



Characterization and processing of kaolin from southern Brazil for potential strategic application in the porcelain stoneware tiles manufacturing

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ABSTRACT

The ceramic industry in the productive hub of Santa Catarina needs some raw materials with specific characteristics such as kaolin to manufacture its products. This clay mineral with a naturally variable chemical composition in relation to its geolocation (in extraction area composed by rock and combined with sediment materials), attributes significant increases in transport costs due to its distance from the main productive areas in southern Brazil. Thus, as a strategic alternative, the improvement of kaolin with a high quartz content (69.9%) from the northern region of the state of Santa Catarina (~400 km from ceramic hub in the south of the state), more precisely from the municipality of Garuva, was sought. Unit processing operations by classifying this mineral for suitability for use in industry, resulted in an increase in the concentration of kaolinite. The results indicated that the proposed method reduced the amount of quartz and consequently increased the kaolinite content, resulting in an aluminum oxide content of approximately 28.20%, a value higher than the normal natural content of ~19%.

1. Introduction

Brazil is one of the main global market players for ceramic tiles, ranking third in production, second in the consumption, and seventh in the export rankings (ANFACER, 2022). To meet the high demand in this field, a significant volume of natural raw materials is required to increase the production (Dondi et al., 2021). Among the materials developed and commercialized, porcelain stoneware tile is an advanced type of ceramic, which stands out owing to their technical characteristics. In particular, the growth in their annual production made them one of the most popular construction products in the market (Ke et al., 2016; Pinter Junior et al., 2022). The complexity of the porcelain stoneware tile manufacturing process is associated with the characteristics of their raw materials, which require higher purity and quality (Luz and Ribeiro, 2007; Molinari et al., 2023). Moreover, a significant problem is the

scarcity of ceramic raw material reserves and their distance to production plants, which strongly influence the final products costs (Luz and Ribeiro, 2007).

The availability of raw materials for the porcelain stoneware tile production has been the focus of scientific studies since the late 1990s (Bedoni et al., 1999; Dondi, 1999). However, with the increasing demand for industrial ceramic materials and increased requirements and quality, the responsibility falls on the miners to develop cost-effective products by applying satisfactory mining and beneficiation techniques that improve the deposits utilization (Agus et al., 2000; Comin et al., 2021b).

Porcelain stoneware tile are vitreous ceramic products commonly composed of a triaxial clay minerals blend, feldspars, and quartz (Ochen et al., 2021), which play crucial roles in achieving high performance (Njindam et al., 2018). Kaolin with the chemical formula of

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$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ crystallizes into mullite needles ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$) when fired at temperatures above 1100 °C, increasing the flexural strength (Conte et al., 2022; De Noni Junior et al., 2023; Magagnin et al., 2014).

Currently, kaolin beneficiation is a process based on traditional technology. In general, the main steps are: kaolinite sands washing (residual or sedimentary), flocculation-decantation in dedicated tanks, and dewatering kaolin slurries by filter-pressing. However, there are now some new technologies that allow purer kaolin to be obtained.

Hosseini and Ahmadi (2015) controlled the kaolin quality by the reducing impurity contents, through a biological beneficiation, and obtained a high quality of kaolin with low iron-bearing minerals, decreasing the iron oxides presence, which adversely affects the kaolin color, and reduces its brightness and refractoriness.

Other work, presented by Çiftçi (2023), developed a new method for the dry beneficiation of clay bentonites, by a laboratory-scale dynamic separator. The new beneficiation equipment (a stirred mill) was used for grinding raw bentonite, and through the airflow rate and the rotor rotation speed, obtained a good beneficiation efficiency by removing quartz, opal, muscovite, and feldspars from the raw bentonite samples.

The potential impurities in raw materials (De Noni et al., 2010; Natri et al., 2011), which naturally exist within the deposit owing to the insufficient mineralogical and chemical consistency between batches (Motta et al., 1998), should be minimized in the developing ceramic products. The entire process of developing products begins with the qualitative and quantitative feasibility study of the geological and mining engineering aspects of the deposit, including the required investments for the mining enterprise. Therefore, it becomes extremely important for ceramic tile production regions, such as the productive hub in Santa Catarina state, to obtain alternative sources of kaolinite raw materials. To achieve this, the beneficiation of materials from the northern region of the same Brazilian state, specifically from the Garuva city, provides a possible solution. In particular, the raw material at that region holds a mining title and have an estimated mining concession of 1.5 million tons. However, despite this large volume, limited studies are focused on the ore beneficiation to add value to existing raw kaolin (Kr),

which is currently limited mainly by its excessive silica and iron oxide content, for use in porous ceramic products. Thus, research efforts have focused on the development of technological solutions that allow the kaolinite fraction concentration, improving its application in porcelain stoneware tile ceramic products.

2. Materials and methods

2.1. Geological characterization of the extraction region

The mineral extraction area is located in the northeastern region of Santa Catarina state (Fig. 1). Existing studies (Kaul and Teixeira, 1982) revealed the geological scenario composed by rock and combined sediment, with the emergence of numerous tectonic structures in this area. These processes occurred in two distinct phases of the crustal evolution. The first phase involves the formation of the crystalline basement, and the second phase involves the formation of the sedimentary cover.

In the researched area, there are colluvial-alluvial sedimentary deposits and granitoids belonging to the Paranaguá Complex or Paranaguá Terrane, specifically originating from the Rio do Poço Granite and Morro Inglês Suite. Additionally, there are remnants of supracrustal rocks represented by paragneisses, schists, and phyllites related to the Succession Rio das Cobras. This corresponds to the Coastal Domain and is located to the east of the Garuva Lineament. These are likely roots of probable Brasiliano-age calc-alkaline batholiths, which were accreted during the collisional episodes of the same cycle.

Therefore, in this area all the found sediments have been mapped as Holocene colluvial-alluvial deposits on the slopes and foot of the hills. These sediments are predominantly fine-grained with varying proportions of sand and angular pebbles without visible structures. Lithological characteristics of the area encompass conglomerates, conglomeratic sandstones, coarse to fine sands, and silt-clay sediments. Moreover, significant deposits of kaolinitic clays were observed and characterized throughout the central-northern portion of the area in the

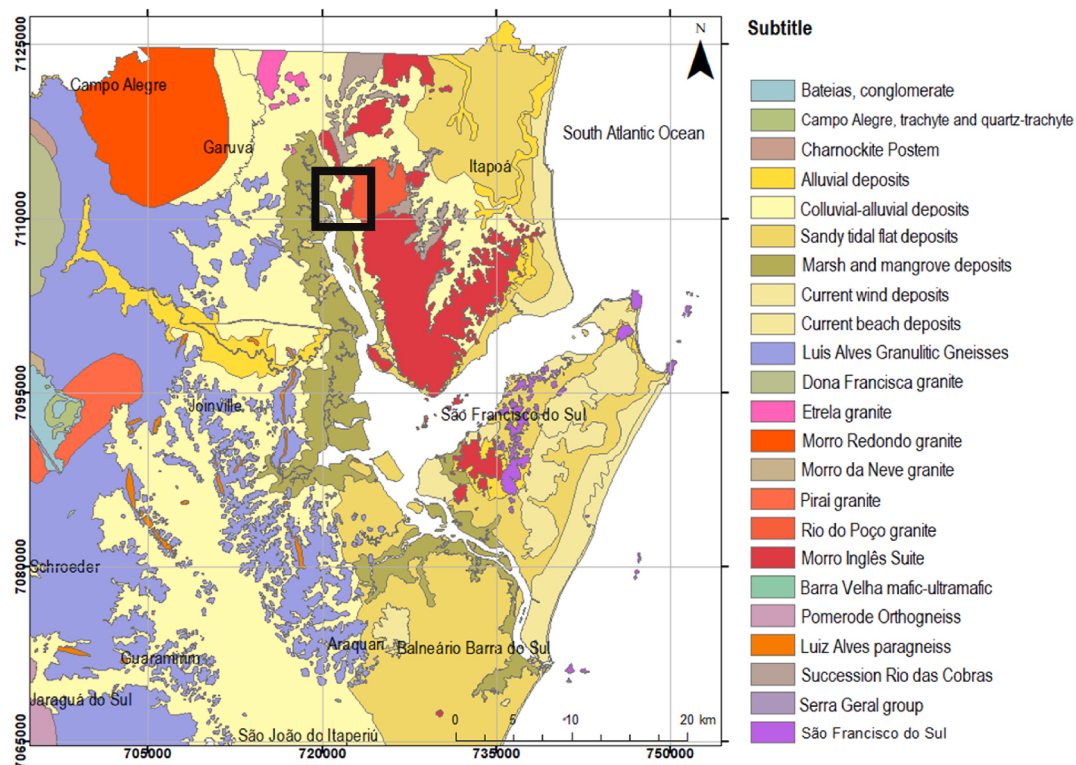


Fig. 1. Geological map of the study area (CPRM, 2022).

topographical lower and flatter regions (Angulo et al., 2009; Caruso Jr. et al., 2000; Maack, 2001).

The kaolinization process has been in detail described by Dill (2016), and denoted different kaolinization environments: primary, secondary and tertiary. After this the author referred that the kaolin and kaolinitic clays are taken as the type-lithology of the near-surface continental environments. Together with bentonites, bentonitic clays and a varied spectrum of argillites they form part of a group of lithologies, encompassing besides bauxite, ferrites and laterite, all of which developed close to the interface between atmosphere, pedosphere, hydrosphere, and lithosphere. In the state of Santa Catarina, there is a predominance of primary kaolin, originating both from the alteration of pegmatites and the weathering of granites (Benvindo da Luz and Lins, 2005).

On the other hand, Varajão et al. (2001) investigated the clay particles in a kaolin deposit from Brazil by different techniques, to study the relationships between morphological and chemical properties of the crystals and to relate these properties to formation conditions. They conclude that in the base of the deposit, kaolinite has a low-defect density whereas in the middle and at the top of the profile, kaolinite has a high-defect density, and show that the results are consistent with the deposit consisting of a transported pedogenic kaolinite over saprolite consisting of in situ kaolinized phyllite.

2.2. Collection of the Kr samples

The Kr samples were directly collected from the mining face at three different points with an approximate quantity of 10 kg, which was later quartered and packaged.

2.3. Beneficiation and characterization

The particle size distribution of the collected Kr sample was determined by laser diffraction using a CILAS 1064 instrument in the range of 0.04–500 μm for a period of 60 s. Sodium polyacrylate (Disperlan LP/G, Lamberti Brazil) was used as dispersant.

On a laboratory scale, a batch of collected raw kaolin, Kr sample (~5 kg), was transformed into a slurry by adding around 6 L of tap water, with the aid of a semi-industrial diluter. Subsequently, the obtained slurry was sieved at 300 μm (#50 ASTM mesh sieve) to achieve two different samples: the materials retained on the sieve (referred to as coarse kaolin, Kc) and those passing through the sieve (referred to as passing kaolin, Kp). After that, the two different fractions (Kc and kp), were removed from the sieve with the aid of a water spray and decanted after standing for about 6 h, to remove a large part of the water. With this beneficiation process, the aim was to separate the majority of the sand present in the sample and obtain a finer fraction made mainly of kaolin (desirably with a kaolinite content $\geq 50\%$).

All the samples (Kr, Kc, and Kp) underwent wet sieving distribution using #4, #8, #16, #30, #50, #100, #200, and #325 ASTM mesh sieve and were chemically characterized by X-ray fluorescence spectroscopy (XRF, PW2400, FRX Philips). The loss on ignition (LoI) method was performed after calcination at 1000 $^{\circ}\text{C}$, with 3 h of dwell-time. The mineralogical characterization was performed by X-ray diffraction (XRD, XRD-6100, Shimadzu) with the measurements taken in the 4–70 $^{\circ}2\theta$ range at a speed of 2 $^{\circ}\theta$ min $^{-1}$, acceleration voltage of 40 kV, current of 30 mA, with an incident radiation CuK α 1 of $\lambda = 1.5406 \text{ \AA}$. The mineralogical phase quantification was obtained by Rietveld refinement, by using the TOPAS software, version 5 (Bruker AXS, Karlsruhe, Germany) was used with a fundamental parameters approach, by using for this purpose the structural models of the ICDD files.

Differential thermal analysis (DTA) and thermogravimetric analysis (TGA) of the raw materials were performed using a simultaneous analyzer (Netzsch 409 Jupiter F3, Germany) with a heating rate of 10 $^{\circ}\text{C}/\text{min}$ from room temperature to 1100 $^{\circ}\text{C}$ in synthetic atmosphere, which is a synthetic mixture of approximately 20% oxygen and 80% nitrogen.

Initially, the samples were subjected to wet grinding in an eccentric jar mill for 35 min and then dried on a drying chamber at $50 \pm 5 \text{ }^{\circ}\text{C}$. The samples were humidified with 7 % of mass water. The granulated compositions were pressed using a laboratory electrohydraulic press (Nannetti Mignon SSN/EA) at 350 kgf/cm 2 . After pressing, the samples were dried on a drying chamber at $100 \pm 5 \text{ }^{\circ}\text{C}$ for 24 h. Finally, the dried bodies were fired in a laboratory roller kiln (Servitech CT094S150) with three controlled zones: heating zone in the entrance of the kiln at 300 $^{\circ}\text{C}$, firing zone at 1190 $^{\circ}\text{C}$, and cooling zone by natural convection. The kiln firing cycle was adjusted to 47 min (Rosso et al., 2005; Sánchez-Muñoz et al., 2002), corresponding to the reference parameters. After firing, each sample was characterized by their water absorption (WA) according ASTM C373-88 (ASTM, 2016), shrinkage after firing (Sh) according ASTM C372 (ASTM, 2020), and colorimetric analysis (L^* , a^* , and b^* coordinates) using a BYK-Gardner 45/0 Gloss spectrophotometer. The rectangular coordinate system Lab was defined by the Commission Internationale de l'Eclairage - International Commission on Illumination (ISO/CIE: 11664-4,2019). D65 was used as the illuminant, and the standard observer was a focus on the retina ($A/10^{\circ}$).

3. Results and discussion

The result obtained for the particle size distribution of the natural Kr sample is present in Table 1. The results demonstrated the heterogeneous grain size distribution of the Kr sample (Fig. 2) showing that there are 50% of particles with a size smaller than 15.25 μm and an average particle size of 66 μm . Considering that a clay material is one whose average particle size is less than 2 μm , the Kc sample is a non-clay material and can be used industrially as a non-plastic material (see Table 2), and the addition of which can help the drying process and regulate the thermal expansion coefficient of ceramic pastes (given their high SiO $_2$ content, see Table 3).

The samples underwent wet sieving (Table 2) for the subsequent mineralogical analysis of different particle size fractions. The Kr sample comprise a Kc mass of 37.59% and Kp mass of 62.40% achieved using a #50 mesh (300 μm). Kc exhibits a coarser particle size distribution with the approximately uniform size of 30% for three size ranges. In contrast, Kp is characterized by a particle size range of less than 45 μm , and therefore must contain mostly kaolinite particles and can be used as a plastic material (kaolin) in the formulation of plastic ceramic pastes where the white color is required (such as porcelain stoneware tiles).

The chemical analysis (Table 3) of Kr characterizes the presence of the clay minerals base oxides (Abubakar et al., 2020) with 69.89% SiO $_2$ and 19.05% Al $_2$ O $_3$. The content of the alkali oxides (K $_2$ O + Na $_2$ O) and alkaline-earth oxides (CaO + MgO), which act as fluxing agents in the ceramic systems (Dondi et al., 2008), reached values of 4.98% and 0.11%, respectively. The chromophore oxide (Fe $_2$ O $_3$ + TiO $_2$) responsible for the ceramic pieces coloration, and the consequent aesthetic properties, which are also significant influence in their production (Piccolo et al., 2022), has a content of 1.29%. The low LoI of 4.64% indicates the loss of constituent water (OH-ions), which is typical for clay minerals.

For the samples obtained from the beneficiation process, the SiO $_2$ content in Kc, increased to 83.93% and that in Kp decreased to 56.42%. Meanwhile, Al $_2$ O $_3$ decreased to 9.87% for Kc and increased to 28.21% for Kp. The alkalis (K $_2$ O + Na $_2$ O + CaO + MgO) content was 5.09% for Kr, which increased to 5.50% for Kc and slightly decreased to 5.01% for Kp. The chromophore oxide content decreased to 0.25% for Kc and increased to 2.39% for Kp. For the LoI a considerable reduction occurred

Table 1
Size distribution and average particle diameter of the Kr sample.

Raw material	Diameter (μm)				
	D $_{10}$	D $_{50}$	D $_{90}$	D $_{100}$	D $_{\text{average}}$
Kr	1.21	15.25	265	500	66

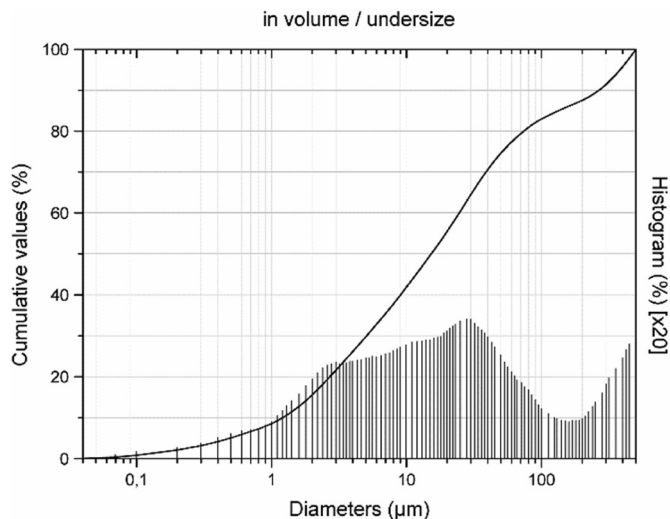


Fig. 2. Cumulative distribution and particle size density of the Kr sample.

to Kc, reaching 0.51%, whereas that in Kp increased to 8.64%. As confirmed in the mineralogical characterization, this result indicates the higher concentration of clay minerals in the Kp sample.

The mineralogical analysis (Fig. 3 and Table 4) showed that the Kr sample is a kaolinitic clay ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})$; PDF card 00-014-0164) with the presence of quartz (SiO_2 ; PDF Card 00-046-1045), feldspar (microcline: $\text{K}(\text{AlSi}_3\text{O}_8)$; PDF Card 00-022-0687), and a small fraction of montmorillonite ($(\text{Na,Ca})_{0.3}(\text{Al,Mg})_2\text{Si}_4\text{O}_{10}(\text{OH})_2 \cdot n\text{H}_2\text{O}$; PDF Card 01-073-6746). For the Kc sample, the higher quartz content (84.10%), following feldspar (11.09%), and lower clay minerals content (4.81%). During the wet sieving process, it's important to consider the possible retention of montmorillonite in the form of clusters in the retained material (Kc). Moreover, the presence of microcline confirms the presence of K_2O in the chemical analysis (Table 3), indicating that feldspar has not undergone complete physical and chemical alterations to clay minerals, such as kaolinite, in this deposit. In contrast, the Kp sample is characterized by a finer fraction in the sieving tests (Table 2), higher concentration of kaolinite, and fine particles of feldspar and quartz. Therefore, the increase in the alumina content from kaolinite and the silica (quartz) reduction content are positively affected by the granulometric separation process using a #50 mesh sieve.

The samples thermal analysis (Fig. 4) illustrates the mass loss of ~1% for Kr (TGA) at 200 °C, corresponding to an endothermic peak at ~100 °C in the DTA curve. Meanwhile, Kp has a mass loss of 1.37% with an endothermic peak at ~100 °C, and Kc has a mass loss of 0.15%, which is attributed to the loss of the adsorbed water (Acchar et al., 2013). In the range of 200–400 °C, Kr exhibits a mass loss of 0.37%, whereas Kp has a mass loss of 0.51%. In contrast, Kc has a mass loss of 0.30% between 200 and 600 °C, which can be associated with the burning of some organic matter. The presence of organic matter in porcelain stoneware tile pastes in fast firing cycles is not recommended due to the strong release of gases it causes during burning, which could cause cracks or bursts in the material (Jaramillo Nieves et al., 2022). For Kr, the mass loss between 400 and 750 °C is 7.29% with a noted one endothermic peak in DTA

curve at 585 °C is due to the removal of hydroxyl groups of kaolinite (Diko et al., 2015; Njoya et al., 2016). For Kp, the mass loss between 400 and 700 °C is ~10% with an endothermic peak at 590 °C, also corresponding to the removal of hydroxyl groups of kaolinite (Comin et al., 2021a). The very small endothermic peak of Kc at ~570 °C (visible in DTA curve) is associated with the α - β quartz transition (Amorós et al., 2022; Zaccaron et al., 2020). Finally, typical exothermic peaks without mass loss related to the structural reorganization of a spinel phase resulting from the kaolinite decomposition (also known as the first stage of mullite crystallization) are noted at around 970 °C for Kr and Kp samples (Yadav et al., 2019).

The physical properties of the ceramic materials after firing (Table 5) demonstrate a typical refractory behavior for the samples with higher kaolinite contents, that is, the shrinkage after firing (Sh) of 2.63% for Kr and 5.61% for Kp. A water absorption (WA) of more than 15% for Kc is also ascribed to its refractory properties and poor packing, associated with the shaping process and to the very low content of clay minerals and high content of quartz. Meanwhile, Kp has a lower WA owing to the higher concentration of kaolinite and microcline feldspar, which act as an alkaline flux.

Furthermore, it is important to evaluate and determine the firing color evolution of the specimens to complement the previous tests (Fig. 5). Lighter shades are characteristic of materials with a higher kaolinite content and low iron content, whereas reddish tones originate from coloring minerals, which hinder the application of clay in porcelain stoneware tile bodies. In this case, the samples should have low levels of chromophore oxides, which allow for lighter shades of the fired samples.

The samples were tested using an instrumental color-measurement (spectrophotometry, CIELAB system) to observe the color variations, and the results are shown in Fig. 6. Considering Kr as the reference and examining the results of the $L^* a^* b^*$ coordinates, Kc exhibits a whiter, less red, and less yellow color after firing at 1190 °C. In contrast, Kp has coordinates that are less white, redder, and more yellow, corresponding with the chemical analysis mentioned above, whereby the higher percentage of chemical elements, such as iron and titanium, resulted in darker shades after firing.

4. Conclusion

Industrial mineral exploitation provides a potential source for raw

Table 3

Chemical composition of the studied samples determined by XRF.

Oxides	Kr	Kc	Kp
SiO_2	69.90	83.93	56.42
Al_2O_3	19.05	9.87	28.21
K_2O	4.75	5.20	3.87
Fe_2O_3	1.01	0.14	1.88
Na_2O	0.23	0.20	0.22
MgO	0.05	<0.05	0.12
CaO	0.06	0.05	0.08
TiO_2	0.28	0.11	0.51
MnO	<0.05	–	<0.05
P_2O_5	<0.05	<0.05	<0.05
Loi	4.65	0.51	8.64
Total	100.08	100.11	100.05

Table 2

Particle size analysis by wet sieving the samples.

Sample (%)	Sieve (ASTM mesh)							
	#4 4.76 mm	#8 2.38 mm	#16 1.19 mm	#30 595 μm	#50 300 μm	#100 149 μm	#200 74 μm	#325 44 μm
Kr	–	0.62	12.08	12.21	12.68	11.88	5.68	5.72
Kc	–	1.62	32.13	33.74	31.17	1.01	–	–
Kp	–	–	–	–	–	19.08	9.13	9.18
								base
								39.12
								0.30
								62.59

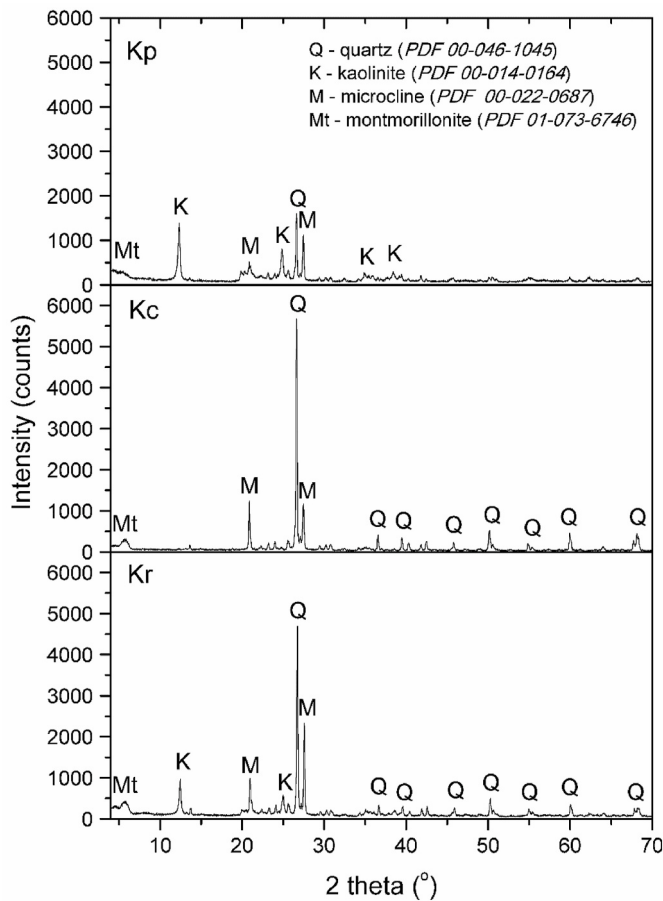


Fig. 3. Mineralogical analysis results obtained by XRD.

Table 4

Quantification of the mineralogical phases obtained by Rietveld refinement.

phase	Quantification (%)		
	Kr	Kc	Kp
quartz	42.72	84.10	18.14
kaolinite	22.01	3.61	53.56
microcline	29.82	11.09	24.53
montmorillonite	5.45	1.20	3.76

materials for the ceramic industry. In this sense, it is imperative to increase its application and give due importance to the resulting by-products and take into account the extent of their reserves. The mineral class analyzed in this study has numerous beneficiation routes to allow different applications and increase the commercial value of newly formed raw materials.

In this study, a region with the geological potential for kaolin clay minerals and is located close to the productive hub for porcelain floor tiles in Santa Catarina state was strategically chosen. However, it was necessary to beneficiate the minerals in this reserve by increasing the kaolinite content to realize technical functionality.

The results showed the feasibility of the applied beneficiation technique, which substantially increased the alumina content by 9.15%, resulting in the cumulative alumina content of 28.20%, which is well above the desired value of 25%. In porcelain stoneware tile ceramic bodies, high alumina content in clay minerals is an important factor, especially for the thermal stability, to the crystalline phase's formation, such as mullite, and to the reducing the pyroplastic deformation. Moreover, other factors that are considered essential for the application in ceramic bodies, such as reduction in the silica content by

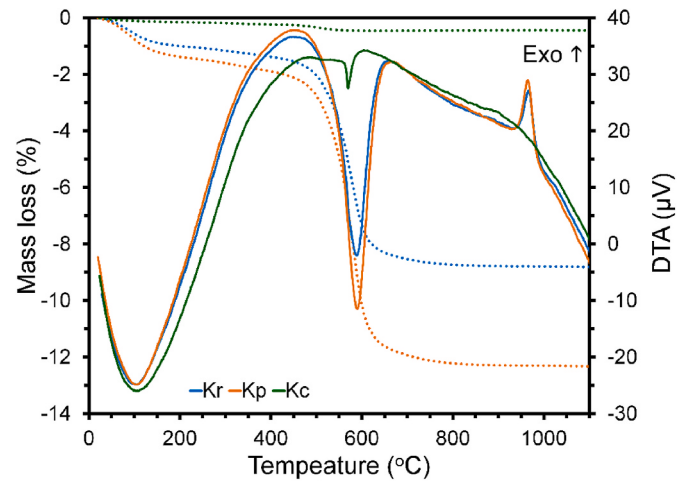


Fig. 4. TDA and TG curves of the samples.
(... ●●) Mass; (—) DTA.

Table 5

Tests results of the samples fired at 1190 °C: shrinkage after firing (Sh) and water absorption (WA).

Technical test	Samples		
	Kr	Kc	Kp
Sh (%)	2.63	0.78	5.61
WA (%)	17.3	29.5	11.7



Fig. 5. Samples photographs fired at 1190 °C, for the visual coloration analysis.

approximately 13.77%, resulting in a final cumulative percentage of 56.42% SiO₂, were achieved. Furthermore, the physical characteristics after firing, such as water absorption, shrinkage and firing color, were highly satisfactory for the predominantly kaolinitic minerals.

Therefore, the proposed method to enhance new raw materials offers a promising potential for cost reduction in ceramic body formulations, especially in terms of logistic costs. Moreover, despite the need for associated beneficiation, the new raw material was obtained through a simple production technique, with great potential for industrial applications in the southern region of the Santa Catarina state. The results allow to conclude that, technologically, the Kp fraction of Garuva kaolin, composed mainly by kaolinite (>50%), is perfectly compatible with the compositional characteristics of kaolin commonly used in the

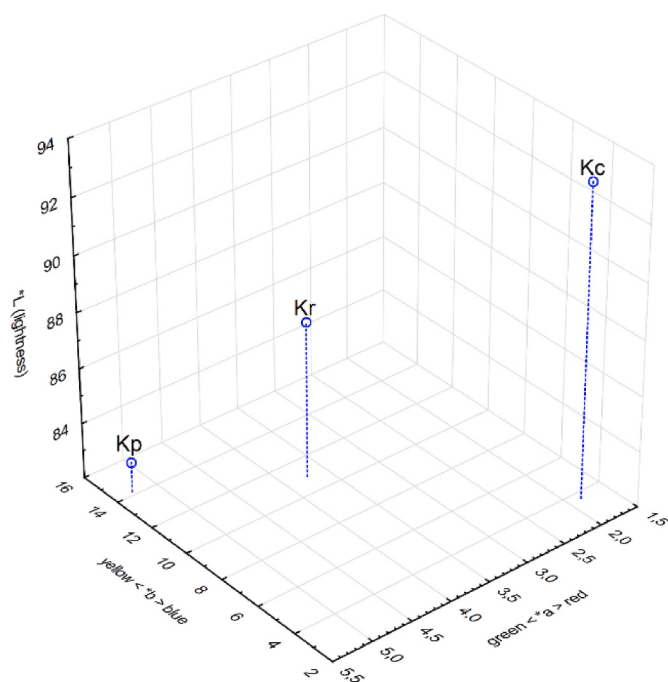


Fig. 6. Colorimetric coordinates in the CIELAB system of the samples fired at 1190 °C.

production of white or off-white ceramic products, such as: white porcelain products, porcelain stoneware tiles, or utility stoneware.

Finally, the sustainability of the washing-settling process, proposed in this work, is based on the guarantee that the increase in the cost of this processing is clearly supported by the commercialization of kaolin that had any commercial value in the past. On the other hand, the environmental impact is greatly reduced by this beneficiation process as it allows for better viability of the research area (kaolin deposit) and, on the other hand, the washing, sieving and decantation are traditional techniques, non-polluting and of reduced environmental impact.

CRedit authorship contribution statement

André Biava Comin: Conceptualization. **Alexandre Zaccaron:** Writing – review & editing, Writing – original draft, Investigation. **Emily Saviatto:** Software, Data curation. **Juliana Acordi:** Visualization, Formal analysis. **Fabiano Raupp-Pereira:** Writing – original draft, Funding acquisition. **João Carlos de Castro Abrantes:** Software, Formal analysis, Data curation. **Manuel Joaquim Ribeiro:** Writing – original draft, Visualization, Investigation. **Guilherme Silva de Souza:** Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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