INCREASED "GREEN VALUE" OF GLAZED PORCELAIN STONEWARE TILES THROUGH THE ECO-DESIGN OF RAW MATERIALS

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1. ABSTRACT

The aim of this research is to change the design paradigm for glazed porcelain stoneware ceramic bodies in order to rationalize the choice of raw materials and reduce the environmental impact and risks associated with supply sources.

The study was carried out using Life Cycle Assessment (LCA) and the eco-design approach to propose two innovative body formulations compared to the traditional one, preferring the use of local raw materials and transport by train.

Using a complete analysis of the environmental impact assessment of the ceramic process, from the extraction of raw materials to the finished product, it was possible to predict the environmental and technological performance of alternative scenarios and choose the best solution for testing in an industrial environment.

Some innovative aspects characterize this research: (1) the adoption of the LCA tool as a basis for eco-design, (2) the inclusion of the element of technological sustainability as a key element in decision making, (3) the simultaneous use of the simulation, laboratory and industrial environment for the validation of results.

This study has the limit of having validated the best body solution in an industrial environment in only two formats (30x60 and 60x120 cm). To verify the real effectiveness of the proposed design solution, it will therefore be necessary to carry out industrial testing in even larger formats.

2. INTRODUCTION

The ceramic industry is a materials and energy-intensive sector due to the manufacturing process itself which requires, in addition to mineral raw materials, huge volumes of water that evaporates during the production cycle and significant amounts of fuel for spray-drying, drying and firing [1].

Historically, the ceramic industrial districts of Sassuolo in Italy [2] and Castellón in Spain [3], were born and developed thanks to the proximity of the sources of supply of raw materials to the tile factories. Since the early 1990s this production paradigm, which was based on the use of local red firing clays, has changed: the market has begun to demand clear firing tiles. This has led to a review of production technologies, body formulations and the choice of sources of raw material supply. The producers had to look for raw materials with a low content in chromophore oxides (Fe_2O_3 and TiO_2) mainly outside the national area, mainly in Eurasia. Therefore, the consumption of extra-European clay and feldspar materials has grown rapidly to almost two decades, moving large quantities of minerals by sea and truck [4]. This phenomenon has led to the development of environmental regulations in a very restrictive sense, especially in Italy [5], whose regulatory framework for the ceramic industry has become the reference point for legislation in other European countries [1], primarily Spain [6]. Today, sustainability is seen as one of the great global challenges [7] and the intentions of environmental legislation are no longer seen as a series of prohibitions, but as an incentive to do better to improve the quality of life for all. Consistent with this new approach, the results of a recent environmental impact assessment study [8] on the Italian ceramic industry have shown that a careful choice of energy sources, both in thermal and electrical terms, is essential to reduce the overall environmental impact of the product. In addition to energy issues, an accurate selection of raw materials, considering their environmental impact, has the potential to significantly increase the final "green value" of the ceramic product. Starting from this result, an eco-design [9] approach is proposed to rationalize the raw material supply system from an environmental point of view, preferring the use of national natural resources and those delivered by train, which is the most environmentally friendly means of transport.

3. DESIGN OF THE CERAMIC BODY COMPOSITIONS

The aim of the eco-design applied in this study is to rebalance the quantity of imported raw materials, to the advantage of national and European ones, with the twofold intention of reducing the environmental impact due to the effect of transport and to mitigate the supply risk linked to the volatility of the prices of extra-EU raw materials and the unavailability of the volumes necessary to meet demand [10]. Table 1 shows the formulations of the bodies studied and their chemical compositions. The composition 1 is representative of a typical porcelain stoneware body [11] where the raw materials imported from outside the EU (which represent 65% of the total) have the function of giving the ceramic body a high plasticity (Illitic-kaolinitic clay) and good meltability (sodium feldspar). The remaining part of the body is made up of an Illitic-kaolinitic clay, a potash feldspar and a feldspar sand, both of national origin. With respect to the criteria of environmental sustainability, which penalizes factors such as the distance from the source of supply (extra-EU) and the type of transport (ship and road transport compared to the train), raw materials were indicated, considered to have a lower impact, as "*eco-raw-materials*", in order to proceed with the formulation of new bodies. In fact, domestic raw materials meet the criterion of the short distance between the factory and the mine, while EU clay uses rail transport.

SOURCES OF RAW MATERIAL SUPPLY		COMPOSITION 1	COMPOSITION 2	COMPOSITION 3
RAW MATERIALS	Extra-EU clay	30	15	0
	Extra-EU Na-feldspar	35	25	20
ECO-RAW-MATERIALS	EU clay	10	25	30
	National clay	0	5	25
	National K-feldspar	10	15	20
	National K-sand	15	15	5

OXIDES (wt%)	COMPOSITION 1	COMPOSITION 2	COMPOSITION 3
SiO_2	67,0	69,0	70,5
Al_2O_3	20,0	18,6	17,5
Fe_2O_3	0,9	1,0	1,0
TiO_2	0,6	0,7	0,6
MgO	0,4	0,4	0,4
CaO	0,6	0,5	0,5
Na_2O	4,2	3,1	2,5
K_2O	2,4	2,8	3,1
L.O.I.	4,3	4,1	3,9

Table 1*: Composition of the ceramic bodies studied and theoretical projection of the corresponding chemical analysis.*

Following this logic, two steps have been taken to improve the environmental performance of the body. Composition 2 provides for a significant reduction in extra-EU clay (15%) to the advantage of European clay (25%), in addition to the introduction of a national kaolinitic clay (5%). In the same way, extra-EU sodium feldspar has also been reduced (25%), partially replacing it with national potassium feldspar (15%). Composition 3, on the other hand, provided for a radical change: extra-EU clay was eliminated and replaced by European (30%) and national (20%) clays, as was the partial replacement of extra-EU sodium feldspar (20%) with national potassium feldspar (20%). The quantity of feldspar sand has also been reduced (even though it belongs to the eco-raw-materials category) to minimize the quantity of free silica in the form of quartz, since quartz in a non-free form (therefore not risky in the sintering phase of the ceramic body) is already made available by all the materials in appropriate quantities as shown by chemical analyses.

Chemical analyses also show a progressive rebalancing of the Na_2O/K_2O ratio [12] starting from the hypothesis, to be confirmed experimentally, that to obtain a high degree of vitrification of porcelain stoneware it is not necessary to go into sodium feldspar saturation, but it is better to optimize the quantities of albite and orthoclase to also have maximum dimensional stability.

4. LIFE CYCLE ASSESSMENT

The objective of the Life Cycle Assessment (LCA) study is the predictive environmental assessment [13] of the damage associated with the production process of glazed porcelain stoneware tiles by simulating the use of the three Compositions described above. In accordance with ISO 14040, which defines the methodological framework for life cycle assessment (LCA), the following parameters were adopted:

- **Function of the system**: covering of floors and walls of buildings;
- **System to be studied**: production of ceramic tiles;
- **Functional unit**: 1sqm of floor or wall covering of buildings;
- **Reference flow**: 1sqm of glazed porcelain stoneware tile;
- **System boundaries**: these range from the extraction of raw materials to produce porcelain stoneware tiles to the end of their life after 50 years of use; after use, it is assumed that the tiles will be recovered and used as a road surface.

SimaPro code 8.5.2.2 (PRé, 2018) was used to calculate the LCA. The data relating to the quantity of tiles produced, the mass of the mixture, water and energy consumption, emissions and waste were supplied by an Italian manufacturer of glazed porcelain stoneware. The IMPACT 2002+ method [14] was used to calculate the damage. It was modified for this research in order to make the system analysed more representative and to improve the final quality of the study results.

The results of the LCA analysis, simulating three different scenarios using Compositions 1, 2, and 3, indicate that the processes that contribute most to environmental damage are the procurement of raw materials for the mixture, the distribution of tiles to the customer, installation and maintenance. As far as the damage related to the supply of raw materials for the mixture is concerned, this is mainly due to the distances between the places where the raw materials are extracted and the production plants and to the transport system used.

Figure 1 shows, in a comparative way, the evolution of the damage produced when the composition of the ceramic mixture used for manufacture is modified. The diagrams show all the phases of the production process, and it is clear that the use of eco-raw-materials significantly reduces the environmental impact. Regarding the supply phase, the total environmental damage goes from 11.5 kPt (kilopoints) of Composition 1, to 9.5 kPt for Composition 2, to be reduced to 7.2 kPt for Composition 3.

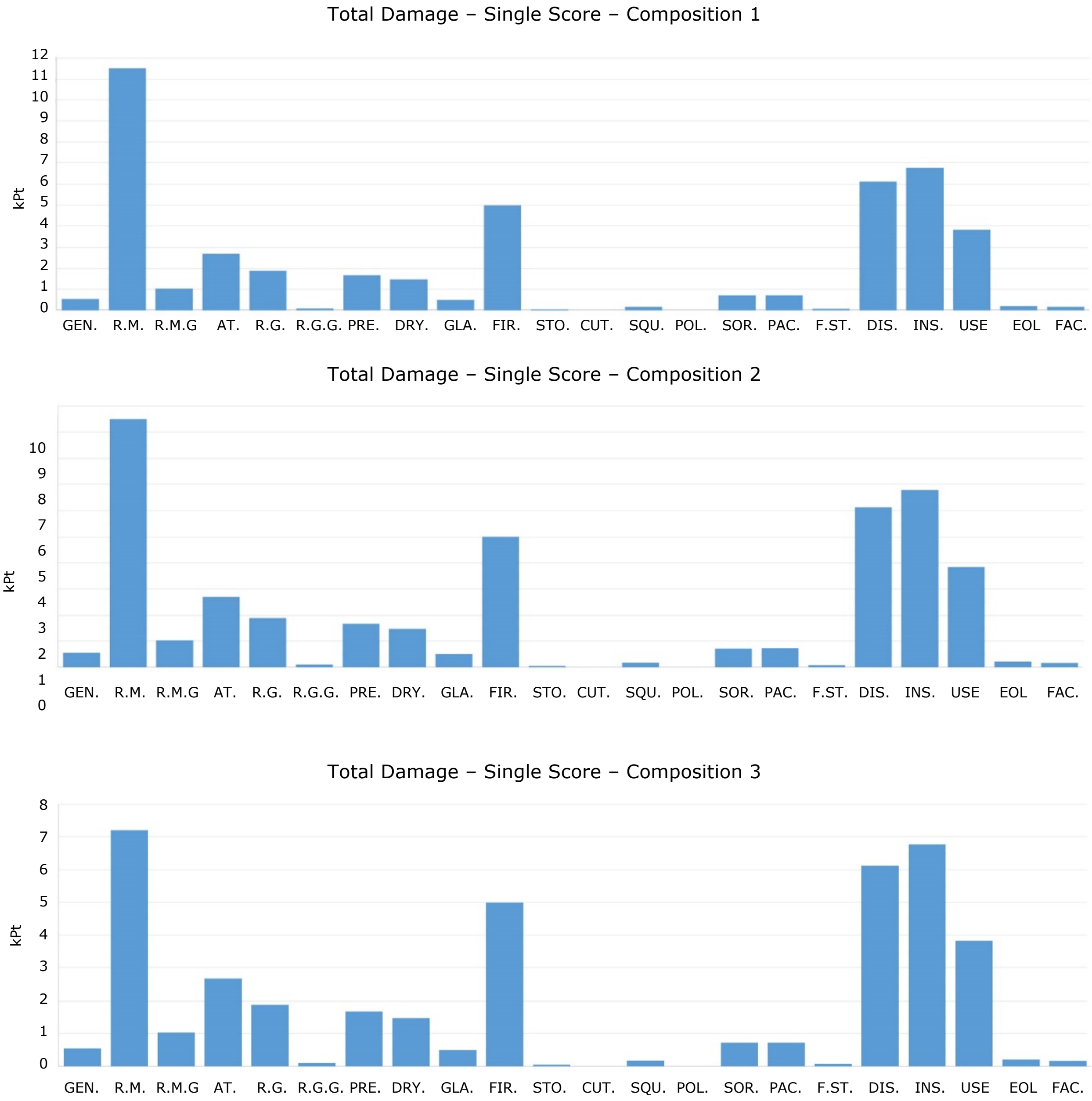

Figure 1: *Single Score graphs relating to the entire process according to the three simulated body compositions. [LEGENDA: GEN.: General; R.M.: Raw materials for the Mixture; R.M.G.: Grinding of Raw materials for the Mixture; AT.: Atomization; R.G.: Raw materials for the Glaze; R.G.G.: Grinding of Raw materials for the Glaze; PRE.: Pressing; DRY.: Drying; GLA.: Glazing; FIR.: Firing; STO.: Storage; CUT.: Cutting; SQU.: Squaring; POL.: Polishing; SOR.: Sorting; PAC.: Packaging, F.ST.: Final Storage; DIS.: Distribution; INS.: Installation; EOL: End of Life; FAC.: Factory]*

The IMPACT 2002+ impact assessment methodology foresees a combined midpoint/damage approach. It connects through 14 midpoint categories, all the process information collected during the inventory analysis phase (material flows, energy consumption, emissions, machinery characteristics), with four damage categories that are shown in Table 2. The midpoint categories evaluate the impacts earlier along the cause-effect chain in terms of, for example, acidification, eutrophication, global warming, etc. while the endpoint categories consider the effects at the end of the cause-effect chain, focusing for instance on human health, ecosystem quality, climate change, etc. The former provide more detailed results but they are more difficult to communicate and interpret; the latter do not require an in-depth knowledge in the field of environmental impacts and provide results that are easier to understand but less specific. "Human health" damage category evaluates the impact on the human health in terms of "DALY" ("Disability-Adjusted Life Years") that represent the years of life lost due to premature death and the time of life with lower quality due to an illness for example while "ecosystem quality" considers the impact on the ecosystem quality in terms of "PDF·m2·year" that represent the fraction of species disappeared (PDF) on 1sqm of earth surface during one year. "Climate change" damage category evaluates the impact on the climate change in terms of "kg CO_{2eq}", using carbon dioxide as a reference substance and "resources" takes into account the impact on the consumption of resources in terms of "MJ primary" related to the amount of energy extracted or needed to extract the resource. Finally, it is possible to aggregate the endpoint results in a single score, expressed in points, that provide a unique environmental result, particularly useful to make comparisons.

DAMAGE CATEGORIES	COMPOSITION 1	COMPOSITION 2	COMPOSITION 3
HUMAN HEALTH [DALY]	1,35E-05	1,27E-05	1,18E-05
ECOSYSTEM QUALITY [PDF*m^2*yr]	5,45E+00	5,24E+00	5,02E+00
CLIMATE CHANGE [kg CO_2 eq]	1,39E+01	1,33E+01	1,27E+01
RESOURCES [MJ primary]	2,60E+02	2,51E+02	2,42E+02
TOTAL DAMAGE [kPt]	**5,54E-06**	**5,30E-06**	**5,02E-06**

Table 2: *Environmental impact in relation to the four damage categories for each body composition and total impact, in accordance with the IMPACT 2002+ methodology.*

Overall, the total damage expressed in kPt decreases by 4% in Composition 2 and 9% in Composition 3 compared to Composition 1 of reference.

5. PREPARATION OF THE CERAMIC BODY

After the environmental sustainability analysis, the technological characteristics of the bodies were initially verified in a laboratory environment. The three ceramic body compositions were milled in a laboratory jar with a dry quantity of 65% and water of 35% to reproduce the industrial conditions, with the addition of 0.4% of solid fluidifying agent (sodium tripolyphosphate) up to a residue of 5% on a sieve at 45μ. The slips were dried and crushed, the powders moistened at 6% and pressed into 50 mm diameter discs at a pressure of 470 kg/cm^2. The samples were then fired in an industrial kiln for subsequent technological characterization as shown in Table 3 below.

CERAMIC BODIES COMPOSTITION	Wetness of the powders (%)	Loss on Ignition LOI (%)	Water Absorption (%)	Linear Shrinkage First Firing (%)	Linear Shrinkage Second Firing (%)	Linear Expansion D (%)
COMPOSITION 1	5,6	3,9	0,23	6,6	6,1	0,5
COMPOSITION 2	5,5	3,8	0,35	5	4,1	0,9
COMPOSITION 3	5,7	3,7	0,26	5,6	5,5	0,1

Table 3*: Technological parameters of the ceramic bodies studied (Bullers temperature 1100°C).*

For each test composition the humidity of the powders was determined before pressing, and after firing LOI, water absorption and linear shrinkage were measured. In addition, linear expansion was determined to test firing behaviour and above all dimensional stability, which is very important to produce large rectangular formats. After firing, the samples were re-fired under the same operating conditions to determine the shrinkage variations: the difference between the shrinkages in the first and second firings indicates the expansion of the ceramic body. The closer the expansion is to zero, the more stable the body is to be considered.

The data show that Composition 3 is comparable to the initial Composition 1 in terms of sintering [15], as it has a water absorption similar to and suitable for the type of glazed porcelain stoneware; moreover, the shrinkage is lower and the expansion almost zero. This shows that the optimization of the ratio between Na_2O/K_2O can allow excellent sintering results without having to resort to large quantities of sodium feldspar. Composition 2, on the other hand, is characterized by higher absorption and, above all, by a greater tendency towards linear expansion [16].

No mechanical strength tests were carried out on the test samples before and after firing due to the difficulty of carrying out these analyses on laboratory samples; it was therefore decided to investigate this aspect through industrial experimentation with the manufacture of a significant quantity of ceramic tiles.

Table 4 below shows the results obtained from the mineralogical analysis of the crystalline part (semi-quantitative) carried out with the Rietveld method on each composition after firing. Qualitatively, all samples contain albite and quartz as residual phases, and mullite as a neo-formation phase. Traces of rutile and cristobalite are also detected in Compositions 1 and 2. It can be seen that the amount of residual albite is higher in Composition 2 than in other cases, thus demonstrating that a similar result (Composition 3) can be obtained without saturating the body with sodium feldspar.

CERAMIC BODIES COMPOSTION	ALBITE Wt. (%)	MULLITE Wt. (%)	QUARTZ Wt. (%)	RUTILE Wt. (%)	CRISTOBALITE Wt. (%)
COMPOSITION 1	8,1	24,6	67,1	<1	<1
COMPOSITION 2	7,9	22,5	69,3	<1	<1
COMPOSITION 3	7,6	24,3	67,8	-	-

Table 4: *Mineralogical composition (% by weight) of the crystalline part.*

6. TESTING IN INDUSTRIAL ENVIRONMENT

The previous phase of eco-design has shown that Composition 3 represents the best solution to pursue a triple objective:

1) reduce the distances and risk of the sources of supply;
2) favour rail over ship transport;
3) improve the environmental performance of current production.

Composition 3, while showing improved environmental sustainability indexes, must also have technological performance features comparable to those of the production body with Composition 1, to be confirmed through an industrial test. To be meaningful, it requires grinding and spray-drying of no less than 120 tons of ceramic body, to allow optimal adjustment of the machines with the parameters of current production. Composition 3 was then ground, and the slip stored in an aerial steel tank to be spray dried. Table 5 shows the characteristics of the slip and the particle size of the spray-dried powder obtained, compared to the production body with Composition 1.

The rheological properties of the new body under test are fully comparable to that of production, as well as the particle size distribution of the two spray-dried products. The table also indicates, for each particle size class, the range of acceptability provided by the quality protocol of the company where the test was conducted. It can therefore be concluded that the spray-dried powder of the test body conforms to the company's quality standards and is comparable to current production.

CERAMIC SLIP	COMPOSITION 1	COMPOSITION 3
Residue on 45*m* (%)	6,2	6,0
Viscosity (seconds)	28	23
Density (g/l)	1699	1706

SPRAY-DRIED CERAMIC BODIES			
Particle Size Classes	Standard Distribution (%)	COMPOSITION 1	COMPOSITION 3
0,710	**10 ÷ 15**	9,9	12,7
0,400	**35 ÷ 45**	30,0	33,5
0,315	**20 ÷ 30**	25,0	28,0
0,250	**5 ÷ 15**	13,8	11,5
0,180	**5 ÷ 10**	13,4	9,2
0,125	**3 ÷ 5**	6,0	3,8
0,090	**0,3 ÷ 0,9**	1,0	0,3
FINAL	**0,5 ÷ 1,5**	0,9	1,0
Humidity	**7,0 ÷ 7,5**	6,9	7,8

***Table 5**: Technological characteristics of slips and spray-dried powders.*

Size 30x60 cm	COMPOSITION 1	COMPOSITION 3
Nominal thickness (mm)	8,2	8,2
Short side caliber (mm)	300,0	300,1
Long side caliber (mm)	602,6	603,6
Water absorption (%)	0,28	0,21
Breaking load (N)	802	853
Breaking strength (N)	1551	1648
Bending resistance (N/mm^2)	53	55
Coefficient of linear thermal expansion	$7,9 \times 10^{-5}$ K^{-1}	$8,1 \times 10^{-5}$ K^{-1}

Size 60x120 cm	COMPOSITION 1	COMPOSITION 3
Nominal thickness (mm)	10,0	10,0
Short side caliber (mm)	608,2	609,5
Long side caliber (mm)	1211,8	1214,8
Water absorption (%)	0,22	0,08
Breaking load (N)	1062	1204
Breaking strength (N)	2079	2366
Bending resistance (N/mm^2)	48	55
Coefficient of linear thermal expansion	$8,3 \times 10^{-5}$ K^{-1}	$8,6 \times 10^{-5}$ K^{-1}

***Table 6**: Technological characteristics of the pressed bodies.*

The spray-dried powder of Composition 3 was subsequently pressed, glazed and fired in two rectangular sizes: 30x60 and 60x120 cm. The choice of the rectangular format compared to the square was adopted to better highlight any geometric and/or mechanical problems. The results of the characterizations on pressed and fired tiles in the two formats are detailed in Table 6. Pressing and firing tests in an industrial environment confirm the data obtained in the laboratory: the lower linear shrinkage translates into a longer caliber compared to current production in both the 30x60 and 60x120 cm formats. The mechanical properties are slightly better than production and comply with international standards for porcelain stoneware tiles. The coefficient of linear expansion can also be considered similar between the two compositions. During the production of a total of about 6000 square meters in the two tile formats, there were no increases in breakage of the pieces during handling and decoration, compared to the current production. This behaviour indicates that the plasticity of the new body is still adequate for the production standards.

7. CONCLUSIONS

This study was aimed at assessing the environmental sustainability of porcelain stoneware tiles considering different scenarios for the supply of raw materials, in order to evaluate whether and to what extent the use of local and nearby raw materials could improve the environmental performances of the tiles. An important and non-secondary benefit of this eco-design approach is the valorization of alternative raw materials not considered before.

Based on new ceramic body formulations, different life cycle processes have been modelled, considering the different ceramic body compositions and the relative supply of raw materials, without changing the other phases along the life cycle; this in order to evaluate the incidence of the raw materials on the results. Three alternatives with 63%, 42% and 20% of extra-EU raw materials respectively have been compared, with their relative means of transport. The environmental impact of the alternatives was assessed with the Life Cycle Assessment methodology, considering the entire life cycle of the tiles. The results highlighted that the scenario with 63% of extra-EU raw materials had the higher impacts, decreasing for the second and third scenarios by 2.8% and 9.1% respectively.

As the second step of the analysis, the three different compositional alternatives were analysed from a chemical and microstructural point of view, with the aim of ensuring the technical and technological feasibility of the product and its final performances. The samples were obtained both on laboratory and industrial scale by standard powder processing routes and sintering conditions (1200 °C, 45 min) and technological parameters were measured in order to assure compliance with the requirements of the BIa class.

This analysis underlined the importance of the design phase of the ceramic body, which can reduce the overall environmental damage due to shorter supply distances of raw materials and more sustainable types of transportation (i.e. train). Both technological and environmental sustainability are fundamental to ensure the feasibility of a product and represent a guide towards the design and manufacturing of more sustainable products.

This research has shown empirically, with laboratory tests and industrial production, that by changing the traditional design logic of ceramic mixes, it is possible to minimize the use of extra-EU raw materials which, in addition to being a source of critical supply, also have a negative effect on the environmental impact of the entire process.

Experimental tests have also shown that a high level of sintering can be achieved by optimizing the Na_2O/K_2O ratio in the mix, making the over-dosage of sodium feldspar superfluous. This technical result therefore allows the introduction of locally sourced feldspars and clay materials into the compositions.

Even the complete elimination of the very plastic clays of extra-EU origin did not compromise the mechanical performance of the unfired products. The complete digitization of the decoration, without subjecting the tiles to mechanical stress like traditional silk-screen printing decoration systems, makes it less necessary than in the past to have very plastic unfired and dried tile bodies with high mechanical strength. Composition 3, which can be considered eco-friendlier, nevertheless requires further industrial testing in sizes larger than the 60x120 cm tested, in order to verify whether the mechanical performance and dimensional stability are confirmed.

Finally, the eco-design approach has proved to be a powerful tool for developing ceramic products with a higher environmental sustainability index, drawing different scenarios that can be evaluated as alternative strategic options.

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