

Sustainable production of ceramic products: the use of by-products in traditional ceramic mixture

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Abstract: The ceramic industry is capable of reusing many of the by-products generated during the production, within its processes. The by-products adopted in the ceramic industry include raw and fired tile scraps, exhausted lime, washing line sludge, polishing sludge, and dried milling residues. Recent developments and innovations in the ceramic production industry allow to replace raw materials with such by-products, in a circular economy perspective. The literature analysis shows that several studies have been conducted to combine different by-products in the ceramic mixture preserving the technical and the mechanical performances of the ceramic products. The purpose of this research is to identify the most relevant by-products allowing the sustainable production of the ceramic products. The adoption of a sustainable ceramic mixture using specific by-products allows the reduction of greenhouse gas emissions (mainly carbon dioxide) requiring lower energy for the production process. However, several by-products require a pre-treatment phase before the addition into the ceramic mixtures with higher energy consumption and economic charge. This paper proposes the definition of a framework that summarizes the advantages and the disadvantages of the sustainable production of ceramic products leading the selection of the most suitable by-product for a specific sustainable ceramic mixture. This selection is supported by the evaluation of the technical and the mechanical performances and the aesthetical characteristics of the ceramic product, the energy performance of the production process and the pre-treatment phase on the by-product. Future developments will concern the selection of specific by-products for the laboratory production of a sustainable ceramic mixture in order to analyse the main characteristics.

Keywords: ceramic industry; circular economy; sustainable production.

I. INTRODUCTION

The problems related to the scarcity of resources, the exploitation of non-renewable energy sources, the complexity of using renewable sources to preserve natural ecosystems, represent topical issues. The inefficient use of raw materials and energy from non-renewable sources are the basis of a production system that is no longer sustainable, which requires a shift from the traditional linear economy to the more sustainable circular economy [1]. According to this perspective, waste acquires a new value as by-product in a new production process [2]. Environmental sustainability is closely related to the cautious use of natural resources. Giving new life to waste using recycled materials characterized by high performance is a process based on the principles of the circular economy. The approach is feasible to any production system and aims at minimizing the consumption of raw materials and the amount of waste in the production processes.

The scheme of reuse, recovery, re-manufacturing and recycling extends the waste life and functionality as much as possible, maximizing its value, reducing the consumption of energy, eliminating the disposal costs, as well as the definition of new opportunities for innovating and optimizing the production processes [4]. The ceramic industry represents a model considering sustainable and circular economy. The reuse of the by-products generated during the production within its process includes raw and fired tile scraps, exhausted lime, washing line sludge, polishing sludge, and dried milling residues. Recent developments and innovations in the ceramic production technologies allow to replace raw materials and natural resources with such by-products in a circular perspective. Also, the adoption of a circular approach to produce ceramic products reduces the intensity of other activities, including the extraction of natural resources and the transport of materials [3]. Several studies focus

on the reuse of by-products in order to preserve the technical and the mechanical performances of the ceramic products. The reuse of by-products in the ceramic production process allows the obtaining of a sustainable ceramic mixture and the realization of innovative and ecological ceramic products. Also, the reuse of specific by-products determines the reduction in energy consumption and greenhouse gas emissions of the ceramic production process. However, a large number of by-products requires a pre-treatment phase before the addition in the ceramic mixture, i.e. the thermal pre-treatment. This pre-treatment implies higher energy consumption that contributes to the increasing costs of the ceramic production process reducing the beneficial effects related to the use of by-products. This paper aims at defining the main by-products to insert in the ceramic mixtures comparing the advantages and the disadvantages of the sustainable production of the ceramic products.

II. LITERATURE REVIEW

The purpose of this research is the definition of a framework to identify the most relevant by-products generated during different production processes allowing the sustainable production of the ceramic products. The ceramic industry reuses the by-products generated during the production within its process in a sustainable perspective. Following the circular economy principles, raw materials and natural resources commonly used in the ceramic production process can be replaced with such by-products considered as industrial waste. The approach allows the minimization in raw materials consumption and the reduction of the industrial waste production. However, the execution of a transformation pre-treatment phase must be considered for several by-products before the addition in the ceramic mixture, representing an additional economic charge to the ceramic production process. Thus, the transformation pre-treatment reduces the beneficial effects associated with the sustainable production of the ceramic products. The framework proposed in this research consists in a path for each by-product inserted in a ceramic mixture considering the advantages and the disadvantages related to the sustainable ceramic production. The definition of the most suitable by-products to insert in a specific sustainable ceramic mixture leads to the obtaining of innovative and ecological ceramic products.

In January 2023, the number of papers published between 2001 and 2022 and available in the Elsevier’s database and Scopus database

considering the keywords “innovative ceramic mixture” and “industrial waste in ceramic industry” is respectively 14,523 and 28,151. A sample of 65 papers have been selected among the publications in the recent literature to include research studies, conference proceedings, reviews, guidelines, research projects and reports with the focus on the addition of specific waste materials. Specially, hazardous waste difficult to manage and waste produced in huge quantities in different industrial production processes were studied to select the main by-products for the formulation of sustainable ceramic mixtures. Considering the complexity in following the landfill disposal chain of hazardous waste and the evaluation of the amount of waste produced in different industrial production processes, the recycled materials selected in this analysis are cement-asbestos, man-made vitreous fibres, fly ash and bottom ash, glass scraps and exhausted foundry sands.

Cement-asbestos waste must be treated with an inertisation process to be used as by-product. Thermal pre-treatment removes the dangerous characteristics from this material. Thermally inerted cement-asbestos shows a phase composition similar to natural clinker with a higher content of aluminium, iron and magnesium [5, 6]. The scraps of man-made vitreous fibres, i.e. mineral wool and glass wool, are considered as hazardous waste due to the high breathability of the fibres. Therefore, the thermal inertisation is necessary to eliminate the dangerous properties of this by-product. The inerted fibres are similar to the materials with glassy characteristics and can be used as fluxes in the ceramic mixtures [7]. Fly ash and bottom ash considered in this research are the residues of the production processes inside the thermoelectric power plants and the municipal solid waste incinerators [8, 9, 10, 11]. Glass scraps of various origins can be used in the ceramic production process as a sintering promoting material. Various studies have proved that the use of this by-product in place of natural sand allows the reduction of the temperature normally applied to obtain the ceramic products [12, 13]. Foundry sands are used to realize molds or cores for casting iron-based alloys. High quality sands are generally considered, after the cooling of the molten material, the mold is broken to take the metal element and the foundry sand has completed its life cycle. The composition of the exhausted foundry sands may significantly vary according to the metallic melt. Hence, a preliminary phase of chemical characterization and particle-size analysis is necessary to identify the most suitable exhausted

foundry sand to insert in the ceramic mixture [14, 15, 16, 17].

The findings of the literature review refer to the definition of a sustainable ceramic mixture adding separately one by-product per time or a combination of different by-products. The sustainable ceramic mixture has been modified adding increasing quantities of recycled materials and reducing the amounts of natural resources to produce different ceramic products.

In this study, the technical and mechanical performances and the aesthetical characteristics of the ceramic products, the energy performances of the ceramic production process and the execution of a transformation pre-treatment on the by-products are considered in order to select the most suitable by-products to use for the definition of a sustainable ceramic mixture and the production of innovative and ecological ceramic products.

III. MATERIALS AND METHOD

The ceramic products are obtained from non-metallic inorganic raw materials mixed and processed by forming and heat treatment. The ceramic products represent a variety of goods with different applications depending on the characteristics of raw materials selected for the mixture and the technological production process. Raw materials involved in the process are clay, sand, kaolin and alumina, degreasers to reduce plasticity and facilitate the cohesion of the elements during the heat treatment, fluxes which reduce the level of refractoriness, raw materials for the glaze applied on the surface. The technological production process consists in the mixture preparation led to the final use of the product, forming and dehumidification to reduce the water content, firing to compact the mixture, glazing and superficial decoration. The chemical composition and the technological production process determine the porosity and the mechanical properties, impermeability, frost resistance and technical properties, as well as the aesthetic characteristics of the ceramic products. The aim of this research is to identify the main by-products generated during different production processes to obtain a sustainable ceramic mixture and support the reduction of the industrial waste production. The definition of a sustainable ceramic mixture allows the production of innovative and ecological ceramic products. The use of specific by-products also supports the reduction in energy consumption and greenhouse gas emissions, contributing to the decrement of the economic charge of the ceramic

production process. These by-products have been analysed in order to characterize the by-products themselves and evaluate the amount of the substitution.

Specifically, this paper refers to the replacement of raw materials with such by-products, i.e. cement-*asbestos*, man-made vitreous fibres, fly ash and bottom ash, glass scraps and exhausted foundry sands.

The information obtained in this research are collected considering: (i) the typologies of by-product added to the ceramic mixture and the percentage of the substitution; (ii) the execution of a pre-treatment phase on the by-product; (iii) the typologies of ceramic product in which the by-product insertion was tested; (iv) the main results focusing on the technical and the mechanical performances and the aesthetical characteristics of the ceramic products; (v) the energy performance of the ceramic production process.

The technical and the mechanical performances of the ceramic products are assessed measuring linear shrinkage, water absorption, bulk density and rupture modulus. These indicators determine information on the sintering process of the samples. Sintering is a densification process aimed at removing interstitial porosity and allowing the development of bonds between adjacent particles. Linear shrinkage is a measure of the percentage dimensional variation of the samples after sintering. Water absorption and bulk density refer to the quality of the samples as it is strictly related to the compactness and therefore to the mechanical strength. Rupture modulus represents the limit load supported without cracking. The aesthetical characteristics are evaluated comparing the difference in colour between the samples and the reference product. Finally, the energy performances of the ceramic production process are analysed considering time and temperature of the heat treatment.

IV. RESULTS AND DISCUSSION

This section introduces the results of the literature analysis on the production of innovative and ecological ceramic products obtained introducing various recycled materials in different ceramic mixtures. The aim is the selection of the most suitable industrial by-products for the formulation of sustainable ceramic mixtures. The information collected on the main by-products, the innovative ceramic products, and the variation on the ceramic production process are considered to define a framework shown in Figure 1.

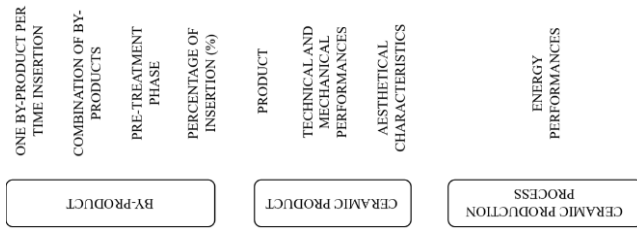


Figure 1. Structure of the framework considering the information collected on the by-products, the ceramic products and the ceramic production process

The addition of one by-product per time or a combination of by-products, the percentage of the insertion and the requirement of a pre-treatment phase on the by-products are reported in the first section. The second section refers to the typology of ceramic product realized through the substitution, the mechanical and the technical performances and the aesthetical characteristics of the ceramic product. Finally, the third section deals with the energy performances of the ceramic production process.

The main by-products considered in this research are cement-asbestos, man-made vitreous fibres, fly ash and bottom ash, glass scraps and exhausted foundry sands. Many of these selected by-products are classified as hazardous waste, contributing considerably to the increasing amount of waste to dispose. Figure 2 collects the results about the insertion in the porcelain stoneware mixture of cement-asbestos, a combination of cement-asbestos and glass scraps, and man-made vitreous fibres.

Cement-asbestos and man-made vitreous fibres must be subjected to a thermal pre-treatment. The danger of using fibrous materials is due to the high breathability of the fibres which can lead serious consequences on health of the operators working with this material. The thermal pre-treatment leads the transformation of the hazardous fibres, which lose the typical fibrous conformation allowing the reuse as by-product. The firing temperature of the thermal transformation is about 1200-1300°C. This process represents a valid alternative to the hazardous waste disposal. However, it contributes to the increasing costs of the ceramic products as the thermal pre-treatment implies an increase in energy consumption of the ceramic production process and consequently higher greenhouse gas emissions. These factors reduce the beneficial effects associated with minimizing the waste sending to landfill sites.

Adding increasing quantities of inerted cement-asbestos (3, 5, 10% wt) replacing quartz sand to

the porcelain stoneware mixture, the mechanical and the technical performances are preserved [6]. Cement-asbestos and glass scraps have been used for the production of frits (1, 3, 5% wt) to insert in the porcelain stoneware mixture. Specifically, the addition of 5% wt of the new frit led to better mechanical strength, stain resistance and higher productivity [5].

Linear shrinkage, bulk density, water absorption, open porosity and crack resistance were tested after the insertion of inerted man-made vitreous fibres (3, 6, 9% wt) in the porcelain stoneware mixture replacing fluxes. The mechanical and the technical performances of the samples are comparable to the reference product. Furthermore, the addition of inerted man-made vitreous fibres allows the reduction of the firing temperature by 40°C [7].

Reducing the firing temperature, the economic charge related to the energy consumption decrease. Also the greenhouse gas emissions connected to the ceramic production process are reduced. Finally, the aesthetical characteristics of the samples obtained with the substitution of cement-asbestos and man-made vitreous fibres differ from the reference product. The use of these by-products leads the realization of darker samples.

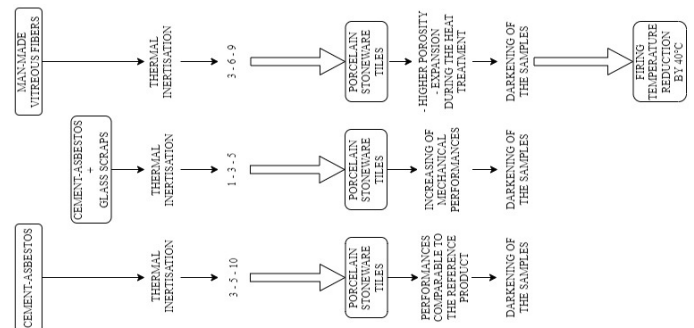


Figure 2. Framework related to the use of cement-asbestos and man-made vitreous fibres in the porcelain stoneware mixture

Fly ash and bottom ash are generated in various production processes and are considered singularly or in combination with other by-products, i.e. furnace blast slag. The results are reported in Figure 3.

Coal fly ash shows properties similar to quartz and can be added to the ceramic mixtures as untreated by-product or after an alkali activation pre-treatment. Treated coal fly ash replacing feldspar in the porcelain stoneware mixture (15, 30, 50% wt) exhibits low firing temperature with large sintering range. Specifically, the samples show optimal post-sintering properties by the temperature of 1100°C [8]. Reducing the sintering

temperature, firing time and temperature are reduced obtaining samples with good mechanical and technical performances. These interesting results lead the reduction of the costs connected to the energy consumption of the production process, allowing the minimization of the greenhouse gas emissions. However, the samples are darker than the reference product.

A combination of fly ash from thermal power plant and blast furnace slag from steel plant (25, 30% wt) replaces respectively quartz and feldspar for the realization of porcelain stoneware mixture. Linear shrinkage, bulk density, water absorption and flexural strength were measured recording beneficial effects by the temperature of 1175°C rather than 1200°C. Increasing temperature, the flexural strength drastically decreases due to the formation of glassy phases [9]. The results suggest that the appropriate regulation of firing time and temperature favours sintering avoiding excessive vitreous formation phenomena and the resistance reduction due to the higher fusibility of the mixture.

Fly ash and bottom ash from the municipal solid waste incineration (MSI) can be inserted in the ceramic mixtures both as untreated by-product or as vitrified by-product. The mineralogical and microstructural investigations must be conducted in order to define the chemical characteristics and the possible presence of hazardous fractions. MSI fly ash is considered for the realization of the porcelain stoneware mixture (10% wt) replacing clay and feldspar. The mechanical and the technical performances of the samples are comparable to the reference product [10]. MSI bottom ash is used both treated and untreated in the porcelain stoneware mixture and in porous single firing mixture. Water absorption and linear shrinkage have been measured. Untreated MSI bottom ash, up to 5% wt, does not affect these indicators. Treated MSI bottom ash (5, 10% wt), inserted as a strong flux, favours the realization of porous single firing samples with mechanical and technical performances comparable to the reference product.

Considering the porcelain stoneware mixture, treated MSI bottom ash favours the sintering process allowing the reduction of firing time and temperature, implying the reduction in energy consumption and greenhouse gas emissions. Furthermore, the leaching tests confirm the non-hazardousness of the samples [11].

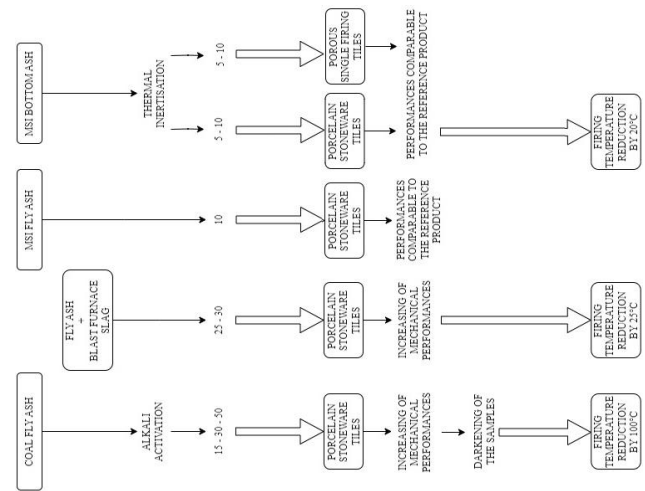


Figure 3. Framework related to the use of fly ash and bottom ash in different ceramic mixtures

The use of glass scraps of various origin as by-product replacing raw materials commonly used to formulate ceramic mixtures is reported in Figure 4. Glass scraps are a valid alternative to fluxes representing a sintering promoting by-product. Various studies have proved that the use of glass scraps replacing natural sand allows the reduction of the firing temperature with economic savings in energy. The advantage of reducing the firing temperature is associated to the environmental sustainability thanks to the reduction of greenhouse gas emissions. The introduction of glass scraps is applied to the production of various ceramic mixtures, i.e. porcelain stoneware and bricks. The insertion of soda-lime-silica glass and borosilicate glass up to 41% wt is evaluated for the production of porcelain stoneware mixture. The samples are highly vitrified at a firing temperature almost 140°C lower than the normally temperature applied to the process [13]. Natural sand has been replaced by glass scraps up to 20% in ceramic tiles and bricks mixtures. Density, water absorption and flexural strength improve with the addition of glass scraps that contributes to the porosity reduction due to the vitreous phase formation. Thus, faster sintering allows the reduction of the firing temperature by 200°C [12].

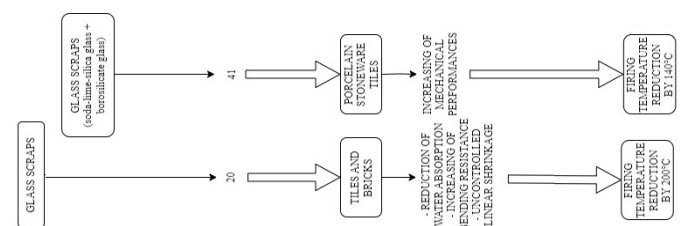


Figure 4. Framework related to the use of glass scraps in different ceramic mixtures

Exhausted foundry sand is a high quality by-product for the substitution of raw materials

commonly used in the ceramic production process. Chemical characterization and particle-size analysis lead the usability evaluation of various exhausted foundry sands in the ceramic mixtures. Figure 5 collects the results about the insertion of exhausted foundry sand singularly or in combination with glass scraps and sewage sludge in different ceramic mixtures.

The porcelain stoneware samples obtained by replacing clay and kaolin with foundry sand up to 15% wt show higher density, linear shrinkage and fire resistance. Higher water absorption and plasticity reduction are also detected. The colour of the samples is slightly darker than the reference material [14]. Interesting results are achieved considering the substitution of clay with 15% of foundry sand in the porcelain stoneware mixture. The reduction of the firing temperature by 50°C leads the reduction in energy consumption and greenhouse gas emissions [16]. Foundry sands can be also used for the production of bricks and tiles replacing clay with increasing percentages up to 50%. The innovative samples meet the requirement for the commercial products. Decreasing the sintering temperature, the volume variations decrease, as well as the weight loss. Consequently, the mechanical performances of the samples and the productivity of the ceramic production process are improved [15]. A combination of foundry sand, glass scraps and sewage sludge from waste water treatment is considered for the formulation of the porcelain stoneware mixture. Glass scraps has positive effects on flexural strength, water absorption and firing weight loss, while foundry sand contributes to reducing linear shrinkage. The combination of 10-15% foundry sand, 15-20% glass scraps and 10% sewage sludge, at the firing temperature of 1000-1050°C, is the most suitable for the production of samples with performances comparable to the reference product [17].

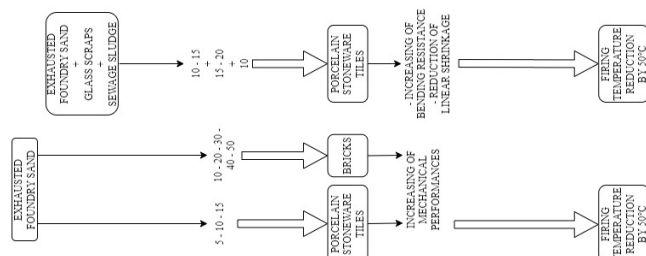


Figure 5. Framework related to the use of exhausted foundry sand in different ceramic mixtures

Finally, the chemical characterization of the various exhausted foundry sands is useful to identify the by-product with the best characteristics to be used in the ceramic production process. The

selected by-product can be added both separately or in combination with other by-products to formulate different ceramic mixtures. Generally, the samples show balanced technical, mechanical and aesthetic performances. The faster sintering determines the possibility to reduce firing time and temperature and consequently involve economic and environmental advantages.

V. CONCLUSION

This paper introduces the results of a literature analysis concerning the advantages and the disadvantages of the insertion in the ceramic mixtures of various industrial by-products. The collection of data allows the definition of a framework that summarizes the advantages and the disadvantages of the sustainable production of the ceramic products. The definition of the most suitable by-product to insert in a specific sustainable ceramic mixture is supported by the evaluation of the typologies of by-product and the percentage of the substitution, the execution of a pre-treatment phase on the by-product, the typologies of innovative and ecological ceramic product and its properties, the energy performance of the ceramic production process. The by-products selected in this research are cement-asbestos, man-made vitreous fibres, fly ash and bottom ash, glass scraps and exhausted foundry sands. The direct use of by-products in different production processes is rarely possible. The transformation pre-treatment is generally required, mainly the thermal inertization of hazardous by-products, in order to ensure the feasibility of reuse. The thermal pre-treatment of cement-asbestos and man-made vitreous fibres provides a valid alternative to landfill disposal of hazardous waste. However, it represents an additional step to the production process with higher economic charge, contributing to the increasing costs of the by-product. Furthermore, the thermal pre-treatment requires higher energy consumption affecting adversely on environmental sustainability due to the higher greenhouse gas emissions.

These consequences reduce the beneficial effects of reusing the by-products instead of sending waste to landfill sites. The chemical characterization of fly ash and bottom ash is essential to identify any hazardous fractions affecting the toxicity level. Milling is a pre-treatment to ensure the suitable particle size fraction of glass scraps and exhausted foundry sand with lower impact on the ceramic production process and the environment. The insertion of sintering promoting by-products in the

ceramic mixture replacing fluxes allows to improve productivity and energy savings of the ceramic production process, reducing firing time and temperature, i.e. man-made vitreous fibres and glass scraps, treated fly ash and treated bottom ash. The economic and environmental benefits refer respectively to the decrement of the ceramic production costs and the greenhouse gas emissions. However, the firing process must be controlled through the correct setting of time and temperature to avoid the excessive fusibility of the ceramic mixture. The drawback refers to the aesthetical characteristics of the samples, which are darker than the reference product. Exhausted foundry sands represent a valid alternative to raw materials normally used for the formulation of ceramic mixtures. The chemical characterization is essential to evaluate the presence of metallic traces and select the most suitable exhausted foundry sand to match the technical and the mechanical performances and the aesthetical characteristics.

The results reported in this paper mainly refer to a laboratory production of ceramic samples. Therefore, limited range of evidence is available considering the applications to the industrial process. Several studies deal with the insertion of industrial by-products in the porcelain stoneware mixture. Porcelain stoneware products show excellent technical and mechanical properties and high application versatility. The commercial demand for porcelain stoneware products constantly increases, thus the high application for experiments concern the study of sustainable ceramic mixtures for the production of innovative and ecological porcelain stoneware products.

Generally, the replacement of raw materials with industrial by-products coming from different production processes does not completely solve the problem of the raw materials extraction. The beneficial effects associated with the reuse of by-products is strictly connected to the realization of innovative and ecological ceramic products formulating a sustainable ceramic mixture with materials regarded as waste to dispose. Future developments will concern the selection of specific by-products for the laboratory production of a sustainable ceramic mixture in order to analyse and produce innovative and ecological ceramic product.

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