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# Circular economy in marble industry: from stone scraps to sustainable water-based paints

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## Abstract

Huge amounts of sludge waste are produced every day during the sawing and processing of carbonate ornamental stones. Most of these materials are landfilled and create serious environmental and health hazards. Thus, it is mandatory to find alternative solutions to recycle them. Since calcium carbonate is the most widely used mineral filler for interior and exterior emulsion paints, this investigation focused on the feasibility of trying out innovative methods of circular economy in a waste processing area. Focusing in the Orosei Marbles' producing area (North-East Sardinia, Italy), the aim of this study was to compare the performance of water-based paints, through the use of marble sludge waste, as mineral fillers, rather than commercial CaCO<sub>3</sub>. According to standard normative tests were carried out on paint samples, prepared according to an experimental formulation, after a preliminary chemical and physical characterization of the fillers. The results indicate that the marble sludge waste can be successfully used as mineral filler in water-based paints, and that it requires only light pre-treatments consisting of deagglomeration and mechanical dispersion in a wet medium, thus confirming its environmental and economic benefits.

**Keywords:** marble sludge, calcium carbonate filler, water-based paint, dimension stone, circular economy, stone industry sustainability.

## 1. Introduction

After stalling due to the SARS-CoV-2 pandemic, the Dimension Stone Industry is increasing again: the world net product could exceed one hundred million tons in 2025 [1]. However, we have to take into consideration that the world gross quarrying production was about 316 million tons in 2019; about 53 % of that amount, however, is regarded as quarrying waste.

With regards to the stone-processing stage, we have noticed that the world production has reached more than ninety million tons, and consequently this means that more than sixty-three million tons of stone-processing scraps is produced. Therefore, we can say that, on a global level, if the quantity of material extracted in the quarry is 100, the total percentage of waste is about 73 % [2]. This raises a substantial problem from the environmental, economical and social point of view.

With regards to commodities, carbonate stones (such as marble, limestone and travertine) account for 58 percent of the whole production of dimension stone, followed by granites (38 %) and other stones (as basalt, trachytes, sandstones, etc.). The latest statistics [1] indicate that the yearly production of carbonate stone waste/scrap by the dimension stone industry amounts to 174 million tons; however, only a small portion of it has a high grade (> 95 %) of calcium carbonate.

The last sixty years have seen an increasing development of the Sardinian ornamental stones sector through a spontaneous process of growth of an important number of small and medium-sized enterprises, mainly active in the Northern Sardinia area (granite sector) and in the *marble*

area of Orosei (which is shown in Fig. 1), North East Sardinia [3]. In both mining districts, the companies mainly deal with the quarried materials extraction and processing.

Ornamental stones companies have to face the economic burden of their waste disposal (about 270,000 t per year in the producing area of Orosei); these materials mainly consist of non-commercial blocks (due to their poor quality), unshaped blocks (unsuitable to block cutter sawing), rubble, crushed slabs/strips/tiles and sludges derived from the water treatment plants [4]. The latter, consisting of micro-fine sawdust sludge (about 12,000 t per year in Orosei), has over time been considered as *real waste* and therefore landfilled. This solution is not no longer compatible with the increasing and pressing need for environmental care and eco-sustainability. Following this perspective, in the recent past many researchers have undertaken studies for the re-use of this kind of "waste" in specific industrial sectors linked with building. The first step of the research focused on the building sector. State of the art in recovery and utilization of carbonate sawdust mainly relates to its use as a substitute for more expensive ingredient in the building sector [5, 6]. Marras et al. [7], André et al. [8] and Martínez-Martínez et al. [9], have reported the feasibility in adding marble industry scraps in concrete admixtures, while Topçu et al. [10] and Felekoğlu [11] in self compacting concrete, Gencil et al. [12] in concrete paving blocks, Buyuksagis et al. [13] in cement-based adhesive mortar, Marras et al. [14] in gypsum plaster and Marvila et al. [15] in cementitious ceramic. Applications in red ceramic have been investigated by Montero et al. [16] and Devant et al. [17]. Some studies have been carried out in brick ceramic field [18-20]. Further investigation has regarded porcelain stoneware [21], artificial stone slabs [22] and composites [23]. Table 1 summarizes the percentages of carbonate sludge used in the main studies cited above.

Table 1. Carbonate sludge percentages used in the main studies (ordered by year)

Authors	Product	Range of carbonate sludge content [% by weight]	Original carbonate stone
Díaz and Torrecillas, 2007 [21]	Ceramic blocks	100	dunite-serpentinite
Felekoğlu, 2007 [11]	Concrete	20 - 38	limestone
Saboya et al., 2007 [20]	Ceramic bricks	5 - 20	marble
Montero et al., 2009 [16]	Ceramics	15 - 35	marble
Topçu et al., 2009 [10]	Concrete	5 - 30	marble
Marras et al. 2010a [19]	Ceramic bricks	3 - 10	limestone
Marras et al., 2010b [7]	Concrete	5 - 15	limestone
Devant et al., 2011 [17]	Ceramics	n.a.	marble
Galekatis et al., 2012 [5]	Paving blocks	18 - 52	limestone
Gencil et al., 2012 [12]	Paving blocks	10 - 40	marble
Buyuksagis et al., 2017 [13]	Bonding mortars	14 - 74	marble
Marras et al., 2017a [14]	Gypsum plaster	5 - 15	limestone
Singh et al., 2017 [6]	Cement	10 - 25	marble
Cobo-Ceacero et al., 2019 [18]	Ceramic bricks	2.5 - 10	marble
Martínez-Martínez et al. 2020 [9]	Clinker	2.5 - 10	limestone
Pappu et al., 2020 [23]	Composite materials	60	marble
Marvila et al., 2021 [15]	Cementitious ceramics	2 - 10.5	marble

All the above-mentioned applications involve a huge amount of materials to be reutilized, although with low economic value, being all building products. For this reason, the aim of the research is to demonstrate the feasibility of reusing these marble scraps in the production of high value-added products [24].

Currently, no other mineral pigment provides an overall performance equal to that of ground calcium carbonate (GCC) - the high calcium type - and it is the most extensively used in the largest number of industries [25]. With regards to this point, this investigation moved to high economic value areas such as tyre one, with a particularly encouraging outcome [26]; the current study turns then to the paint sector, in which the calcium carbonate content accounts for 50 – 70% of the emulsion paint used for interiors.

In paint, GCC is used as extender and filler to improve hiding power, brightness, washability and application properties, and to reduce consumption of expensive pigments, such as titanium dioxide [27-28]. An estimated 8.8 Mt of GCC and PCC were used for the production of paint in 2011, nearly all of which in architectural products. The majority, 6.95 Mt, was in the form of GCC [27].

According to ISO 4618: 2014 and EN ISO 3262-1, extenders/fillers are materials in granular or powder form, practically insoluble in the application medium and used as a constituent of paints in order to modify or influence certain physical properties. It is therefore clear that fillers not only offer economic advantages, but also they are capable of bringing technical-applicative properties to the final formulations. Fillers strongly affect some of the main final properties of paints, including rheological properties, hiding power, mechanical properties, chemical resistance, resistance to atmospheric agents (and therefore durability) and colorimetric properties [29-30]. It should be noted that the market value of dry GCC with an average particle size of 4-9 microns is about 80 €/t (FOB price, bagged) [31-32].

For this reason, this high market value of micronized CaCO<sub>3</sub> has now driven researchers into experimenting the re-use of this micro-fine sludge, provided that its characteristics meet the industrial, sometimes rigid, standard requirements.

Mineralogy, physical characteristics such as whiteness index, pH, oil absorption, granulometric properties such as shape, size and particle size distribution, play a very important role in the performance of the paints, especially for their weathering resistance in low pH environments.

According to this concept, the marble sludge (MS) was analyzed in order to compare its properties with the GCC [33-34].

After verifying compatibility between MS and GCC, the experimental phase began in cooperation with a Sardinian paint producer Company. Two white wall paints for interiors were characterized and compared, introducing MS instead of GCC.

## 2. Geological settings



Figure 1. Aerial view of Orosei Marble basin (source: Google Earth – own elaboration)

The Orosei Marble, which is associated with the Formation of M. Bardia emerges on the eastern and southern slopes of Mount Tuttavista (locality Oroe), in the Municipality of Orosei where an important extractive basin of stone materials for ornamental use is developed. These quarries are located on the Mesozoic limestones unearthed following the excavation of the Pleistocene conoids. These ornamental rocks, known as *Orosei marble*, are sold under the name of "Marble"; however although this term is commercially acceptable, it is geologically improper, since it is a non-metamorphic limestone. These "Marbles", once cut and polished, are placed on the market

with the generic commercial name of "Biancone Tirreno", but the remarkable lithological variety allows them to be differentiated in relation to the textural differences (e.g.: *Perlato*, *Nuvolato*, *Venato*, *Brecciato*) and colors (from light to dark beige). Currently, the commercial lithotype named "Fiocco di neve" (snowflake), which is characterized by a cream color tending to white, is the market's most requested range [35].

All these materials are used all over the world and exported to all continents to be used both indoors and in the external cladding of buildings, in urban planning works as well as for urban furniture, for the creation of sculptures and public art.

The main lithotypes emerging in the area are bioclastic and ooid calcarenites/calcirudites (with typically bimodal ooid grainstone), mostly in centimeter/decimetric layers. The layers are often graded and erosive based and they result in parallel to crisscrossing at a small angle. The most common granules are ooids, foraminifera, algae, ketetide and mollusc bioclasts, coated grains, oncoids. Sometimes small ketetide bioconstructions may be present. In the coarser levels without micritic matrix it is possible to observe pores filled with calcite (like keystone vugs). Sub parallel fractures (sheet cracks) or transverse to stratification, discontinuous and with micritic granular fillings and/or sparitic cements, attributable to early and syndimentary diagenesis phases are very frequent [36].

The macro-fossil content consists of frequent colonies of hexacorals and porifers (including ketetids and stromatoporids), gastropods (in particular nerineids), bivalves (including diceratids) and corals. In the upper part, microfacies with ostracods, oogones of caraceous algae, and benthic foraminifera prevail, including miliolides. The age established mainly on microfacies, in particular foraminifera and green algae, would extend from the Kimmeridgian to the lower Berriasian according to Dieni and Massari [37-38]. Recent biostratigraphic analyzes have shown a distribution between the upper Titonian and the lower Valanginian.

### 3. Materials and Methods

#### 3.1 $\text{CaCO}_3$ characterization

As mentioned in the introduction, the MS suffered the same characterization analyses as GCC [34].

The MS sample has been collected at the end of the filter press section in a factory belonging to Orosei marble district. Before starting the analysis phase, MS samples, containing about 25% moisture, were dried in oven at a temperature of  $110 \pm 5^\circ\text{C}$  until they reached the constant weight, and then quartering reduction was made, in order to obtain representative samples.

Specimens were measured for major elements by an X-Ray Rigaku Spectrometer (mod. Primus IV) on pellets (about 4 g). Using the instrument software, the concentrations were calculated by fundamental parameters method, after calibration with international standards set.

Diffraction analyses were performed on powdered samples (micronized in agate jar and grinded below  $63 \mu\text{m}$ ) using a Rigaku MiniflexII X-ray diffractometer operating with Cu anticathode, voltage of 30 kV and 15 mA of current. Raw data have been processed using Match Crystal Impact software recognising the mineralogical phases by comparison with the ICDD (International Centre for Diffraction Data) database [39]. In order to maximize the resolution, the XRPD spectrum was collected with a speed of  $0.2^\circ/2\theta/\text{min}$  sampling at each  $0.02^\circ/2\theta$ .

The particle size distributions were determined using a laser light scattering analyzer CILAS 1180 (Orleans, France).

Apparent specific weights were determined by a Micromeritics AccuPyc II 1340 He Pycnometer in accordance with EN 1097-7 [40].

Thin layers of MS and GCC were spread on a stub and graphitized by LEICA EM SCD005 apparatus equipped with a CEA035 carbon evaporation supply.

SEM observations of the samples were carried out using a Zeiss Evo LS15 Scanning Electron Microscopy equipped with tungsten filament as electron source.

Colorimetric measurements were carried out by using a Konica Minolta CM-700d spectrophotometer (illuminant D65). The instrument has been adjusted to repeat the measurement 6 times on each point: for each sample eight points have been identified and measured. The results were evaluated as  $L^*$  (brightness),  $a^*$  (redness colour), and  $b^*$  (yellowness colour) coordinates.

The total colour difference  $\Delta E$  was calculated according to the CIE-76 and CIE-2000 colour space standards [41-42].

### 3.2 Water-based paint mix-design

The white wall paint for interiors recipe was studied and designed together with the paints producer Company; the components are listed in Tab. 2, with the alternative replacement of GCC with MS.

The constituents were mixed in the same order in which they appear in table. Each ingredient was weighted, gradually combined in a mixer to form a homogeneous paste.

The first emulsion was obtained adding GCC and was named EDI; the second emulsion based from MS was named ORO, and since the MS was delivered dehydrated and in compacted lumps, the sample required a light pre-treatment consisting of deagglomeration and mechanical dispersion in a wet medium.

Table 2. White wall paint for interiors recipe [kg]

White washable wall paint			
Constituent	kg	Weight [%]	Progressive
H <sub>2</sub> O	2.614	26.14	2.61
Orgal PST 65	0.818	8.18	3.43
Till-Disp	0.011	0.11	3.44
Tioxide TR 92	0.318	3.18	3.76
Talc CHB2	0.227	2.27	3.99
CaCO <sub>3</sub> (GCC or MS)	5.691	56.91	9.68
Tillplast STA	0.068	0.68	9.75
Butylglycules	0.034	0.34	9.78
Ammonia	0.023	0.23	9.8
Sodium Hypochlorite	0.136	1.36	9.97
Anti-bubbles	0.025	0.25	9.97
Intercid Cmed	0.009	0.09	9.97
Carbocell	0.025	0.25	10.00
<b>Total</b>	<b>10.00</b>	<b>100.00</b>	<b>10.00</b>

Paints are composed by pigments, binders, solvents, additives and extenders/fillers [43].

Binders create a film that protects the substrate and keeps the pigment in place:

- ORGAL PST 65 is a commercial styrene acrylic copolymer emulsion introduced to form soft and elastic films when dried above 0°C. Exhibits very good elasticity, water resistance, tensile strength and adhesion.

Pigments give the colour of the finished product:

- TIOXIDE TR92 is a multipurpose white pigment of titanium dioxide, characterized by high opacity giving an excellent hiding power.

Solvents affect the application of the coating by thinning the paint, allowing it to be brushed or sprayed. Once on the substrate, the solvent evaporates leaving the dry film containing the binder and pigment. In this case multicomponent solvent mixture was used, particularly:

- DISTILLED WATER.
- BUTILGLYCULES is an excellent co-solvent in aqueous coating systems (water-based paints).
- AMMONIA is a basic aqueous solvent, pH stabilizer, and has the advantage of slowing down the chemical degradation of the thickener.

Additives and fillers are used to customize the properties of the paint:

- CARBOCELL is used as water-retentive and thixotropic agent.
- SODIUM HYPOCHLORITE is used on a large scale as a disinfectant.
- INTERCID CMED is a family of isothiazolinone (chloro-methyl and methyl isothiazolinone) and ethylenedioxydimethanol (EDDM) combination based biocides.
- TALC is used as filler and extender providing reinforcement.
- ANTIBUBBLES avoid air-bubble formation acting on surface tension reduction.
- TILL-DISP promotes the particle dispersion and their wettability favouring the paint stability.
- TILLPLAST STA enhances the paint elasticity.

- $\text{CaCO}_3$  (GCC and MS) is used as filler, white pigment and extender also providing coating reinforcement.

### 3.3 Experimental phase

The paint samples of ORO and EDI suffered four specific characterization tests, according to UNI EN ISO 10795, that are the same used from the paint Company in order to commercialize their water-based paints, as follows: alkali resistance, paint hiding power, paint dirt pick-up and wet scrub resistance.

In addition, a qualitative evaluation was made in order to verify the workability and durability of our samples (hand-brushing test).

#### 3.3.1 Alkali resistance

This parameter refers to the property of a paint that describes its ability to resist fading, discoloration or deterioration when exposed to alkaline substances, i.e. low pH.

The alkali resistance tests were carried out according to the UNI 10795:1999 standard [44]. Specimens films (200 microns thickness) of both paints (EDI and ORO) were prepared and cured for 28 days in a climatic chamber (Angelantoni mod. Discovery DY250) at a temperature of  $23 \pm 2$  °C and a relative humidity of  $50 \pm 5\%$ .

After completing the first curing cycle, the specimens were immersed one part in deionized water for about 100 mm of immersion, the second part in a 0.5 M sodium hydroxide solution inside hermetically sealed glass containers and stored at  $23 \pm 2$ °C for 15 days.

At the end of the retention period, the consistency and appearance of the film have been visually evaluated. The results have been related to the classification reported by the technical normative following the scheme reported in Tab. 3.

Table 3. Alkali resistance classification [44]

Classification	Observations
Alkali resistant paint	It can be also applied on cement substrates
Not alkali resistant paint	Check the substrate alkalinity before application
Paint not resistant to water immersion	Alkali resistance determination not possible

#### 3.3.2 Paint hiding power

Hiding power is the ability of pigmented paints to cover the colour or the colour differences of a background; it also has pigment properties. The value is determined by photometric methods.

The procedure indicated in the UNI 11271:2008 standard [45] was followed, using the method A. The standard specifies a method for evaluating the hiding power of white or light-colored emulsion paints. One hundred parts per mass of paint are diluted with 10 parts by mass of distilled water and applied on a black and white support with a wet film thickness of about 100 micrometers.

After the preparation of specimens on Leneta's white/black cards, they were subjected to a curing period of 28 days in the climatic chamber at a temperature of  $23 \pm 2$  °C and a relative humidity of  $50 \pm 5\%$ .

At the end of the curing, the tristimulus Y values on white and black areas have been measured using the spectrophotometer and their percentage contrast ratio was calculated (RC).

The data were compared with the values reported in the reference standard table (Tab. 4).

Table 4. Paint hiding power classification [45]

Classification	Tristimulus Y Value(Rc)
Great	$\geq 98 \%$

Good	$\geq 96 - < 98 \%$
Medium	$\geq 93 \% - < 96 \%$
Not sufficient	$< 93 \%$

### 3.3.3 Paint dirt pick-up

The factors that influence the tendency of coatings to collect dirt are complex and multiple and depend also on the environmental conditions.

The correct mix-design of the coating, through the choice and proportion of the components (e.g. binders, fillers, pigments and additives) as well as their physical characteristics, can guarantee a high paint performance, minimizing the problem of dirt pick-up mostly on external coatings.

The paint dirt pick-up measurements were carried out in accordance with the reference standard UNI 10792:1999 [46].

The specimens were prepared with a spiral-type film stretcher, calibrated with a bar for the release of a film thickness of 200 microns, on Leneta cards and then conditioned for 28 days in the climatic chamber at a temperature of  $23 \pm 2 \text{ }^\circ\text{C}$  and a relative humidity of  $50 \pm 5\%$ .

At the end of the curing phase, the specimens have been half immersed in an aqueous solution of carbon black pigment (Corax N326) for  $30\text{s} \pm 1\text{s}$ , after they have been rinsed and dried for 24 h. At the end of the drying phase, the different brightness ( $\Delta L$  index) between the two parts of the Leneta card has been evaluated. The final values for EDI and ORO paints were the averages compared with those reported in classification reference standard (Tab. 5).

Table 5. Dirt pick-up classification [46]

Classification	$\Delta L$
Very Low	$\leq 3$
Low	$3 \div 9$
Medium	$9 \div 15$
High	$> 15$

### 3.3.4 Wet scrub resistance

The tests were conducted on EDI and ORO specimens in duplicate, according to UNI 10560:1996 [47], using brush method. This test is necessary to determine the number of washing cycles, with brush and surfactant solution, which cause the removal of the film and the uncovering the interior paint substrate.

The obtained mean values were compared with the classification table according to standards (Tab. 6).

Table 6. Wet scrub resistance classification [47]

Classification	Reference values
Excellent wet scrub resistance	$> 5000$
Wet scrub resistant	$1000 \div 5000$
Suitable for applications that do not require wet scrub resistance	$< 1000$

### 3.3.5 Hand-Brushing test

A qualitative evaluation of the brush strokes was done after two years to assess the workability, durability and homogeneity of the paints, when applied in double coating on the vertical surface of an outdoor concrete pillar in an industrial warehouse.

## 4. Results and discussion



#### 4.1 GCC and MS chemical-physical characterization and their comparison

The GCC is a calcium carbonate with rhombohedral structure (Calcite) of excellent quality, derived from calcareous rocks of a sedimentary nature in the territory of the province of Trapani (Italy).

A comparison between GCC and MS chemical composition and physical properties is reported in Tab. 7, being general filler requirements for paint industry.

Table 7. Chemical composition and physical properties of MS and GCC

Parameter	MS	GCC
CaCO <sub>3</sub> [wt %]	99.14	98.67
MgCO <sub>3</sub> [wt %]	0.94	0.37
SiO <sub>2</sub> [wt %]	0.15	0.15
Fe [wt %]	0.02	0,005
Loss of Ignition [wt %]	43.91	43.88
pH in sature water solution	9.20	8.36
Specific weight (on the granulate) [kg/dm <sup>3</sup> ]	2.71	2.71
Hardness [Mohs] on the granulate	3	3
Oil intake (cooked linen) [%]	19.46	19.40
Distilled water intake [%]	27.62	27.50
Luminance Y [%]	95.32	94.02

Some of the peculiar characteristics should be outlined: uniform particle size distribution, that allows an excellent uniformity and constancy of behaviour by mixing homogeneously and assuming the prerogatives of the products with which it is mixed, low apparent specific weight, low absorption, which allows a saving of binders, excellent whiteness.

Underlining that the average particle diameter for calcium carbonate in emulsion paints rounded 0.9 ÷ 70 micrometers, this physical property is fulfilled for both MS and GCC distribution, as shown in Fig. 2.

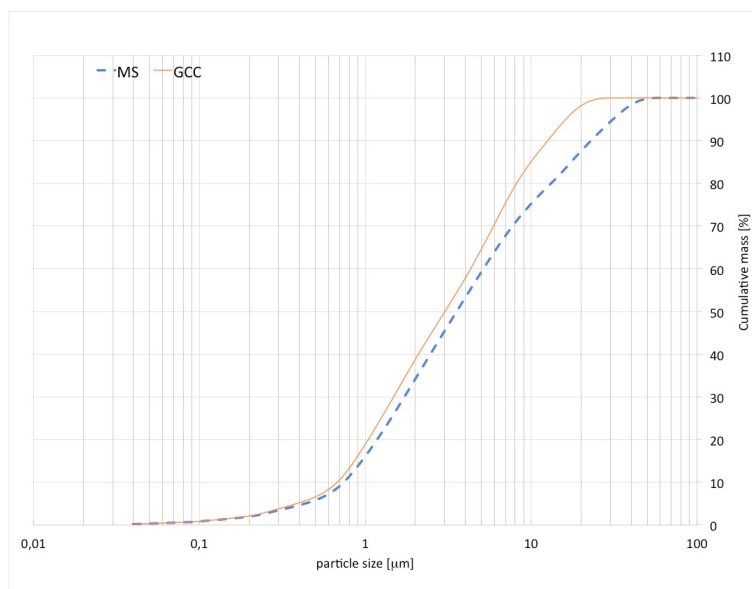
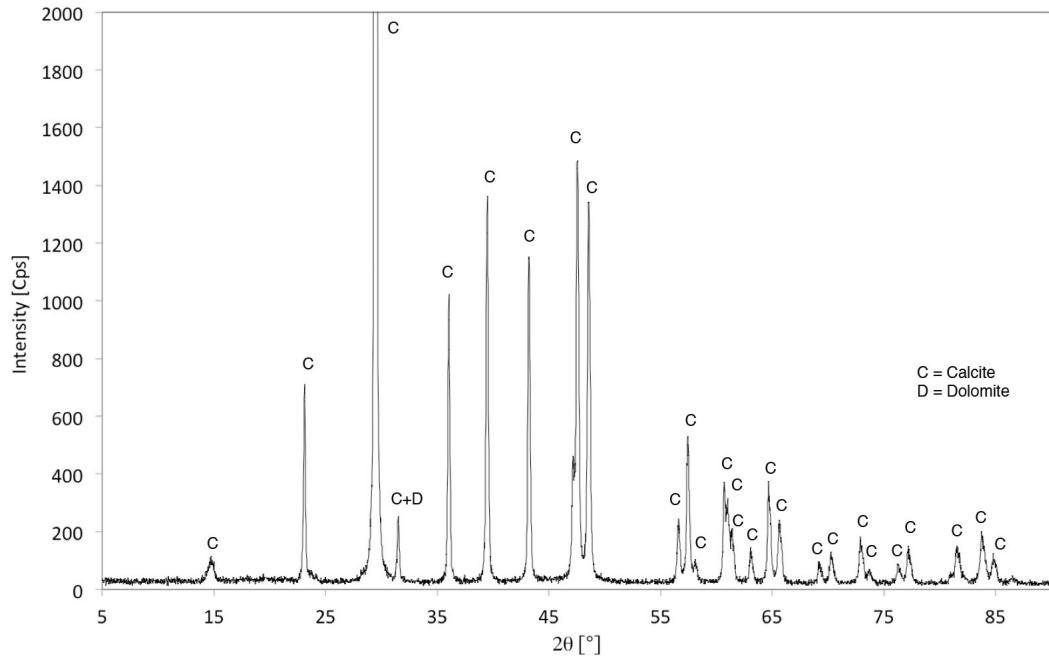


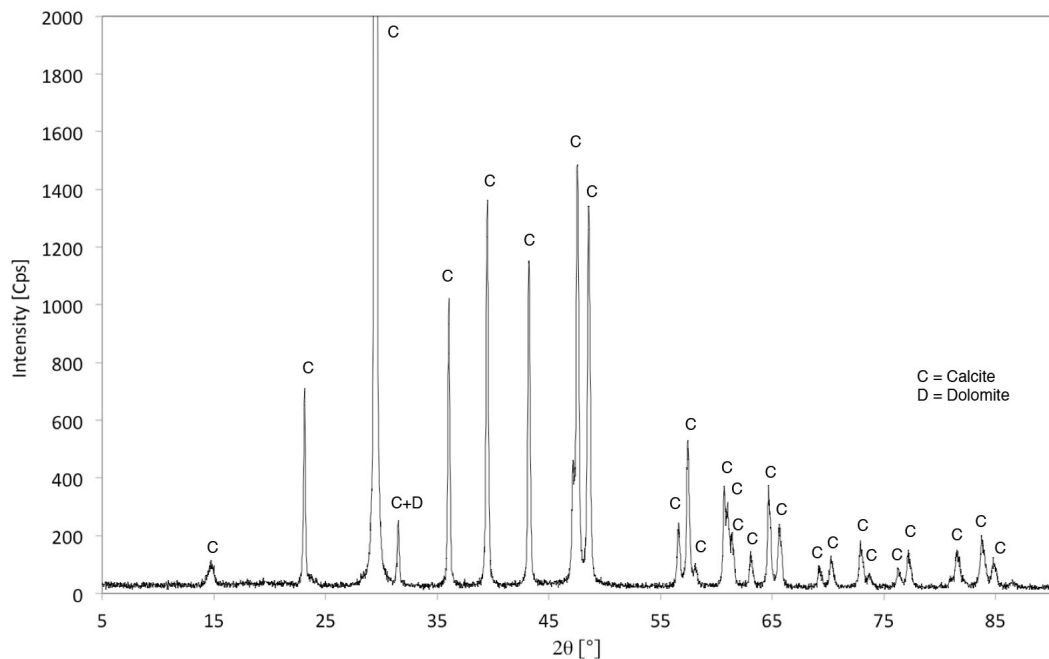
Figure 2. Granulometric distribution curves of MS and GCC

As verified by previous studies [33, 48], it is important to emphasize, that although there may be differences in the processing cycle of blocks and slabs (sawing, size-cutting, surface-finishing), the sludge always presents almost identical/very similar particle size curves.

Both MS and GCC exhibit a XRPD pattern where  $\text{CaCO}_3$  is the quite exclusive mineralogical phase apart from extremely small amount of dolomite in both samples (Fig. 3 a and b), highlighted by the presence of the  $I/I_0 = 1$  peak at  $d = 2.883 \text{ \AA}$  ( $31.62^\circ 2\theta$ ) while the other principal reflections (e.g.  $I/I_0 = 0.6$  and  $I/I_0 = 0.50$  peaks) are overlapped by the predominant  $\text{CaCO}_3$  phase. This mineralogical composition reflects the MS and GCC chemical composition (see Table 7).



a)



b)

Figure 3. XRPD pattern of GCC a) and MS b)

SEM image shows that both GCC and MS have particles from angular to sub-rounded. The dimensional distribution is quite bimodal ranging from micrometric and prevailing sub-micrometer in size (Fig. 4 a and b).

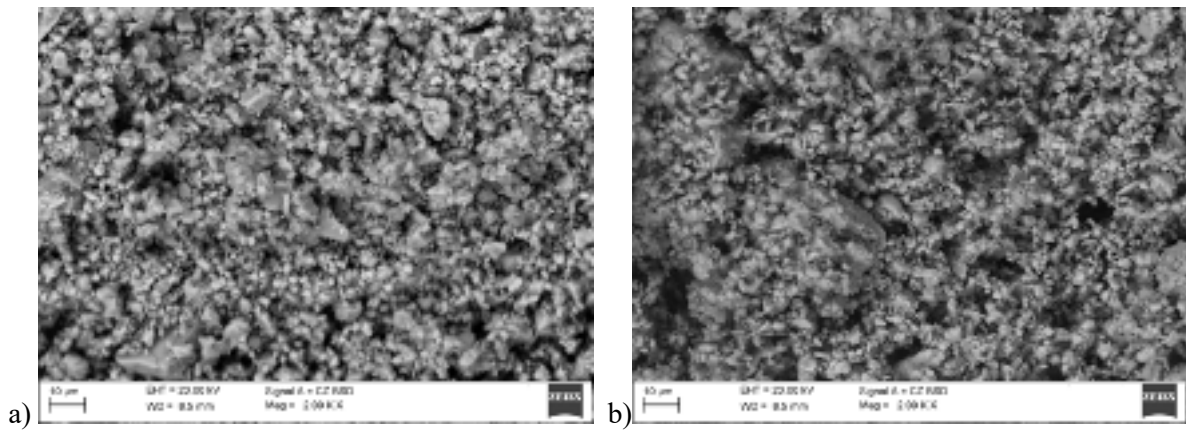


Figure 4. Back scattered electron images of GCC a) and MS b)

#### 4.2 Paint characterization and performance comparison

After verifying the similarity between GCC and MS, comparing the specifications for calcium carbonate used in paint industry, specific tests were carried out on paint samples as follows.

##### 4.2.1 Alkali Resistance

After the curing time, the coating surfaces were carefully observed and analyzed; the paint films still appeared intact, coherent and with no tendency to dissolve. Both the analyzed samples are classified as *alkali resistant* and they can be also applied on cement substrates.

##### 4.2.2 Paint hiding power

As shown in the data reported in Tab. 8, the tristimulus (Y) values, calculated from colorimetric data, make it possible to classify the ORO with a light improvement over EDI.

Table 8. Paint hiding power measurements data

		ORO		EDI	
		White	Black	White	Black
1	L*	95.65	93.11	96.28	93.45
	a*	-0.25	-0.52	-0.49	-0.58
	b*	2.52	0.55	1.49	-0.29
	Y	96.25	94.06	96.79	94.35
2	L*	95.64	93.87	96.23	93.32
	a*	-0.21	-0.48	-0.50	-0.58
	b*	2.61	0.96	1.48	-0.36
	Y	96.24	94.72	96.75	94.24
3	L*	95.62	94.11	96.22	93.46
	a*	-0.30	-0.45	-0.50	-0.58
	b*	2.45	1.18	1.49	-0.29
	Y	96.22	94.92	96.74	94.36
4	L*	95.55	93.37	96.15	93.69
	a*	-0.32	-0.50	-0.51	-0.57
	b*	2.49	0.74	1.48	-0.19
	Y	96.16	94.28	96.68	94.56
Mean	L*	95.62±0.05	93.62±0.46	96.22±0.05	93.48±0.15
	a*	-0.27±0.05	-0.49±0.03	-0.50±0.01	-0.58±0.01
	b*	2.52±0.07	0.86±0.27	1.49±0.05	-0.28±0.07
	Y	96.22±0.04	94.50±0.39	96.74±0.05	94.38±0.13
RC (%)		98.21		97.56	

Note: RC - contrast ratio

A better hiding power is to be ascribed to a good dispersion of the pigments in the binder media, because white pigments absorb incident visible light of all wavelengths.

The color of the paint appears to be affected by the color of the marble. However, since marble scrap is mostly derived from processing the commercial lithotype “Fiocco di neve”, which tends to white, the scrap also has a consistent color that tends to white [33].

#### 4.2.3 Dirt pick-up of the paint

The outcomes for EDI and ORO specimens, as obtained values average, are reported in Tab. 9. When comparing the dirt pick-up test results for EDI and ORO with the reference classification, paints can be categorized as: *very low dirt pick-up*.

Table 9. Paint dirt pick-up

		ORO		EDI	
		Clean	Dirty	Clean	Dirty
1	L*	95.50	93.90	96.39	95.08
	a*	0.06	0.17	-0.32	-0.22
	b*	3.50	3.49	2.28	2.23
2	L*	95.62	93.93	96.32	94.39
	a*	0.08	0.15	-0.32	-0.16
	b*	3.50	3.45	2.24	2.20
3	L*	95.58	93.73	96.34	94.28
	a*	0.07	0.08	-0.33	-0.16
	b*	3.50	3.15	2.24	2.18
4	L*	95.49	93.58	96.55	94.91
	a*	0.05	0.11	-0.18	-0.15
	b*	3.55	3.19	2.62	2.38
Mean	L*	95.55±0.06	93.62±0.16	96.40±0.10	94.67±0.39
	a*	0.07	0.13	-0.29	-0.17
	b*	3.51	3.32	2.35	2.25
ΔL		1.76		1.74	

#### 4.2.4 Wet scrub resistance

The classification for this specific parameter is reported in Tab. 10, both for EDI and ORO samples.

Table 10. Wet scrub resistance classification

Sample	Classification	Reference values
EDI	< 1000	Suitable for applications that do not require wet scrub resistance
ORO	< 1000	Suitable for applications that do not require wet scrub resistance

#### 4.2.5 Hand brushing test

The paints, equally EDI and ORO, were found to be easy to be brushed on a concrete background, presenting very similar characteristics in the application. The overall colour was also homogeneous and similar for both paints and such it retained, as confirmed by the visual observation carried out after a period of 2 years (Fig. 5). Furthermore, the paint looks well adherent and does not release material to the touch.

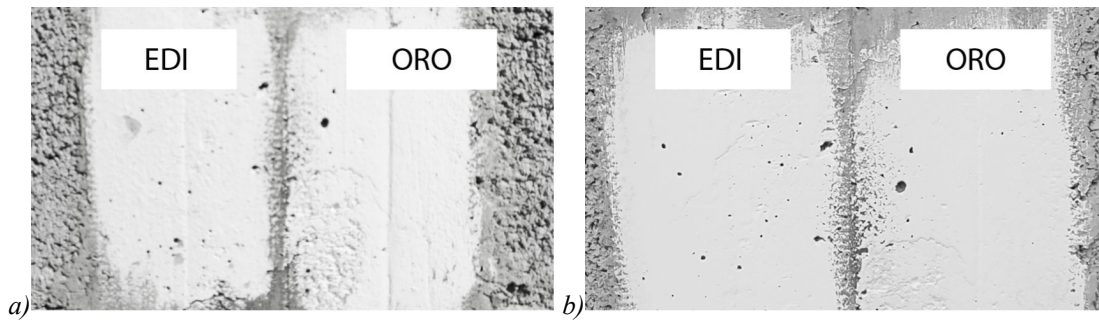


Figure 5. Photographs of the paint applications a) and after two years of exposition b)

## 5. Conclusion

In paint production, GCC is used as extender and filler to improve hiding power, brightness, washability and application properties, and to reduce consumption of expensive pigments, such as titanium dioxide.

The characterization phase showed the almost overlapping characteristics between MS and GCC. The specific tests carried out on the paints have given encouraging results on the reuse of marble waste in the industrial field of paints, specifically for white wall paints for interiors.

In some cases, the paints, made with sawdust, exhibited slightly better test performance than the one obtained for reference paint, i.e. reference parameters is very important, considering that it results in a better performance of the paint, and subsequent cost-saving.

In addition, to the time-passing, the analysed paint has shown good durability with regard to adhesion to the support and its coloration with respect to a visual evaluation.

It is important to note that water-based white paints are the most difficult to make because they are strongly affected by the color of the additives. In this study, it has emerged that the use of the Orosei marble sludge instead of commercially available calcium carbonate produces a paint with the same characteristics as the one on the market. It is therefore reasonable to argue that the same type of study can be applied to the production of colored water-based paints.

The idea that a large amount of material not only does not go to landfill, but also can find its way back into a suitable economic relocation is a very reassuring one, which might support the idea of sustainability as a corporate responsibility.

Our waste becomes a by-product that can be re-used by setting aside a slight and inexpensive pre-treatment on carbonate particles to counteract the compacting effect of the flocculant used in the filter press section. So, marble waste becomes a by-product.

The study is ongoing, so further applications in this area are being tested. A LCA analysis will be carried out at a later date.

Through the transformation of the produced waste, waste piles are reduced, degraded soils recovered, improving the land in terms of landscape and environment, in a perspective of circular economy.

The processing waste deriving from carbonate stone quarries will have a future.

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