



Omrane Benjeddou^{1,2,*} and Mamdooh Alwetaishi³

- ¹ Prince Sattam bin Abdulaziz University, College of Engineering, Department of Civil Engineering, Alkharj 16273, Saudi Arabia
- ² University of Tunis El Manar, National Engineering School of Tunis, Civil Engineering Laboratory, BP 37, Tunis-Belvédère 1002, Tunisia
- ³ Department of Civil Engineering, College of Engineering, Taif University, P.O. Box 11099, Taif 21944, Saudi Arabia; m.alwetaishi@tu.edu.sa
- * Correspondence: benjeddou.omrane@gmail.com

Abstract: Stone industry has generated about 200 million tons of marble waste by marble processing industries whether in the form of sludge or solid blocks. The accumulated marble wastes contaminate water and air and have harmful effects on human health, plants, and animals. This study focuses on exploring the uses of powder obtained by drying and grinding marble sludge waste, generated from marble manufacturing processes, as a mineral filler for other construction materials. First, physical characterization was performed on marble sludge. Second, the powder preparation process was presented. Thereafter, a set of tests was carried out to identify the chemical, mineralogical, and physical properties of marble powder. By doing so, tests such as chemical analysis, calcium carbonate content, and methylene blue test, as well as mineralogical characterization using X-ray diffraction (XRD), Atterberg limits, particle size analysis, densities, Blaine specific surface, hydraulic property, as well as reaction with admixture, cement, and activity index were conducted. In the last part of this work, the obtained powder properties are compared to the standards requirements to confirm its suitability as mineral filler. The test results showed that the obtained marble powder is too rich in calcite; it is poor of any clay minerals fraction; it is very well graded; it is not reactive; and it does not have any effects on concrete strength; consequently, it can be considered as a mineral filler.

Keywords: environment; marble sludge waste; filler; construction materials; valorization

1. Introduction

During cutting and polishing processes, marble industry units generate huge quantities of marble sludge [1,2]. Marble sludge is a substance consisting of grains derived from the sawing and the polishing processes and water used to cool and lubricate machines [3]. About 25% of the processed marble turns into powder or dust during the sawing, shaping, and polishing process [4]. The large amounts of marble sludge wastes have a considerable impact both on the environment and on public health. In addition, these accumulated wastes contaminate the surface and underground water reserves [5,6].

The main solution to avoid the risks of marble sludge wastes is reusing them in order to produce new products; they can also be used as admixtures to other construction materials. As a consequence of waste recycling, both the need for raw materials and energy consumption decrease. Thus, recycling preserves natural resources for future generations. In addition, for the products made using recycled materials, the cost is reduced [7–9]. There are several reuses and recycling solutions for marble waste materials: they can be used as aggregates, as powder, or in their sludge state.

Marble sludge wastes are mainly used as fine or coarse aggregates for ordinary cement concrete [9–11]. The results showed that marble aggregates do not affect either the mechanical properties or ther heology of the concrete. In addition, marble aggregates can



Citation: Benjeddou, O.; Alwetaishi, M. Valorization of Powder Obtained from Marble Sludge Waste and Its Suitability as a Mineral Filler. *Crystals* 2021, *11*, 619. https://doi.org/ 10.3390/cryst11060619

Academic Editors: Assem Barakat and Alexander S. Novikov

Received: 2 May 2021 Accepted: 27 May 2021 Published: 31 May 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). be used as compounds of self-compacting concrete because they do not affect its rheological behavior in the fresh state and its mechanical behavior in the hard state [12].

Another solution for marble wastes is their reuse as powder in different applications such as an additive material for bricks manufacturing [13–15]. In this case, Aukour, F.J. [13] showed that physical and mechanical properties of the produced bricks are well improved. In addition, marble powder was used as a mineral filler incorporated in self-compacting concrete to improve its rheological behavior in fresh state and to increase its compressive strength both in early age and in long term [16,17]. Marble powder is also used as an additive in cement manufacturing which greatly contributes to the economy and the ecology of countries [18–20].

Regarding the use of marble wastes in its sludge state, there are several recycling solutions. As an example, Rashwan, M.A. et al. [21] used marble sludge in its wet state as cement replacement on concrete mixes. The results of its work showed an improvement toward the water penetration depth and sulphuric acid attack resistance. The results of all hardened concrete mixes are compatible with the requirements of concrete bricks and concrete paving units in terms of water absorption and compressive strength. These results are confirmed by Prošeka et al. [22] and Yuting et al. [23], all of whom demonstrate that inert marble sludge addition reinforces cement concrete matrix and acts as a filler. Moreover, the results of the work of Ahmed, O.M. et al. 2016 [24] showed that using up to 20% of marble sludge improves both physical and mechanical properties of concrete products. In addition, marble sludge was used as precursor for new alkali-activated materials [25,26]. The results of other research projects confirm the possibility of the reuse of marble waste in tire mixture production [27,28].

In addition, Sardinha et al. [29] and Rodrigues et al. [30] demonstrated that fine aggregates obtained from dried marble sludge improve very well both the physical and the mechanical performances of concrete in fresh and in hard states [29,30]. Moreover, these fine aggregates can be used in the production of lean masonry mortars [31]. Other research results showed that 15% of marble sludge is deemed optimum for the manufacturing of burnt clay bricks leading toward sustainable, economical, environment-friendly, and energy-efficient construction [32,33].El-Alfi, E.A. and Gado, R.A. [34] confirm the possibility of the production of cement by raw mix contained in 25% kaolin, 20% gypsum, and 55% marble sludge waste at the firing temperature of 1200 °C.This new process leadsto the production of less CO₂ emission cement known as sulfoaluminate-belite cement.

Finally, marble sludge and urban sewage sludge can be added in different proportions to clay in a ceramic body. The results showed that the selection of adequate contents of sludge to be added to the ceramic body will be controlled by the usual standards applied to construction materials [35,36].

The main objective of this experimental investigation is to evaluate the suitability of powder, obtained by drying and grinding marble sludge waste as mineral filler incorporated in different construction materials. To this end, an experimental study was conducted in two phases. In the first phase, physical properties of marble sludge were evaluated. In second phase, chemical, mineralogical, and physical properties of marble powder were evaluated through a complete set of experimental tests such as chemical analysis, calcium carbonate content, methylene blue test, mineralogical composition using microstructure analysis, Atterberg limits, particle size analysis, densities, Blaine specific surface, hydraulic property, and reaction with admixture, cement, and activity index.

2. Materials and Methods

2.1. Characterizations of Marble Sludge

The marble sludge used in this work was collected from a local marble manufacturing unit in Tunisia. The frequently manufactured marbles are from the marble quarry of Thala in Central Tunisia. The geological indication of marble rocks of this quarry is "Cretaceous", which are marked by diversified colors. Marble sludge waste was generated in its wet state during the process of cutting and polishing of marble stones (Figure 1). In this part, the physical properties of marble sludge were evaluated through density measurement and Atterberg limits tests.



Figure 1. Marble sludge waste in wet state.

2.1.1. Density Measurement

The bulk density of marble sludge was determined according to the requirements of NF EN 1097-7 standard [37], via the following steps:

- Weigh the dry sample: M_S;
- Put the sample in packing material;
- Weigh the packed sample: M_{SP};
- Calculate the weight of packing material, M_p, using this relation:

$$M_{\rm P} = M_{\rm SP} - M_{\rm S} \tag{1}$$

- Calculate the volume of packing material, V_p, by this equation:

$$V_{\rm P} = \frac{M_{\rm P}}{\rho_{\rm p}} \tag{2}$$

where ρ_P is the packing material density;

- Place the sludge sample in graduated cylinder and record the sample volume, Vs;
- Finally, calculate the marble sludge sample density, ρ_s , using the following relation:

$$\rho_{s} = \frac{M_{s}}{V_{s} - V_{p}} \tag{3}$$

2.1.2. Atterberg Limits Tests Setup

Atterberg limits of marble sludge were determined to choose the appropriate process of powder preparation. Atterberg limits are the following: liquidity limit (LL), plasticity limit (PL), plasticity index (PI), liquidity index (LI), and consistency index (CI).

The plasticity limit (PL) and the liquidity limit (LL) are determined according to the requirements of ASTM D4318-17 standard [38]. Thereafter, the plasticity index, the liquidity index, and the consistency index are computed as follow:

 $\sqrt{}$ The plasticity index (PI), which is the size of the range of water content where sludge exhibits plastic properties, was calculated using this relation:

$$PI = LL - PL \tag{4}$$

 $\sqrt{}$ The liquidity index (LI), which is used for scaling the natural water content of marble sludge sample to the limit, is calculated using the following equation:

$$LI = \frac{(W - PL)}{(LL - PL)}$$
(5)

where W is the natural water content.

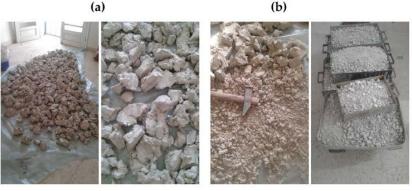
 $\sqrt{}$ The consistency index (CI), which is an indication about marble sludge consistency, is computed using this relation:

$$CI = \frac{(LL - W)}{(LL - PI)}$$
(6)

2.2. Marble Powder Preparation

Marble powder was obtained by drying, crushing, grinding, and sieving marble sludge waste through the following process (Figure 2):

- Marble sludge was dried in open air, for 48 h (Figure 2a).
- The dried sludge was crushed into small blocks using a hammer (Figure 2b).
- Subsequently, in order to expect all water content, the sludge was dried for 24 h in an oven, at the temperature of 80 °C (Figure 2c).
- The dried sludge was ground in fine powder using a crusher (Figure 2d).
- Finally, filler was obtained by sieving the powder using a sieve 63 μm (Figure 2e)







(d)

(e)



Figure 2. Different steps of marble powder preparation: (**a**) Air drying of marble sludge, (**b**) Crushing of marble sludge, (**c**) Drying of marble sludge in oven, (**d**) Grinding of marble sludge, (**e**) Sieving of marble powder.

2.3. Marble Powder Characterization Procedure

In this part, different standards were followed to identify the chemical, mineralogical, physical, and hydraulic properties of the obtained marble powder. The conducted tests are chemical analysis, calcium carbonate (CaCO₃) content, methylene blue test, mineralogical characterization performed by means of X-ray Diffraction (XRD), particle size analysis, densities, Blaine specific surface (BSS), hydraulic properties, reaction with admixture, and reaction with cement and the activity index.

2.3.1. Chemical Analysis

Chemical composition of marble powder was determined using the atomic absorption spectrometry test (AAS), according to the requirements of NF EN ISO 15586 standard [39].

2.3.2. Calcium Carbonate Content CaCO₃

Calcium carbonate (CaCO₃) content measurement was determined according to the requirements of NF P 94-048 standard [40] using a calcimeter apparatus. This test consisted of measuring the CO₂ volume developed by chloridric acid reacting with marble powder. Calcium carbonate content was calculated using this relation:

$$CaCO_{3}(\%) = \frac{120 \times V_{b} \times P}{m \times (\theta_{b} + 273)}$$
(7)

where V_b is the gas volume released in the burette in cm³, P is the atmospheric pressure in kPa, m is the sample mass in g, and θ_b is the ambient temperature in °C.

2.3.3. Methylene Blue Test

The methylene blue test was performed on marble powder samples to check if the powder contains any clay mineral fraction. This test was conducted based on the requirements of the standard NF P 94-068 [41]. It consisted of dispersing bat least 200 g of powder in 500 g of distilled water in a beaker. Subsequently, methylene blue value (MBV) was calculated by the following equation:

$$MB = \frac{V}{M} \times 10 \tag{8}$$

where M is the sample mass, in g, and V is the total volume of dye solution injected, in milliliters.

2.3.4. Mineralogical Composition

The chemical composition and the morphological forms of the main mineral components of marble powder are identified using the X-ray diffraction (XRD) technique and the scanning electron microscope (SEM).

2.3.5. Particle Size Analysis

Grain size distribution was obtained using the sedimentation method, according to the requirements of NF P 94-057standard [42], because the maximum grain size of marble powder is equal to 63 µm.

Subsequently, Hazen coefficient (Cu) and curvature coefficient (Cc) were calculated using the following equations:

$$Cu = \frac{D_{60}}{D_{10}}$$
(9)

$$Cc = \frac{(D_{30})^2}{D_{60} \times D_{10}}$$
(10)

where D_{10} , D_{30} , and D_{60} are the passing grain diameters at 10%, 30%, and 60% finer, respectively.

2.3.6. Densities

Bulk density and absolute density using the pycnometer method were performed on powder samples according to the requirements of NF EN 1097-7 standard [37].

2.3.7. Blaine Specific Surface

Blaine specific surface (BSS) of marble powder was determined using Blaine air permeability apparatus, according to the requirements of the standard NF EN 196-6 [43]. This method assumes that all powder grains have a uniform spherical shape. Subsequently, the specific surface was computed through the measurement of the flow time of a specific quantity of air through a compressed sample layer of a given size and porosity.

2.3.8. Hydraulic Property

Hydraulic property of marble powder with water was checked by measuring the temperature of marble powder paste as function of time [3]. The paste sample was prepared by mixing 500 g of marble powder with 125 mL of water.

2.3.9. Reaction with Cement

This part consisted of studying the reactivity of marble powder with cement. This reactivity was verified by comparing the setting time of cement paste to that of marble powder-cement paste. This test was carried out according to the requirements of the standard NF EN 196-3 [44].

Cement paste was prepared by mixing 500 g of cement and 135 mL of water. Marble powder–cement paste was prepared by mixing 500 g of cement, 250 g of marble powder, and 202 mL of water. Both water/cement and water/(cement + marble powder)were taken equal to 0.27.

In addition, in order to better understand the reactivity between cement and marble powder, an in depth microstructure analysis was made on the two pastes using X-ray Diffraction (XRD) patterns.

2.3.10. Reaction with Admixture

The reactivity of marble powder with admixture was checked by the Marsh cone test, according to the requirements of the standard NF P18-507 [45]. For this, the flow times of different grouts mixing with superplasticizer (SP), water, and marble powder were determined by varying the SP amount. The physicochemical characteristics of the used superplasticizer are presented in Table 1.

Table 1. Physicochemical characteristics of the SP.

Density	pН	Na ₂ O _{eq} (%)	Dry Extract (%)	CI- (%)
1.06 ± 0.01	4.5 to 6.5	$\leq 1\%$	28.0% to31.0%	$\leq 0.1\%$

2.3.11. Activity Index

In this part, the marble filler effects on the compressive strength of concrete or mortar was tested via the determination of the "Activity index (I)". According to the standard NF EN 206-1 [46], the activity index is expressed by the following equation:

$$I = \frac{R}{R_0}$$
(11)

where R and R_0 are the compressive strengths of the cement–marble powder mortar and the control cement mortar, respectively, at the age of 28 days.

The first step of this test consists of preparing the two mortars according to the requirement of the standard NF EN 196-1 [47]. Control mortar was prepared by mixing 450 g of cement, 1350 g of sand, and 225 mL of water. Whereas cement–marble powder

mortar was prepared by mixing 75% of cement (337 g), 25% of marble powder (113 g), 1350 g of sand, and 225 mL of water.

The second step consisted of preparing 3 prismatic specimens with the dimensions of $4 \times 4 \times 16$ cm for each mortar.

Finally, at the age of 28 days, all specimens were tested under compressive strength test and the two compressive strengths R and R_0 were determined.

3. Results

3.1. Characterizations of Marble Sludge

The tests results show that bulk density of marble sludge is about 2.45 g/cm^3 . Moreover, theresults presented in Table 2 shows that the water content of marble sludge in its natural state is about 30%. In addition, the liquidity limit (LL) is equal to about 31%.

Parameter	Water Content	Plasticity Limit	Liquidity	Plasticity	Liquidity	Consistency
	W (%)	PL (%)	Limit LL (%)	Index PI	Index LI	Index CI
Value	30	Undefined	31	Undefined	Undefined	Undefined

Table 2. Atterberg limits results of the marble sludge.

As a first conclusion, the wet state of the collected marble sludge waste represents its liquidity limit. The second conclusion is that marble sludge is free from any fine clay proportion because its plasticity limit was undefined.

3.2. Characterizations of Marble Powder

3.2.1. Chemical Composition

The chemical composition of marble powder, presented in Table 3, shows that marble powder is composed of about 94.88% of calcium carbonate (CaCO₃). Note that the calcium carbonate amount is the sum of loss on ignition (LOI) and carbon dioxide (CaO) amount: In addition, the presence of calcium oxide (CaO) was observed higher (52.28%), which exceeds magnesium oxide (MgO) in minor amount (0.50%), indicating the calcite form of marble powder [25]. This result was confirmed by the XRD test result (Figure 3), which shows also the presence of marble powder in calcite form. Other compounds such as silica, alumina, ferric oxide, sodium oxide, and potassium oxide were also observed in small amounts. The observed loss on ignition (LOI) is about 42.60%, which may be attributed to the loss of carbon dioxide [25].

	Table 3.	Chemical	composition	of mar	ble s	ludge.
--	----------	----------	-------------	--------	-------	--------

Component	CaCO ₃	LOI	CaO	MgO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	MgCO ₃	Sulphur Trioxide (SO ₃)	Moisture
Percentage	94.88	42.60	52.28	0.50	3.00	0.39	0.14	1.04	0.03	0.02

In addition, the calcium carbonate content test result shows that marble powder contains about 90% of CaCO₃. This result confirms the one obtained by the AAS test analysis.

Subsequently, the methylene blue test result shows that marble powder MPV is about 0.42. This result demonstrates that marble powder is poor of any clay mineral fraction. As a consequence, when this powder is added to cement materials, any disorders affected hydration reaction, rheology (both in fresh and in hard states), and durability are avoided.

Finally, as a main result, the obtained filler has a chemical composition similar to that of limestone filler, which was already used as building material. As a consequence, the possibility of reusing this powder as a mineral filler is very high.

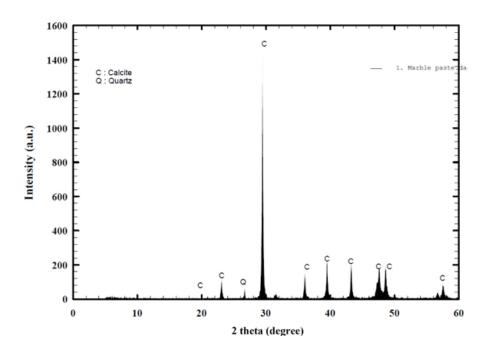


Figure 3. XRD results for marble sludge.

3.2.2. Mineralogical Composition

Figures 3 and 4 show XRD patterns and SEM micrographs of marble paste, respectively. According to these results, the main crystalline mineral of marble paste is calcite mineral (CaO₃) with significant peaks. In addition, quartz mineral is also detected in very low concentration. Traces of SiO₂ and corundum Al_2O_3 were observed in marble powder by Munir, M.J. et al. [25].

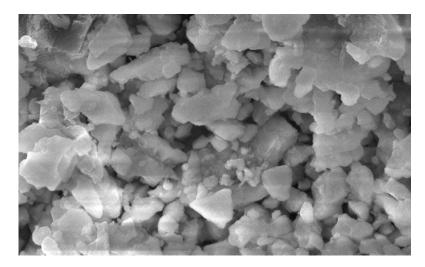


Figure 4. SEM micrographs of marble sludge.

3.2.3. Particle Size Distribution

The particle size distribution curve of marble powder is presented in Figure 5. The result shows that 65% of powder grain sizes are between the split $0/10 \ \mu\text{m}$. This result demonstrates that the tested powder is very fine, and it can be considered as a filler.

According to the grading curve presented in Figure 5, Hazen coefficient and curvature coefficient of marble powder are, respectively, 6.1 (greater than 4) and 0.42 (less than 5). These two results are very encouraging because they demonstrate that the granular distribution of the obtained filler is well graded, and it contains large ranges of particle sizes.

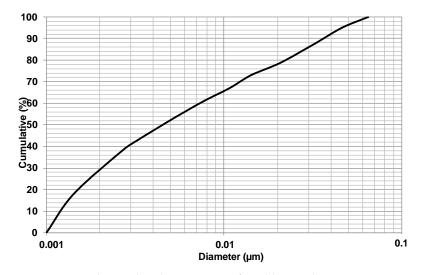


Figure 5. Particle size distribution curve of marble powder.

3.2.4. Physical Properties

The results presented in Table 4 show that bulk and absolute densities of marble powder are 0.63 g/cm^3 and 2.65 g/cm^3 , respectively.

Table 4. Physical parameters of marble powder.

Parameters	Particle Size Absolute Density (mm) (g/cm ³)		Bulk Density (g/cm ³)	Blaine Specific Surface BSS (cm ² /g)	
Values	0/0.063	2.65	0.63	9350	

According to the results presented in Table 4, marble powder BSS is equal to $9350 \text{ cm}^2/\text{g}$. We can conclude that marble powder is thinner than cement and that, indeed, it can be considered as a filler.

3.2.5. Hydraulic Property

The results of this test, presented in Figure 6, show that the paste temperature increases very slowly from 18 to 20 °C during more than 6 hours for a room temperature equal to 18 °C. This result indicates that there are no hydraulic properties between marble powder and water. Indeed, marble powder can be considered as an inert component in cementing materials such as concrete and mortar.

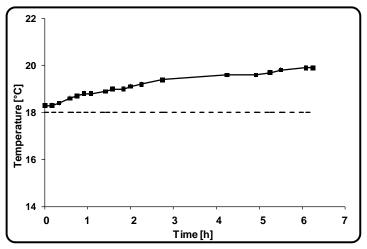


Figure 6. Marble powder paste temperature as function of time.

This last result was confirmed by SEM micrographs and XRD patterns of marble sludge, presented in Figure 3 and in Figure 4, respectively. According to these results, it is clear that marble powder has no pozzolanic activity.

3.2.6. Reaction with Cement

The results presented in Figure 7 indicate that the initial set times of cement paste and marble powder–cement paste are 150 and 140 min, respectively, and the final set times are 250 and 240 min for cement paste and marble powder–cement paste, respectively. These results prove that marble powder has no effects on the cement paste setting time and, as a consequence, it has no reactivity with cement. This result confirms that marble powder is an inert filler.

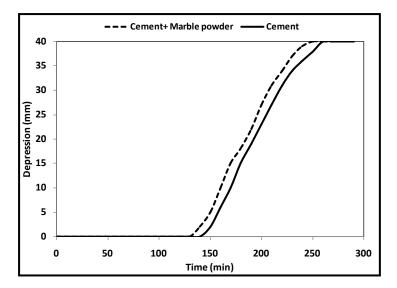


Figure 7. Setting times of cement paste and marble powder-cement paste.

Figures 8 and 9 show the XRD analysis of cement paste and marble powder–cement pattern, respectively. The results show that the presence of tricalcium silicate (C_3S) and dicalcium silicate (C_2S) was higher in cement paste. The results also show the presence of tricalcium aluminate (C_3A), tetracalcium alumino ferrite (C_4AF), and calcium oxide (C) in small amounts (C) for cement paste. However, marble-cement paste shows the presence of calcite (C) in major amounts and the absence of quartz (Q).

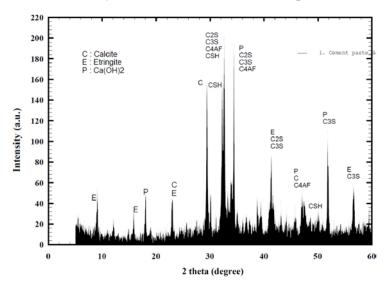


Figure 8. XRD results for cement paste.

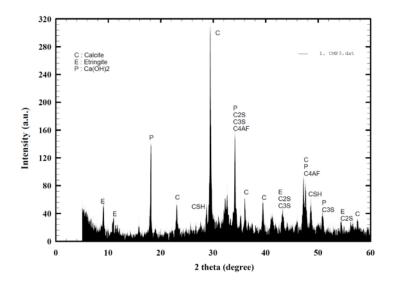


Figure 9. XRD results for marble powder-cement paste.

The results show also that when marble powder is added to cement paste, the phase composition does not change qualitatively. In addition, both calcium hydroxide (CH) and portlandite have a peak for the two pastes.

The absence of quartz showed that marble powder cannot be a part of the hydration process [25]. As a consequence, marble powder has no hydraulic property and it can be considered as an inert material which does not lead to a change in the phase composition of cementing material.

3.2.7. Reaction with Admixture

The result presented in Figure 10 shows that all SP/marble powder ratios give the same flow time. This result indicates that marble powder is not reactive with the superplasticizer.

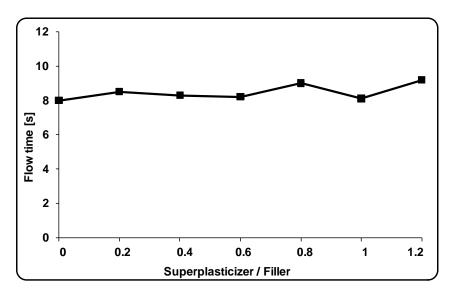


Figure 10. The flow time of marble powder–superplasticizer grout as a function of SP/marble powder ratio.

3.2.8. Activity Index

The result shows that the activity index of marble powder is about 0.88. This is a very encouraging result because, according to the standard NF P18-508 [48], the minimum activity index of mineral filler must be about 0.71 at the age of 28 days for a substitution

rate of K = 25%. As a consequence, the addition of marble powder as a filler in cementing materials does not affect their compressive strengths.

4. Discussion

4.1. Identification of Marble Powder as Mineral Filler

The chemical and physical properties of mineral fillers used in cementing materials, according to standard NF P18-508 [48], are shown in Table 5.

Table 5. Physical and chemical properties of mineral fillers according to standards NF P18-508.

Properties	Passing of Sieve 0.063 mm	Maximum Grain Size (mm)	Blaine Specific Surface	CaCO ₃ Content	Methylene Blue Value	Sulphur Trioxide (SO ₃) Content	Activity Index
Criteria	>70%	<0.125 mm	>2200 cm ² /g	>65%	<1.3 g/100 g	<0.15%	>0.71
Mable Powder	100%	0.063 mm	9350 cm ² /g	93.30%	0.42 g/100 g	0.03%	0.88

As a conclusion, the powder obtained by crushing marble sludge can be considered as a mineral filler added to cementing materials due to the following reasons:

The main criteria set by the standard NF P18-508 [48] are satisfied:

- Maximum grain size: D = 0.063 mm < 0.125 mm;
- Passing of sieve 0.063 mm: 100% > 70%;
- Blaine specific surface: BSS = $9350 \text{ cm}^2/\text{g} > 2200 \text{ cm}^2/\text{g};$
- $CaCO_3$ content = 93.30% > 65%;
- Methylene blue value: MBV = 0.42 g/100 g < 1.3 g/100 g;
- Sulphur trioxide (SO₃) content: 0.03% < 0.15%;
- Activity index: I = 0.88 > 0.71.
- Marble powder is very well graded.
- Marble powder has no hydraulic property. Indeed, it is inert with water.
- Marble powder is not reactive with cement and, as a consequence, is inert. Indeed, there is no effect on the hydraulic reaction of cement when this powder is added. In addition, the mechanical properties of the obtained cementing materials are affected.
- Marble powder is inert with admixture. This property is very encouraging because, in this case, the admixture amount depends only on the cement amount.

4.2. Possibilities of Use of Marble Powder

As a consequence of the recycled marble powder properties, and in order to reduce environmental pollution, the following several uses of this powder as raw material are recommended:

4.2.1. Reuse of Marble Filler in Mortar

Marble powder can be incorporated as mineral addition in mortar to improve its properties [49–51]. These researchers demonstrate that the density, and the compressive and tensile strengths increase when marble filler is added to cement mortar.

4.2.2. Reuse of Marble Filler in Concrete

Marble powder can be incorporated as a filler in concrete to improve its mechanical properties or to reduce the total void content [7,52–54]. The obtained results from these studies show also that the incorporation of marble powder in concrete as a mineral filler does not affect the hydration reaction of cement. This result confirms that marble filler can be considered as an inert material.

In addition, marble powder can be added to self-compacting concrete as a filler to improve the rheological behavior in the fresh state and the mechanical behavior in the hard state [55–59]. The results have shown that marble filler is the best one for increasing the compressive strength of self-compaction concrete, compared with other mineral fillers.

Moreover, the microstructure studies show that marble filler does not have any hydraulic reaction, and it is inert both with cement and with admixtures.

4.2.3. Reuse of Marble Filler as Raw Material for Cement

Marble powder can be used as a component for manufacture cement for the following reasons: First, to limit the environmental impacts of cement plants by the reduction of CO_2 emissions. Second, to reduce the cost by reducing the clinker dosage of cement in favor of marble filler. Finally, to improve the cooking process performances [20,60,61].

4.2.4. Reuse of Marble Filler as Raw Material for Bricks

Marble powder can be used as a filler in the brick industry [32,62]. The results of these studies show that the obtained brick quality was not affected by adding marble filler to the initial mixture. To the contrary, the total void content and water absorption are remarkably reduced. In addition, the mechanical strength of the new bricks was increased compared with clay bricks only. Finally, the results show also that the cooking program of bricks are not affected by marble filler, whileCO₂ emission does not increase.

4.2.5. Reuse of Marble Filler in Soil Pigment

Marble powder can be incorporated as a mineral filler in soil pigment-based paints [63]. The results of this study showed that the quality of soil pigment-based paints is remarkably improved when using marble powder in its composition.

4.3. Design of Marble Filler Production Unit

In this part, a proposal of the design of a production unit of marble filler is presented. The main parts of the unit are the following (Figure 11):

- A sludge storage zone: The sludge collected from marble manufactory is stored in this zone.
- First open-air drying step: The sludge is partially dried in open air in its initial state. The main advantage of open-air drying is its energy saving.
- Crushing step: The partial dried sludge is crushed into small blocks.
- Second open-air drying step: After crushing, small blocks are dried in open air until completely dry. The drying time was calculated according to the water content.
- > Grinding step: The dried small blocks are finely ground in order to produce the filler.
- The sieving step: The grinding powder is sieved in this step into the appropriate granular fraction.

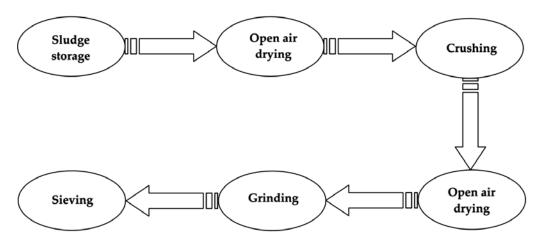


Figure 11. Design of marble filler production unit.

5. Conclusions

This experimental work focuses on the evaluation of the suitability of powder obtained by drying and grinding marble sludge wastea s mineral filler incorporated in different construction materials. This research presents a complete characterization of marble filler via a large set of tests. In addition, it presents a proposal of marble filler production design.

Due to the following results, marble powder can be considered as a mineral filler, and it has several possibilities of application as raw material for construction products:

- Chemical results show that the obtained marble powder has a similar chemical composition compared to commercialized limestone filler, because it is too rich in calcite (CaCO₃) while being poor of any clay mineral fraction.
- Sieve analysis test shows that marble powder is very well graded due to its uniform and curvature coefficients. This characteristic gives the concretes and mortars a low air content, and, as a consequence, their mechanical properties will be improved.
- The activity index shows that when adding marble powder to mortars and concretes, their compressive strength will not be affected.
- The most important result is that marble powder is not reactive with water, with cement, or with admixture. As a consequence, the hydraulic reaction in cementing materials will not be affected. This result was confirmed by a microstructure analysis.
- The obtained powder can be considered as an eco-friendly product because it gives a feasible solution for the great amount of marble sludge waste in the world.

According to the obtained results, and, considering the proposed production unit design, this filler is more economical than normally used fillers because of its low energy consumption during the different production stages. In addition, it can be considered as an eco-friendly product.

Our future research consists of preparing a comparative study of the reuse of marble waste materials in other construction materials in order to evaluate its economic benefits.

Author Contributions: Investigation, O.B.; writing—original draft preparation, O.B. and M.A.; writing—review and editing, O.B. and M.A.; supervision, O.B. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by [Taif University, Researchers Supporting Project] grant number [TURSP-2020/196].

Informed Consent Statement: Not applicable.

Data Availability Statement: No data, models, or code were generated or used during the study.

Acknowledgments: The author would like to acknowledge the finical support provided from Taif University Researchers Supporting Project Number (TURSP-2020/196).

Conflicts of Interest: The authors declare no conflict of interest.

References

- 1. Ashish, D.K.; Verma, S.K.; Kumar, R.; Sharma, N. Properties of concrete incorporating sand and cement with waste marble powder. *Adv. Concr. Constr.* 2016, *4*, 145–160. [CrossRef]
- 2. Mansour, R.; ElAbidine, R.Z.; Brahim, B. Performance of polymer concrete incorporating waste marble and alfa fibers. *Adv. Concr. Constr.* **2017**, *5*, 331–343.
- Alyousef, R.; Benjeddou, O.; Soussi, C.; Khadimallah, M.A.; Mohamed, A.M. Effects of Incorporation of Marble Powder Obtained by Recycling Waste Sludge and Limestone Powder on Rheology, Compressive Strength, and Durability of Self-Compacting Concrete. *Adv. Mater. Sci. Eng.* 2019, 2019, 1–15. [CrossRef]
- 4. Khodabakhshian, A.; De Brito, J.; Ghalehnovi, M.; Shamsabadi, E.A. Mechanical, environmental and economic performance of structural concrete containing silica fume and marble industry waste powder. *Constr. Build. Mater.* **2018**, *169*, 237–251. [CrossRef]
- 5. Benjeddou, O.; Soussi, C.; Jedidi, M.; Benali, M. Experimental and theoretical study of the effect of the particle size of limestone fillers on the rheology of self-compacting concrete. *J. Build. Eng.* **2017**, *10*, 32–41. [CrossRef]
- Alyousef, R.; Benjeddou, O.; Khadimallah, M.A.; Mohamed, A.M.; Soussi, C. Study of the Effects of Marble Powder Amount on the Self-Compacting Concretes Properties by Microstructure Analysis on Cement-Marble Powder Pastes. *Adv. Civ. Eng.* 2018, 2018, 1–13. [CrossRef]
- 7. Singh, M.; Choudhary, K.; Srivastava, A.; Sangwan, K.S.; Bhunia, D. A study on environmental and economic impacts of using waste marble powder in concrete. *J. Build. Eng.* **2017**, *13*, 87–95. [CrossRef]
- 8. Arel, H. Şahan. Recyclability of waste marble in concrete production. J. Clean. Prod. 2016, 131, 179–188. [CrossRef]
- 9. Ouassila, B.; Houria, H.; Leila, K.; Mouloud, B.; Assia, A.; Chaher, R. Valorization of marble. Adv.Concr. Constr. 2020, 9, 217–225.

- 10. Ulubeyli, G.C.; Artir, R. Properties of Hardened Concrete Produced by Waste Marble Powder. *Procedia Soc. Behav. Sci.* 2015, 195, 2181–2190. [CrossRef]
- Kore, S.D.; Vyas, A.K. Impact of marble waste as coarse aggregate on properties of lean cement concrete. *Case Stud. Constr. Mater.* 2016, 4, 85–92. [CrossRef]
- 12. Uygunoğlu, T.; Topçu, I.B.; Çelik, A.G. Use of waste marble and recycled aggregates in self-compacting concrete for environmental sustainability. J. Clean. Prod. 2014, 84, 691–700. [CrossRef]
- 13. Aukour, F.J. Incorporation of Marble Sludge in Industrial Building Eco-blocks or Cement Bricks Formulation. *Jordan J. Civ. Eng.* **2009**, *3*, 58–65.
- Alyousef, R.; Benjeddou, O.; Soussi, C.; Khadimallah, M.A.; Jedidi, M. Experimental Study of New Insulation Lightweight Concrete Block Floor Based on Perlite Aggregate, Natural Sand, and Sand Obtained from Marble Waste. *Adv. Mater. Sci. Eng.* 2019, 2019. [CrossRef]
- 15. Bilgin, N.; Yeprem, H.A.; Arslan, S.; Bilgin, A.; Günay, E.; Marşoglu, M. Use of waste marble powder in brick industry. *Constr. Build. Mater.* **2012**, *29*, 449–457. [CrossRef]
- 16. Belaidi, A.; Azzouz, L.; Kadri, E.; Kenai, S. Effect of natural pozzolana and marble powder on the properties of self-compacting concrete. *Constr. Build. Mater.* **2012**, *31*, 251–257. [CrossRef]
- 17. Esat, K.; Ince, A.R. A preliminary concrete mix design for SCC with marble powders. Constr. Build. Mater. 2009, 23, 1201–1210.
- 18. Aruntaş, H.Y.; Gürü, M.; Day, M.; Tekin, I. Utilization of waste marble dust as an additive in cement production. *Mater. Des.* 2010, *31*, 4039–4042. [CrossRef]
- 19. Aliabdo, A.A.; AbdElmoaty, A.M.; Auda, E.M. Re-use of waste marble dust in the production of cement and concrete. *Constr. Build. Mater.* **2014**, *50*, 28–41. [CrossRef]
- 20. El-Sayed, H.A.; Farag, A.; Kandeel, A.; Younes, A.A.; Yousef, M.M. Characteristics of the marble processing powder waste at Shaq El-Thoaban industrial area, Egypt, and its suitability for cement manufacture. *HBRC J.* **2018**, *14*, 171–179. [CrossRef]
- 21. Rashwan, M.A.; Al-Basiony, T.M.; Mashalya, A.O.; Khalil, M.M. Behaviour of fresh and hardened concrete incorporating marble and granite sludge as cement replacement. *J. Build. Eng.* **2020**, *32*, 101697. [CrossRef]
- 22. Prošek, Z.; Nežerka, V.; Tesárek, P. Enhancing cementitious pastes with waste marble sludge. *Constr. Build. Mater.* 2020, 255, 119372. [CrossRef]
- 23. Yuting, X.; Eleftherios, A.; Aspasia, K.; Stefano, S. Fresh and hardened properties of cement mortars using marble sludge fine sand cement sludge fines. *Constr. Build. Mater.* **2019**, 220, 142–148.
- 24. Ahmed, O.M.; Baher, A.E.K.; Baseln, N.S.; Amr, M.E.; Mohammed, A.R. Effects of marble sludge incorporation on the properties of cement composites and concrete paving blocks. J. Clean. Prod. 2016, 112, 731–741.
- Munir, M.J.; Abbas, S.; Nehdi, M.L.; Kazmi, S.M.S.; Khitab, A. Development of Eco-Friendly Fired Clay Bricks Incorporating Recycled Marble Powder. J. Mater. Civ. Eng. 2018, 30, 04018069. [CrossRef]
- 26. Coppola, B.; Palmero, P.; Montanaro, L.; Tulliani, J.M. Alkali-activation of marble sludge: Influence of curing conditions and waste glass addition. *J. Eur. Ceram. Soc.* 2020, 40, 3776–3787. [CrossRef]
- 27. Marras, G.; Careddu, N. Sustainable reuse of marble sludge in tyre mixtures. Resour. Policy 2018, 59, 77-84. [CrossRef]
- 28. Khalil, A.; Shaikh, S.N.; Nudrat, Z.R.; Farzana, H. The effect of silica on the properties of marble sludge filled hybrid natural rubber composites. *J. King Saud Univ. Sci.* **2013**, *25*, 331–339.
- 29. Sardinha, M.; deBrito, J.; Rodrigues, R. Durability properties of structural concrete containing very fine aggregates of marble sludge. *Constr. Build. Mater.* **2016**, 119, 45–52. [CrossRef]
- 30. Rodrigues, R.; deBrito, J.; Sardinha, M. Mechanical properties of structural concrete containing very fine aggregates from marble cutting sludge. *Constr. Build. Mater.* **2015**, *77*, 349–356. [CrossRef]
- Syed, A.K.K.I.; Ashok, K.V. Experimental investigation on utilization of dried marble slurry as fine aggregate in lean masonry mortars. J. Build. Eng. 2019, 23, 185–192.
- 32. Rupali, B.; Upender, P.; Apoorva, V. Manufacturing of sustainable bricks: Utilization of mill scale and marble slurry. *Mater. Today Proc.* **2020**, *26*, 2136–2139.
- Muhammad, J.M.; Syed, M.S.K.; Yu, F.W.; Asad, H.; Muhammad, U.A.K. Thermally efficient fired clay bricks incorporating waste marble sludge: An industrial-scale study. J. Clean. Prod. 2018, 174, 1122–1135.
- 34. El-Alfi, E.; Gado, R. Preparation of calcium sulfo aluminate-belite cement from marble sludge waste. *Constr. Build. Mater.* **2016**, 113, 764–772. [CrossRef]
- 35. Montero, M.A.; Jordán, M.; Crespo, M.S.H.; Sanfeliu, T. The use of sewage sludge and marble residues in the manufacture of ceramic tile bodies. *Appl. Clay Sci.* **2009**, *46*, 404–408. [CrossRef]
- Acchar, W.; Vieira, F.; Hotza, D. Effect of marble and granite sludge in clay materials. *Mater. Sci. Eng. A* 2006, 419, 306–309. [CrossRef]
- 37. Tests for Mechanical and Physical Properties of Aggregates—Part 7: Determination of the Particle Density of Filler—Pyknometer Method; AFNOR: Paris, France, 2008; NF EN1097-7.
- Standard Test Methods for Liquid Limit, Plastic Limit, and Plasticity Index of Soils; American Society for Testing and Materials: West Conshohocken, PA, USA, 2017; ASTM ASTMD4318-17.
- 39. Water Quality—Determination of Trace Elements using Atomic Absorption Spectrometry with Graphite Furnace; AFNOR: Paris, France, 2004; NF EN ISO 15586.

- 40. Soil: Investigation and Testing—Determination of the Carbonate Content—Calcimeter Method; AFNOR: Paris, France, 1996; NF P94-048.
- 41. Soils: Investigation and Testing. Measuring of the Methylene Blue Adsorption Capacity of à Rocky Soil; AFNOR: Paris, France, 1998; NF P94-068.
- 42. Soils Investigation and Testing. Granulometric Analysis. Hydrometer Method; AFNOR: Paris, France, 1992; NF P94-057.
- 43. Methods of Testing Cements—Part 6: Determination of Fineness; AFNOR: Paris, France, 2012; NF EN 196-6.
- 44. Methods of Testing Cement—Part 3: Determination of Setting Times and Soundness; AFNOR: Paris, France, 2017; NF EN 196-3.
- 45. Additions for Concrete. Water Retention. Method for Measurement of Fluidity by Flowing with the "Cone De Marsh"; AFNOR: Paris, France, 2017; NF P18-507.
- 46. Concrete—Specification, Performance, Production and Conformity; AFNOR: Paris, France, 2014; NF EN 206-1.
- 47. Methods of Testing Cement—Part 1: Determination of Strength; AFNOR: Paris, France, 2016; NF EN 196-1.
- 48. Additions for Concrete—Limestone Additions—Specifications and Conformity Criteria; AFNOR: Paris, France, 2012; NF P18-508.
- 49. Wang, J.; Ma, B.; Tan, H.; Du, C.; Chu, Z.; Luo, Z.; Wang, P. Hydration and mechanical properties of cement-marble powder system incorporating tri isopropanolamine. *Constr. Build. Mater.* **2021**, 266, 121068. [CrossRef]
- 50. Kabeer, K.S.A.; Vyas, A.K. Utilization of marble powder as fine aggregate in mortar mixes. *Constr. Build. Mater.* **2018**, *165*, 321–332. [CrossRef]
- 51. Vardhan, K.; Goyal, S.; Siddique, R.; Singh, M. Mechanical properties and microstructural analysis of cement mortar incorporating marble powder as partial replacement of cement. *Constr. Build. Mater.* **2015**, *96*, 615–621. [CrossRef]
- 52. Atiyeh, M.; Aydin, E. Data for bottom ash and marble powder utilization as an alternative binder for sustainable concrete construction. *Data Brief.* **2020**, *29*, 105160. [CrossRef]
- 53. Ashish, D.K. Concrete made with waste marble powder and supplementary cementitious material for sustainable development. *J.Clean. Prod.* **2019**, *211*, 716–729. [CrossRef]
- 54. Varadharajan, S. Determination of mechanical properties and environmental impact due to inclusion of fly ash and marble waste powder in concrete. *Structures* **2020**, *25*, 613–630. [CrossRef]
- 55. Meeran, M.; AnujDash, A.K.; Gupta, S. Rheological and strength properties of self-compacting concrete incorporating marble and granite powders. *Mater. Today Proc.* 2020, *32*, 1005–1013. [CrossRef]
- 56. Choudhary, R.; Gupta, R.; Nagar, R.; Jain, A. Mechanical and abrasion resistance performance of silica fume, marble slurry powder, and fly ash amalgamated high strength self-consolidating concrete. *Constr. Build. Mater.* 2021, 269, 121282. [CrossRef]
- 57. Alyamaç, K.E.; Ghafari, E.; Ince, R. Development of eco-efficient self-compacting concrete with waste marble powder using the response surface method. *J. Clean. Prod.* 2017, 144, 192–202. [CrossRef]
- Sadek, D.M.; El-Attar, M.M.; Ali, H.A. Reusing of marble and granite powders in self-compacting concrete for sustainable development. J. Clean. Prod. 2016, 121, 19–32. [CrossRef]
- 59. Tennich, M.; Ben Ouezdou, M.; Kallel, A. Thermal effect of marble and tile fillers on self-compacting concrete behavior in the fresh state and at early age. *J. Build. Eng.* **2018**, *20*, 1–7. [CrossRef]
- 60. Alabduljabbar, H.; Benjeddou, O.; Soussi, C.; Khadimallah, M.A.; Alyousef, R. Effects of incorporating wood sawdust on the firing program and the physical and mechanical properties of fired clay bricks. *J. Build. Eng.* **2020**, *35*, 102106. [CrossRef]
- 61. Belouadah, M.; Rahmouni, Z.A.; Tebbal, N. Influence of the addition of glass powder and marble powder on the physical and mechanical behavior of composite cement. *Procedia Comput. Sci.* **2019**, *158*, 366–375. [CrossRef]
- 62. Saboya, F.; Xavier, G.; Alexandre, J. The use of the powder marble by-product to enhance the properties of brick ceramic. *Constr. Build. Mater.* **2007**, *21*, 1950–1960. [CrossRef]
- 63. Tressmann, D.M.G.A.; Pedroti, L.G.; de Carvalho, A.F.; Ribeiro, J.C.L.; Cardoso, F.D.P.; Lopes, M.M.S.; deOliveira, A.F.; Ferreira, S.O. Research into the use of marble waste as mineral filler in soil pigment-based paint sand as an active pigment in water borne paints. *Constr. Build. Mater.* **2020**, *241*, 117976. [CrossRef]