

1. INTRODUCTION

In the ceramic wall and floor tile industry, generally, raw materials are prepared into pressing-powder, which is pressed into green compacts and then fired into ceramic tiles. The pressing-powder properties have significant effects on the quality of the resulting green and fired tiles. In the past four decades, the wet process, consisting of raw material wet milling and spray-drying, has been employed to produce pressing-powder worldwide, benefiting from the technical advantage of raw material wet milling and the favorable property of the spray-dried powder. However, the high energy and water consumption in the spray-drying stage has always been a prominent issue, and the pollution emission from spray-drying also demands the use of a high-efficient cleaning system [1]. The application of cogeneration system contributes significantly to optimize the energy efficiency of pressing-powder preparation [2]; however, the inherent energy consumption of spray-drying in itself is still not changed.

Therefore, it is worthwhile exploring a new process of pressing-powder preparation that is inherently low in energy/water consumption and pollution emission. An alternative process (i.e. droplet-powder granulation process, hereafter DPGP) was recently proposed [3], consisting of raw material wet milling and a cleaner dewatering and granulation stage.

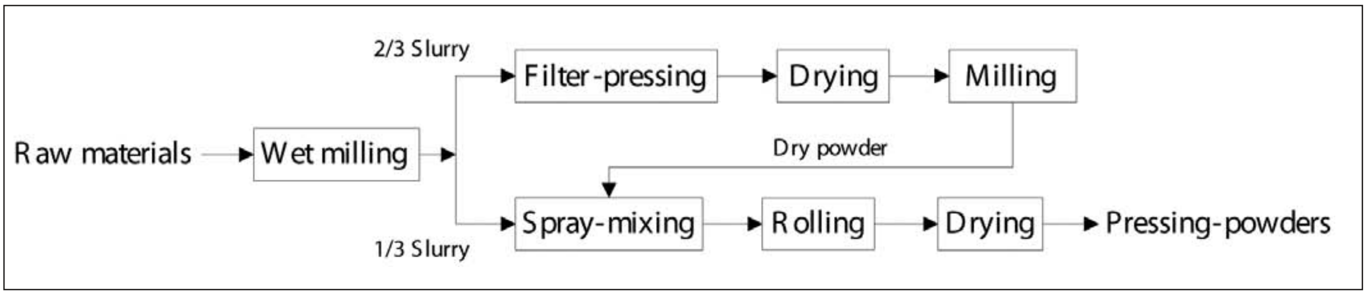

Fig.1. Flowchart of the new process to prepare pressing-powder.

In the new process, a high proportion of the water in wet-milled slurry is removed by filter-pressing and then recycled; subsequently, a novel granulation method is employed to develop pressing-powder.

Thus, this work presents the new process with the focus on these two key steps, i.e. filter-pressing and DPGP granulation. The approach of improving the filter-pressing efficiency is proposed and the mechanism of the DPGP granulation process is shown. The property of as-prepared pressing-powder and its performance in pressing and firing are also studied, along with the evaluation of energy/water consumption and pollution emission of the new process, in comparison with the spray-drying process (hereafter SD).

2. MATERIALS AND METHODS

2.1 Raw materials

A porcelain tile composition was used, consisting of 35 wt% ball clay, 55 wt% sodium feldspar and 10 wt% quartz. Ball clay was dried and slightly milled by hammer mill (5.02 wt% residue on the 63 μm sieve). Finely milled sodium feldspar (0.82 wt% residue on the 63 μm sieve) and quartz (9.80 wt% residue on the 63 μm sieve) were used. Table 1 shows the chemical and mineralogical analysis of raw materials and porcelain tile composition.

2.2 The new process of pressing-powder preparation

The new process of pressing-powder preparation is schematically shown in Fig. 1 and detailed below.

2.2.1 STAGE 1: PREPARING SLURRY BY “WET MILLING”

This stage is equal to the wet milling stage in the traditional wet process, obtaining deflocculated slurry.

In this work, pre-milled raw materials were used and thus they were directly wet-mixed into slurry. Batch-weighed raw materials were mixed in water with 0.2 wt% deflocculant (on the solid basis, sodium metasilicate and sodium tripolyphosphate in a 3:1 ratio by weight) to prepare slurry with a water content of 0.47 kg water/kg solid and a viscosity of 650 centipoise

Table 1. Chemical and mineralogical analysis of raw materials and porcelain tile composition.

		Raw material (wt%)			Porcelain tile composition (wt%)
		Ball clay	Sodium feldspar	Quartz	
Chemical analysis	SiO_2	64.30	69.50	91.00	69.83
	Al_2O_3	23.50	18.00	5.00	18.63
	Fe_2O_3	0.96	0.14	0.12	0.43
	TiO2	1.35	0.28	0.08	0.63
	CaO	0.20	0.50	0.10	0.36
	MgO	0.50	0.20	0.01	0.29
	Na_2O	0.50	10.00	0.10	5.69
	K_2O	2.10	0.80	2.50	1.43
	LOI	6.31	0.50	1.06	2.59
	Others	0.28	0.08	0.03	0.15
	Total	100.00	100.00	100.00	100.00
Mineralogical analysis	Illite	24	0	15	10
	Kaolinite	38	2	6	15
	Quartz	37	10	79	26
	Albite	0	87	0	48
	Others	1	1	0	1
	Total	100	100	100	100

rested after 1 min. The contents of deflocculant and water were determined by a viscosity curve of between 500 and 1000 centipoise, using a Gallenkamp viscosimeter.

2.2.2 STAGE 2: PREPARING DRY POWDER BY "FILTER-PRESSING", "DRYING" AND "MILLING"

The main aim in this stage of the process is to change about two-thirds slurry into dry fine powder, which will be used in a subsequent granulation stage. For this purpose, slurry is firstly filter-pressed into press-cake, then dried and finally milled into fine powder.

In the above operation, a cylinder hydraulic filter press with a 15.8 mm diameter is used. In each trial, a constant weight (800 g) of slurry was filter-pressed for 90 min. While filter-pressing, the weight of filtrate was determined in real time, and the weight ratio between residue-water and total solid could be calculated as the real-time water content of press-cake. The filtrate pH value after filter-pressing was determined using a pH meter. In order to improve the filter-pressing efficiency, pure acetic acid was used as flocculant. The acid dosage and filter-pressing pressure were varied to study their effects on filter-pressing efficiency. The optimal acid dosage and filter-pressing pressure was chosen and used in subsequent experiments.

Press-cake was oven-dried into a water content of 0.02 kg water/kg solid and then milled into a dry fine powder by a hammer mill with a 200-micron sieve.

2.2.3 STAGE 3: PREPARING GRANULES, I.E. GRANULATION, BY "SPRAY-MIXING", "ROLLING" AND "DRYING"

The goal of this stage is to transform the remaining one-third slurry and the as-obtained dry fine powder together into granules (i.e. pressing-powder) suitable for tile pressing. Firstly, the slurry is sprayed into droplets and the dry powder is also sprayed into a dispersed state inside a spray-mixing tower, where the slurry droplets adsorb the dispersed dry powder on their surface to form granules. Then, the raw granules undergo the rolling and drying treatments to become a useable pressing-powder for tile pressing.

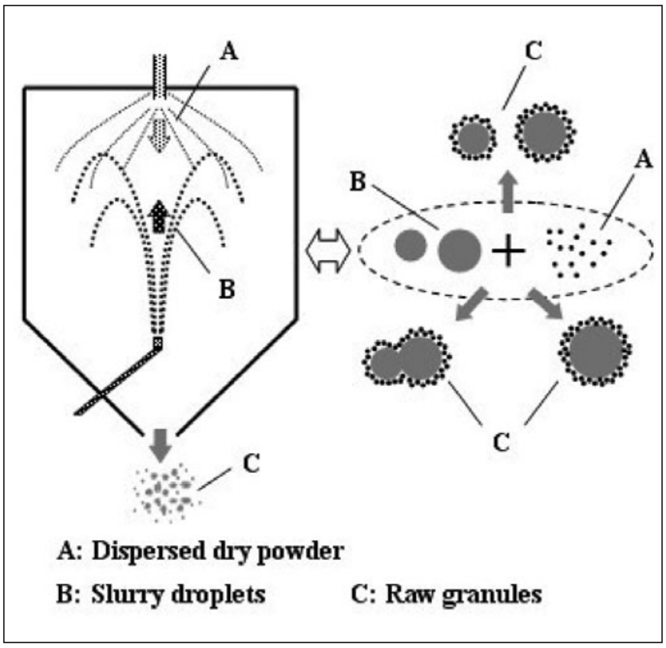

Fig. 2. Granulation principle of spray-mixing.

In this operation, a pilot spray dryer excluding heating system was used as the tower for spray-mixing. As schematically shown in Fig. 2, inside the tower, the slurry was upwards sprayed into droplets from the bottom and the dry powder was downwards sprayed into a dispersed state from the top. Then, the droplets adsorbed the dispersed dry powder to form granules which fell down and went out. It is assumed that a single droplet may adsorb dry powder to form a single spherical granule, and, several droplets may also combine together and adsorb dry powder to form a single granule with larger size or with more irregular shape, and, some dry powder may exist independently without being adsorbed by droplets as well. Thus, the granules obtained in spray-mixing may be considered as a mixture of diversiform granules and dry powder.

The as-obtained raw granules were placed into an empty laboratorial planet mill (without grinding media) to undergo a rolling treatment for 0.5 min. The water content of raw granules, i.e. the weight ratio between sprayed slurry and dry powder, was varied to study its effect on the granulation. The optimal water content was chosen and used for subsequent experiments.

The rolled granules were subsequently dried into a water content of 0.055 kg water/kg solid and became the DPGP pressing-powder.

In addition, the same deflocculated slurry was spray-dried into the SD pressing-powder, which was used as a comparison.

2.3 Characterization of pressing-powder

Pressing-powders were adjusted to a water content of 0.055 kg water/kg solid and stored for 24 h. Pressing-powder was free-fall filled into a measuring cylinder, and then the cylinder was gently tapped until a constant volume was obtained. The powder weight and volume before and after tapping was measured to respectively calculate free-fall bulk density and tapped bulk density, the ratio between former and latter (Hausner index) [4] being calculated to evaluate the powder flowability. Pressing-powder was also placed to flow through a timer-equipped stainless funnel with inner dimensions of 155 mm dia. (at top) × 10 mm dia. (at bottom) × 150 mm height, and the volume flow velocity was calculated to evaluate the flowability in a different way. For each type of pressing-powders, 50 g samples were dried at 110 °C for 2 h and screened through grading sieves. Each screen residue was weighed to evaluate the granule size distribution of pressing-powders. Granule morphology was observed by a digital optical microscope, and intragranular microstructure was determined by scanning electron microscope (SEM) on polished cross-sections of granules mounted in epoxy.

2.4 Pressing and firing

A uniaxial press with a cylinder die of 40 mm diameter was used to press disk specimens. Specimens were respectively pressed at 15, 25 and 40 MPa from each type of pressing-powders and dried at 110 °C to constant weight. Specimen bulk density was then determined to reveal the relationship between bulk density and pressing pressure, i.e. powder pressing behavior. Then, pressing-powders were pressed at 32.5 MPa, dried at 110 °C to constant weight and fired in a laboratory electric kiln at different peak temperatures, ranging from 1180 to 1240 °C. The heating rate was 25 °C/min with a hold of 6 min at the peak temperature. For fired specimens, bulk density was determined by the mercury displacement, and water absorption was determined according to standard UNE-EN ISO 10545-3. The diameters of the specimens before and after firing are measured, and the diameter difference divided by the diameter before firing is calculated to evaluate the linear shrinkage of specimens during firing.

3. RESULTS AND DISCUSSION

3.1 Filter-pressing

Starting from wet-milled slurry (0.47 kg water/kg solid in this work) and ending at pressing-powder (0.055 kg water/kg solid in this work), the DPGP process needs to remove the same quantity of water as the traditional wet process. However, instead of spray-drying in wet process, other three dewatering steps (i.e. slurry filter-pressing, pressed-cake drying and granules drying) are employed in the DPGP process (Fig. 1). Filter-pressing, a high-energy-efficient mechanical technique [5,6], is expected to remove and recycle the water as much as possible, in order to reduce the energy and water consumption of the whole DPGP process.

3.1.1 EFFECT OF FLOCCULANT CONTENT ON FILTER-PRESSING EFFICIENCY

Fig. 3 plots the water content of pressed-cakes versus the filter-pressing time, filter-pressed at 20.31 kg/cm^2 from slurries with different flocculant contents. It shows that, each pressed-cake water content decreases along the filter-pressing time with a decreasing decrement (i.e. gradually flattening of the curve slope), which results from the thickness increase of dewatered layer that is compacter and less permeable than the initial un-dewatered layer in press-cake [7].

In order to analyze the effect of flocculant content on filter-pressing efficiency better, the water content of pressed-cakes after 90 min is detailed in Fig. 4. It shows that, the water content of pressed-cakes from flocculated slurries are much lower than that from the initial deflocculated slurry (without flocculant), indicating that flocculating the slurry improves the filter-pressing efficiency.

It is well documented [8,9] that deflocculated slurry has a microstructure of particles uniformly dispersing in water, which provides the slurry with a low viscosity in favor of wet milling and piping. However, this microstructure is not favorable for filter-pressing. Since particles are independently dispersed, the paths for water transferring inside the slurry are the inter-particle space, resembling numerous fine tubes with high specific surface area. Thus, the total friction between water and particle is high, resulting in low press-cake permeability and, therefore, low filter-pressing efficiency.

After flocculating the slurry with a small amount of flocculant (e.g. 0.0025 kg acetic acid/kg solid), the pressed-cake water content significantly decreases (Fig. 4), i.e. the filter-pressing efficiency increases. This change is assumed to result from the change of slurry microstructure after flocculating, which further stems from the coagulation of clay particles during acidification. It is documented that [9], when acidifying the neutral deflocculated slurry, the edges of clay particles (usually existing as plate crystals) adsorb the protons and change from negatively charged into positively charged due to broken bonds, while the faces of those keep negatively charged. The heteropolar attractions between edges and faces, therefore, lead to the coagulation of clay particles, which may act as bridges and further flocculate the surrounding particles including clay, feldspar and quartz. In this way, the small particles flocculate into large agglomerates and the slurry transforms into a paste consisting of agglomerates. For filter-pressing, the main paths of water transferring inside the as-obtained paste are the inter-agglomerate space, resembling thicker tubes with lower specific surface area. Thus, the total friction between water and agglomerate is low, resulting in higher press-cake permeability and thus higher filter-pressing efficiency, as stated before.

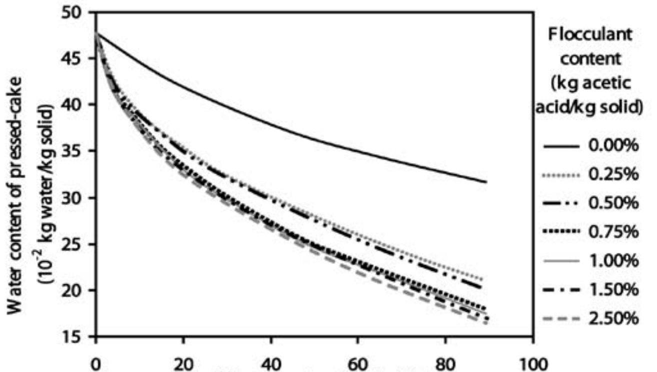

Fig. 3. Water content of pressed-cake versus filter-pressing time, filter-pressed at 20.31 kg/cm^2 from slurries with different flocculant contents (on the solid basis).

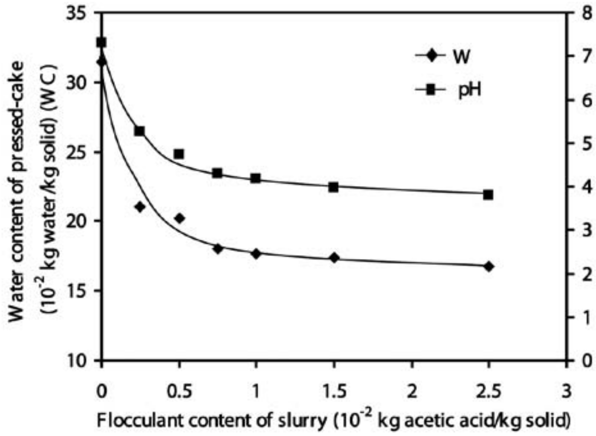

Fig. 4. Pressed-cake water content and filtrate pH versus flocculant content of slurry, filter-pressed at 20.31 kg/cm^2 after 90 min.

Fig. 4 also shows that, the further increase of flocculant content leads to the further decrease of pressed-cake water content with a decreasing change, indicating that the increased amount of acid still helps in the particle flocculation but with decreasing contribution.

Moreover in Fig. 4, it is interesting to notice that, along the increasing of flocculant content, the decrease of filtrate pH is gradually decreasing, similar to the changing trend of the pressed-cake water content. It means that, although contributing little to particle flocculation, the further introduced protons from the acid are still adsorbed onto particles but hardly stay in water.

In view of increasing the filter-pressing efficiency and saving the flocculant cost for industrial application, it is suitable to choose a flocculant content over which the resulting increase of filter-pressing efficiency becomes insignificant, e.g. about 0.0075 kg acetic acid/kg solid in this study. In the lab tests chemical-pure acetic acid (with an estimated cost about 300 €/t) was used as a flocculant. Nevertheless, in industrial applications other cheaper flocculants can be used to minimize the cost, bearing in mind that the optimal content can be deduced following the methodology described in this work.

3.1.2 EFFECT OF FILTER-PRESSING PRESSURE ON FILTER-PRESSING EFFICIENCY

Fig. 5 shows the water content of pressed-cake versus the filter-pressing time, filter-pressed at different pressures from the same slurry with a flocculant content of 0.0075 kg acetic acid/kg solid. For each case, the pressed-cake water content decreases along the filter-pressing time with a decreasing decrement, due to the same reason as above-mentioned for Fig. 3.

In order to analyze the effect of filter-pressing pressure on filter-pressing efficiency better, the water content of pressed-cakes after 90 min at different pressures is shown in Fig. 6. It demonstrates that the increase of filter-pressing pressure results in the decrease of pressed-cake water content (i.e. the increase of filter-pressing efficiency) with a decreasing change.

In view of increasing the filter-pressing efficiency and saving the energy cost for industrial application, it is suitable to choose a filter-pressing pressure over which the specific resulting increase of filter-pressing efficiency becomes almost worthless, e.g. about 20 kg/cm^2 in this study.

According to the previous results in this work, the flocculant content of 0.0075 kg acetic acid/kg solid, the pressure of 20.31 kg/cm^2 and the time of 60 min were used for filter-pressing in subsequent experiments.

3.2 Spray-mixing and Rolling, i.e. granulation

In the DPGP process (Fig. 1), part of slurry is filter-pressed, dried and milled into dry fine powder. The other part of slurry and the as-obtained dry fine powder are spray-mixed in a tower, the slurry droplets adsorbing the dispersed dry powder to form granules (Fig. 2). Rolling is a subsequent treatment to modify the raw granules into final granules.

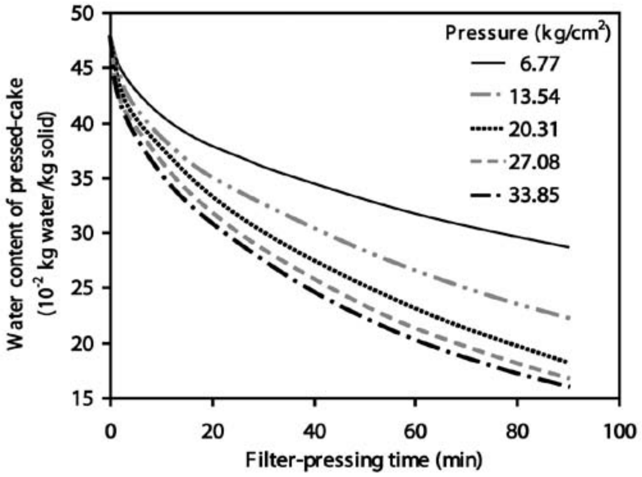

Fig. 5. Water content of pressed-cake versus filter-pressing time, filter-pressed at different pressures from the same slurry with a flocculant content of 0.0075 kg acetic acid/kg solid.

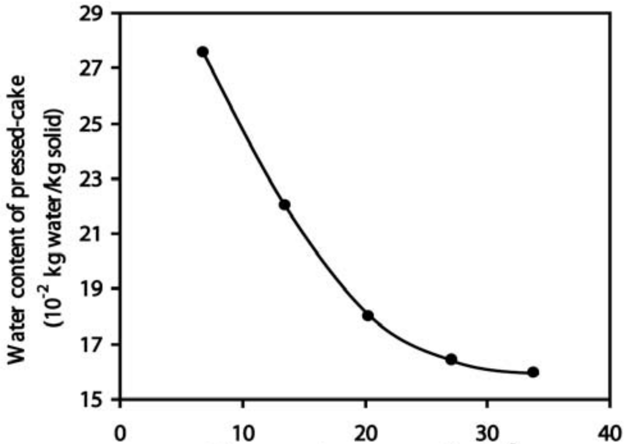

Fig. 6. Water content of pressed-cake versus filter-pressing pressure, filter-pressed after 90 min from the same slurry with a flocculant content of 0.0075 kg acetic acid/kg solid.

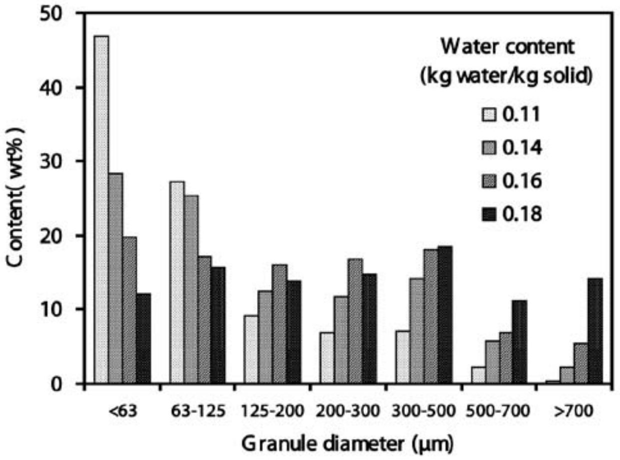

Fig. 7. Granule size distribution of raw granules with different water contents obtained from spray-mixing.

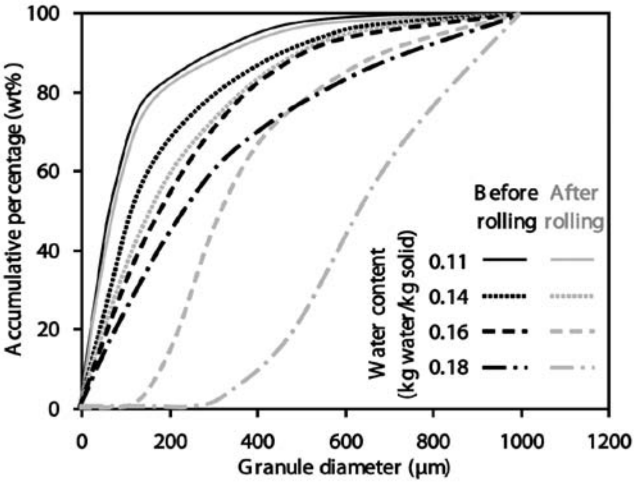

Fig. 8. Granule size distribution of granules before and after rolling treatment.

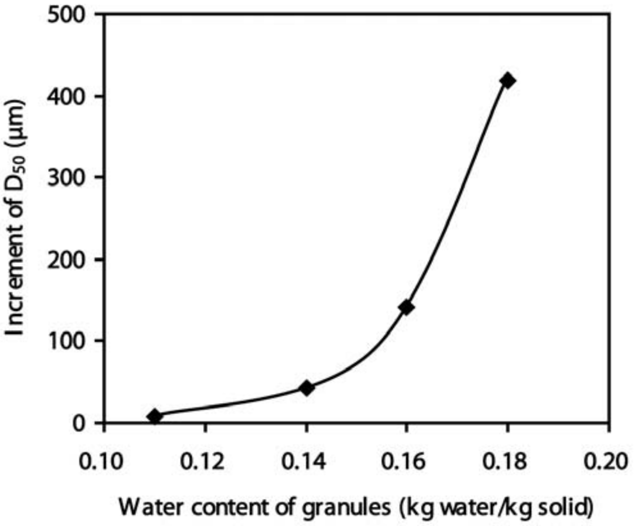

Fig. 9. Increment of D_{50} (the granule diameter under which there are 50 wt% of granules) after rolling treatment versus granule water content.

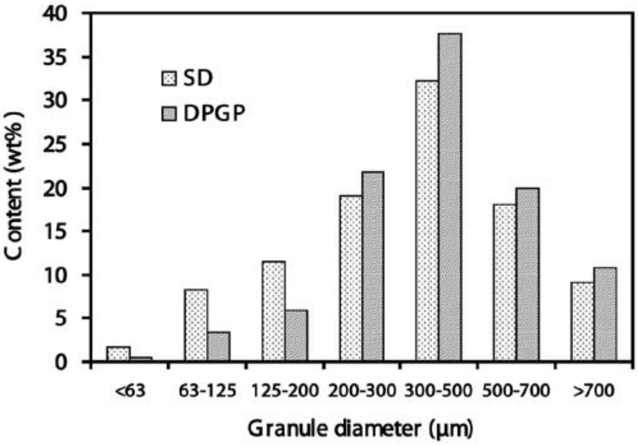

Fig. 10. Granule size distribution of the SD and DPGP powders (a water content of 0.16 kg water/kg solid using in the DPGP spray-mixing and rolling treatment).

3.2.1 EFFECT OF WATER CONTENT ON GRANULATION IN SPRAY-MIXING

By adjusting the weight ratio between the sprayed slurry and dry powder, raw granules with different water contents were obtained from spray-mixing. The size distribution of as-obtained granules is plotted in Fig. 7. It shows that the granule size increases when the water content increases. In fact, the increasing of water content means there are more slurry droplets and less dry powder mixing together, resulting in the granule size growing due to two reasons: on the one hand, since the droplets have larger sizes than the dry fine powder, granule size distribution of their mixture will inherently shift to larger granule sizes; on the other hand, since there are less dry powder to coat and isolate the droplets, there will be more opportunities for droplets to contact with each other and combine into larger granules.

However, within the range of water content shown in Fig. 7, the raw granules always have a large proportion of fine granules (smaller than 125 μm and especially than 63 μm), which is reported to block the deairing paths and compromise the tile pressing [10]. As tested in this work, moreover, the further increase of water content results in large proportion of excessive coarse agglomerations (coarser than 1000 μm), which are also unfit for tile pressing. Therefore, in order to obtain granules with favorable size distribution, a modifying treatment is required. In this work, rolling the raw granules in an empty laboratorial planet mill (without grinding media) for 0.5 min was employed as the modifying treatment.

3.2.2 EFFECT OF WATER CONTENT ON GRANULATION IN ROLLING TREATMENT

Fig. 8 plots the accumulative size distribution of the granules before and after rolling treatment. It shows that, rolling treatment results in the shift of granule size distribution to larger granule sizes. The shift degree, moreover, depends significantly on the water contents.

To analyze the effect of water content on the size-distribution shift of granules during rolling treatment better, Fig. 9 details the increment of D_{50} (the granule diameter under which there are 50 wt% of granules) versus the granule water content. It shows that the D_{50} increment (i.e. granule size increment) also increases as the granule water content increases. This behavior can be explained that, because water promotes the plastic behavior of clays [11], the raw granules with higher water content are much easier to stick together into larger granules during the rolling treatment.

3.2.3 PRESSING-POWDER CHARACTERIZATION

Previous studies have reported [9] that pressing-powder with a granule size distribution of less proportion of fine and coarse granules and more proportion of medium granules is favorable for a high bulk density and, thus, suitable for tile pressing. Fig. 10 shows that the DPGP powder rolled from the raw granules with a water content of 0.16 kg water/kg solid have a granule size distribution similar to that of the SD powder prepared in the pilot plant. Therefore, 0.16 kg water/kg solid was recognized as the optimal water content for spray-mixing and rolling in this work and used in subsequent experiments.

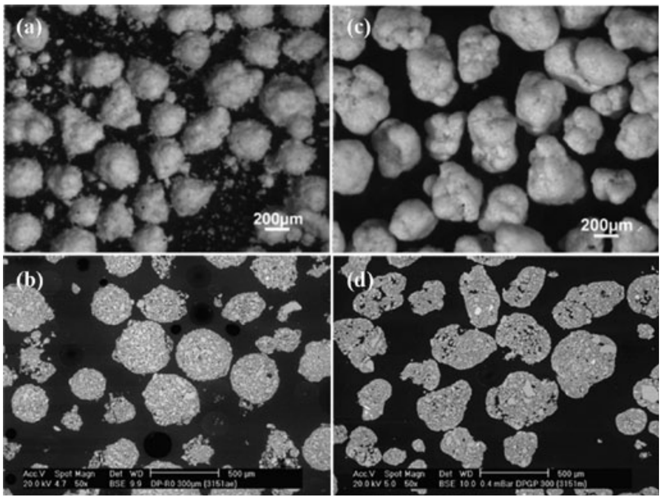

Fig. 11. (a) Morphology and (b) cross-sections of raw granules from spray-mixing, and (c) morphology and (d) cross-sections of final granules after rolling treatment.

Fig. 11 exhibits the morphology and cross-sections of the DPGP granules with a water content of 0.16 kg water/kg solid before and after rolling treatment. It shows that, before rolling treatment, the raw granule is a surface-flocky spherical granule composed of the inner droplet and outer fine powder (Figs. 11a and b); after rolling treatment, the raw granules are combined and compacted into granules with sphericity-irregular shape and smooth surface (Figs. 11c and d). The holes inside the rolled granules (Fig. 11d) are assumed to be the remaining interspaces between the initial finer granules which combine together into final coarser granules during the rolling treatment.

Table 2 exhibits the bulk density and flowability of the DPGP powder with or without rolling treatment. It shows that after rolling treatment, the DPGP granules get a notable increase of bulk density and a slight increase of flowability. The increase of bulk density may be attributed to the improvement of granule size distribution, that is, the decrease of fine granule proportion and the increase of medium granule proportion (Figs. 7 and 10). The increase of flowability is assumed to result synthetically from two positive reasons and one negative reason: the two positive reasons, increasing flowability, are the growing of granule size (Fig. 8) and the smoothing of granule surface (Figs. 11a and c); the one negative reason, reducing flowability, is the irregularity of granule sphericity (Fig 11). In comparison with the SD powder (Table 2), moreover, the DPGP rolled granules are similar in terms of bulk density and slightly lower in terms of flowability.

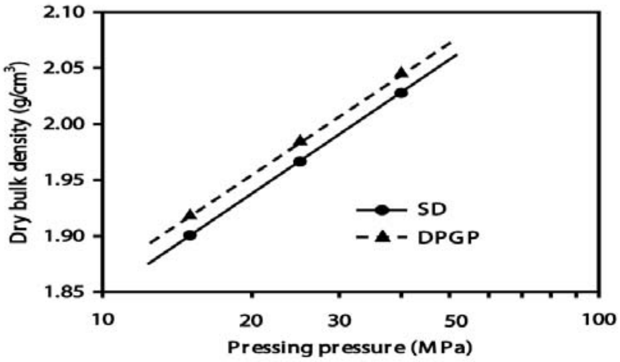

Fig. 12. Bulk density of the DPGP and SD powders versus pressing pressure.

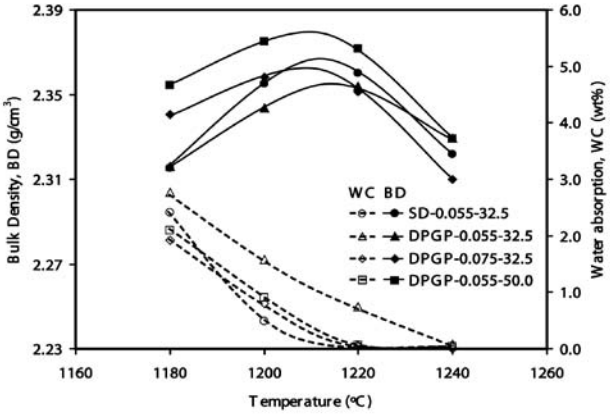

Fig. 13. Bulk density and water absorption versus firing temperature. “DPGP-0.055-32.5” corresponds to specimens pressed from the DPGP powder with a water content of 0.055 kg water/kg solid at 32.5 MPa, the rest being deduced by analogy.

3.3 Performance of pressing-powder in pressing and firing

Fig. 12 plots the pressing behavior of the DPGP and SD powders, pressed at a water content of 0.055 kg water/kg solid. It shows that, at the same pressing pressure, the DPGP powder presents slightly higher bulk density than the SD powder.

Fig. 13 plots the bulk density and water absorption versus firing temperature for both powders (DPGP and SD) pressed at a pressure of 32.5 MPa and a water content of 0.055 kg water/kg solid. The graph shows that the DPGP specimens need a similar maximum-densification temperature as the SD to reach

TABLE 2. BULK DENSITY AND FLOWABILITY OF THE DPGP POWDER SPRAY-MIXED AT WATER CONTENT OF 0.16 KG WATER/KG SOLID WITH OR WITHOUT ROLLING TREATMENT, AND, OF THE SD POWDER.

Pressing-powder	Free-fall bulk density (g/cm^3)	Tapped bulk density (g/cm^3)	Hausner index	Volume flow velocity (cm^3/s)
DPGP without rolling	0.94 ± 0.01	1.17 ± 0.01	1.24	14.03 ± 0.10
DPGP with rolling	1.08 ± 0.01	1.32 ± 0.01	1.23	14.90 ± 0.11
SD	1.11 ± 0.01	1.35 ± 0.01	1.21	16.58 ± 0.10

the maximal bulk density but a higher vitrification temperature to eliminate apparent porosity (i.e. water absorption). For the SD specimens, the vitrification temperature is lower than the maximum-densification temperature. This indicates that the SD specimens can eliminate apparent porosity when fired at maximum-densification temperature, which is the optimum peak firing temperature in porcelain tile industry. For the DPGP specimens, however, the vitrification temperature is higher than the maximum-densification temperature. This means, when fired at maximum-densification temperature, the DPGP specimens will still have open pores, which compromise the frost and stain resistance of fired product [12,13].

Therefore, to produce porcelain tiles from the DPGP powder, it is necessary to modify the processing parameters to reduce the vitrification temperature. In this work, two pressing parameters, including pressing pressure and water content, were respectively increased to reduce the size and volume of green pores in pressed specimens and thus to facilitate vitrification during firing [14,15]. As shown in Fig. 13, when either increasing the water content of pressing-powder (to 0.075 kg water/kg solid) or increasing the pressing pressure (to 50.0 MPa), the vitrification temperature of DPGP specimens can be efficiently reduced to near the maximum-densification temperature. In addition, modifying the starting composition formulation is another major solution to make vitrification temperature lower than maximum-densification temperature for industrial application.

Apart from the bulk density and water absorption, the linear shrinkage of specimens during firing was also characterized. When prepared in the same conditions (press-formed with a water content of 0.055 kg water/kg solid at 32.5 MPa and fired at the maximum-densification temperature), the linear shrinkage of the DPGP specimens is 5.45% which is lower than that of the SD specimens (6.08%). The lower firing linear shrinkage is deemed to lead to a higher dimensional stability which favours the production of large size ceramic tiles.

3.4 Evaluation of energy/water consumption and pollution emission

Table 3 depicts the energy/water consumption and pollution emission of the DPGP and SD processes to produce 1t of pressing-powder. The equipment of each processing step is also proposed. The consumption and emission data are calculated from the relevant data in the literature [5,16-24] and our pilot experimental results. The processing parameters used for calculating are referred to our pilot experimental results as follows.

- In the DPGP process, raw materials are wet milled into the slurry with a water content of 0.47 kg water/kg solid; 68 wt% of the slurry are filter-pressed at 20.31 kg/cm^2 for 1 h into pressed-cake with a water content of 0.22 kg water/kg solid; the pressed-cakes are dried into a water content of 0.02

TABLE 3. ENERGY/WATER CONSUMPTION AND POLLUTION EMISSION FOR PRODUCING 1 T OF PRESSING-POWDER.

Process	Purpose	Step	Equipment	Consumption			Emission (g)		
				Thermal (kWh)	Electric (kWh)	Water (t)	Particle before BAT [e]	Particle after BAT	CO_2
SD	Slurry preparation	Wet milling	Ball mill [a]	---	52.4	0.446	5000	20	---
	Granulation	Spray-drying	Spray dryer [a]	407.5	8	---	4000	60	74164
		BAT application [a]		---	3.9	---	---	---	---
		Total		**407.5**	**64.2**	**0.446**	**9000**	**80**	**74164**
DPGP	Slurry preparation	Wet milling	Ball mill [a]	---	52.4	0.446	5000	20	---
	Dry-powder preparation	Filter-pressing	Chamber filter press [c]	---	4.3	-0.159 [d]	---	---	---
		Drying (Press-cake)	Paddle dryer [a]	134.6	3.9	---	26	26	24499
		Milling (Press-cake)	Raymond mill [b]	---	2.2	---	---	---	---
	Granulation	Spray-mixing	Spray-mixing tower [a]	---	3.0	---	---	---	---
		Rolling	Roller [b]	---	2.8	---	---	---	---
		Drying (Granules)	Vibratory fluid bed dryer [b]	108.4	6.0	---	500	10	19721
		BAT application [a,b]		---	2.5	---	---	---	---
		Total		**243.0**	**77.1**	**0.287**	**5526**	**56**	**44220**

[a] *Consumption and emission data are calculated from the relevant data in the literature [16-22] about the existing wet process of pressing-powder preparation (i.e. raw material wet milling and spray-drying).*

[b] *Consumption and emission data are calculated from the relevant data in the literature [23,24] about the existing dry process of pressing-powder preparation (i.e. raw material dry milling and granulation).*

[c] *Consumption and emission data are calculated from our experimental data and the relevant data in the literature [5] about the filter-pressing process.*

[d] *Minus water consumption corresponds to the recycled filtrate water from the filter-pressing step.*

[e] *BAT (Best Available Techniques) is the use of bag filter [1].*

kg water/kg solid and then milled into dry fine powder; the other 32 wt% of slurry and as-obtained dry powder are spray-mixed into raw granules with a water content of 0.16 kg water/kg solid, which are rolled and then dried into pressing-powder with a water content of 0.055 kg water/kg solid.

- In the SD process, raw materials are wet milled into the slurry with a water content of 0.47 kg water/kg solid and then spray-dried into pressing-powder with a water content of 0.055 kg water/kg solid.

Table 4 further reveals the difference between the DPGP and SD processes in terms of consumption and emission, showing that, the DPGP process has a potential to reduce energy consumption (high decrease in thermal and low increase in electricity), water consumption and pollution emission in comparison with the SD process. These differences result from the difference between the DPGP and SD processes in water-removing procedure, while the total amount of water needed to be removed is the same in the two processes. In the SD process, the water is removed totally by spray-drying, which is an intensive heat treatment entailing significant pollution. In the DPGP process, the water is removed through three steps: one step is a mechanical treatment (i.e. the slurry filter-pressing) which removes about forty percent of the total water in a high-energy-efficient way with no pollution emission but recyclable filtered water; the other two steps (i.e. the pressed-cake drying and the granules drying) are heat treatments which entail much lighter pollution. Therefore, the DPGP process achieves a significant reduction of thermal/water consumption and pollution emission in contrast with the SD process. Of course, the employment of the filter-pressing and other steps complicates the processing procedure and increases the electric consumption; however, the DPGP process is still capable of reducing total energy/water consumption and environmental impacts. In spite of not having included in the study the effect of recycling the tile processing wastes (such as wastewater, sludge and scrap), their reuse should be feasible in the DPGP process, the same as in the existing SD process [25-28], because the wet milling, i.e. the critical step, is also present in the new process (see Fig. 1). In addition, according to an initial estimation, the facility investment of the DPGP process is of the same order of magnitude as that of the SD process at the same output capacity. In rough figures, the investments of the wet mills, tanks and transfers are the same in the two processes, and, the investment of filter-press, dry mill, dryer, spray-mixer and roller in the DPGP process is similar to that of spray-dryer and tail-gas cleaning system in the SD process.

4. CONCLUSION

- To process wet-milled slurry into pressing-powder, the new process (DPGP) introduces two special steps: the filter-pressing step and a novel granulation step consisting of the spray-mixing and rolling treatment.
- The filter-pressing step, a high-energy-efficient mechanical technique, removes and recycles about forty percent of the water needed to be removed in the whole process, contributing to saving energy/water and reducing pollution in the DPGP process, in comparison with the spray-drying process. Either flocculating the wet-milled slurry or increasing the filter-pressing pressure can increase the filter-pressing efficiency, while the specific efficiency increment is decreasing along the increasing of flocculant dosage or filter-pressing pressure.
- The spray-mixing step obtains surface-flocky spherical granules. After rolling treatment, the raw granules transform into granules with larger sizes, sphericity-irregular shape and smoother surface, exhibiting higher bulk density and better flowability. Water content has a major effect on the granulation of both spray-mixing and rolling treatment: the granule sizes increase as the water content increases.
- In comparison with the spray-dried powder, the as above prepared DPGP powder have similar granule size distribution, bulk density, flowability, and pressing/firing behavior, in spite of requiring a higher vitrification temperature for porcelain tile manufacturing.

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TABLE 4. CONSUMPTION AND EMISSION DIFFERENCE BETWEEN THE DPGP AND SD PROCESSES FOR PRODUCING 1 T OF PRESSING-POWDER. [a]

	Consumption			Emission			Total
	Thermal	Electric	Water	Particle before BAT	Particle after BAT	CO_2	
Quantity	-164.5 kWh	12.9 kWh	-0.159 t	-3474 g	-24 g	-29944 g	---
Ratio (%)	-40	20	-36	-39	-30	-40	---
Cost (€) [b]	-6.58	1.29	-0.11	---	---	-0.45	-5.85 [c]

[a] *Negative numbers mean decrease from SD to DPGP, and positive numbers mean increase from SD to DPGP.*

[b] *Considered unit costs are 0.04 €/kWh thermal energy, 0.10 €/kWh electric energy, 0.70 €/t water and 15 €/t* CO_2*, according to the data of Spain in 2010.*

[c] *Since the selling price of spray-dried powder for producing porcelain ceramic tile is about 70€/t in Spain (2011), the percentage of price saving from SD to DPGP can be estimated at 8.4%.*

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