Porcelain: raw materials, technological properties and applications—a review

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Porcelain: raw materials, technological properties and applications—a review

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Abstract: Porcelain is a ceramic product formulated from a typical triaxial mixture: plastic material; fluxes; and degreaser. It is a very compact product, vitrified throughout. This paper described all the significant characteristics, and raw materials of porcelain and enumerated its fields of application in the world. Porcelain is mainly composed of silica, alumina, and alkaline and alkaline-earth oxides which come directly from these raw materials. The typical mineralogical composition of porcelain is a glassy phase, mullite, residual quartz, and only rarely cristobalite. Linear shrinkage (≤15 %) water absorption (< 0.5 wt%), apparent density ($>2.3\,\mathrm{g.cm^{-3}}$), flexural strength (>35 MPa), mineralogy, and microstructure properties of porcelain depend on chemical composition, granulometry, and thermal treatment of raw materials. It is used extensively in tableware, sanitaryware, decorative ware, electrical insulators, and dental prosthetics.

Keywords: applications, characteristics properties, kaolin and porcelain

La porcelaine : matières premières, propriétés technologiques et applications revue

Résumé : La porcelaine est un produit céramique, formulée à partir d'un mélange triaxial : de matière plastique, de fondant et de dégraissant. C'est un produit très compact, entièrement vitrifié. Ce présent document décrit les caractéristiques significatives, les matières premières de la porcelaine et énumère ses domaines d'application. Une porcelaine est principalement composée de silice, d'alumine et d'oxydes alcalins et alcalino-terreux qui proviennent directement de ces matières premières. La composition minéralogique typique de la porcelaine est constituée de phase vitreuse, de mullite, du quartz résiduel et rarement de la cristobalite. Le retrait linéaire (≤15 %), l'absorption d'eau (< 0,5 % en masse), la densité apparente ($>2{,}3\ \mathrm{g.cm^{-3}}$), la résistance à la flexion (>35 MPa), la minéralogie et les propriétés microstructurale de la porcelaine dépendent de la composition chimique, de la granulométrie et du traitement thermique des matières premières. La porcelaine est largement utilisée dans la vaisselle, les sanitaires, les articles décoratifs, les isolants électriques et les prothèses dentaires.

Mots clés : applications, propriétés caractéristiques, kaolin et porcelaine

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1. Introduction

Clays originated as a result of the dissolution of a given mineral or group of minerals composing rocks like granites [1]. Clays and clay minerals are extensively used in a variety of industrial applications because of their inertness and stability in some situations, and their reactivity and catalytic activity. Plastic raw materials of ceramics involve any clay material that when mixed with water reveals the property called plasticity [2]. The traditional ceramic industry has, as its main objective, to satisfy the needs of the consumers in the production of bricks, tiles, flooring solutions, coatings, sanitary ware, and ceramic objects for utilitarian and decorative purposes. The final properties of the ceramic materials are influenced by their chemical and mineralogical composition, as well as by the chemical bonds and structure, resulting from the manufacturing process [3]. The most significant development in the history of ceramics many centuries ago was the production of vitrified, translucent porcelain in China [4]. Fired porcelain properties are the result of the raw material composition, shaping process and heat treatment. Porcelain is involved in several areas of life for its aesthetic properties, chemical inertness, very low porosity and absorption of water but mostly for its mechanical and thermal resistance suited to its use [5, 6]. The term "porcelain" designates a particular ceramic material, which is vitrified, and translucent and has a white paste, fine, closed and homogeneous. It is fired between 1250 °C and 1460 °C [7, 8].

Extensive research on porcelain has been going on for decades, to confirm its complexities and reiterate the important challenges in understanding porcelain about raw materials, processing science, phase equilibrium, and microstructural evolution. In any given ceramic industry, the type of processing tends to be kept constant, with enough flexibility to accommodate eventual changes in raw materials [9]. Nowadays, the interpretation of raw materials microstructure, microstructural and technological properties of porcelain, and the relation between raw materials, manufacturing process and porcelain microstructure continues to draw particular attention in the scientific community [10, 11]. In literature, considerably less attention has been devoted to the raw materials and applications of porcelains.

The invention of porcelain, as well as its later reinvention in the West, was not a simple achievement but a continuous process involving a series of incremental innovations [12]. Porcelain is a glass-ceramic material with a range of applications including whiteware, tableware, electrical porcelain, dental porcelain and porcelain enamel. Some of these applications involve the material in use on deformations, spalling, flaking or weakening due to their exposition to higher temperatures [13]. Ceramics, therefore, despite their fragility, can be extremely useful, both to the societies which have employed them, usually in abundance, and to the scientists who study them [5]. This paper presents raw materials, properties and application of porcelains based on the results obtained from previous studies.

2. Raw materials

The raw materials are the body and glaze recipe components used in porcelain syntheses. Mostly these are natural minerals and rocks sourced from precise mines and locations which gave rise to several names. Natural raw materials include feldspars, china clays, ball clays, and soapstone. Synthetic raw materials include bone ash, pearl ash, soda ash, smalt, lime, and magnesia [14]. The raw materials used for the body compositions of porcelain can be divided into three groups of minerals, each having its function: plastic, non-plastic including melting minerals, and structural ones [15].

2.1. Main raw material and their interest

Kaolin clay, siliceous clay, crumbly and not very plastic, is used in porcelain paste and is mainly composed of kaolinite ($Si_2Al_2O_5(OH)_4$), an aluminum silicate. It has the particularity of presenting only fluxes which make~~s~~ it a refractory clay. The absence or very small quantity of iron oxides gives it, moreover, a characteristic white coloration after firing [16]. The burnt dough formed from kaolin is also partially vitrified and thereby acquires translucency. The use of a particular clay, the greater vitrification and the translucency make it possible to distinguish porcelain from another category of ceramic of great fires: stoneware.

Kaolinitic clay not only confers plasticity on the dough, thereby facilitating shaping operations, but also promotes the formation of mullite during baking [17]. Clay minerals are formed by the decomposition of feldspathic rock via geological processes; a typical reaction sequence using potassium feldspar (microcline or orthoclase) can be written in **equation 1**:

$$2KAlSi_3O_8 + 3H_2O \rightarrow Al_2Si_2O_5(OH)_4 + 4SiO_2 + 2KOH \qquad \textbf{Equation (1)}$$

If potassium is not properly removed after aging, illicit clays (2: 1 sheet silicates) are formed instead of kaolinite [18]. Kaolinite ($Si_2Al_2O_5(OH)_4$) is the most important and most common mineral of the kaolinite group, otherwise made up of the minerals halloysite ($Si_2Al_2O_5(OH)_4 \times 2(H_2O)$), dickite ($Si_2Al_2O_5(OH)_4$) and nacrite ($Si_2Al_2O_5(OH)_4$) [19]. From a structural point of view, kaolinite is a two-layer phyllosilicate of type 1 : 1 (TO), consisting of a tetrahedral layer $(Si_2O_5)^{2-}$ added to an octahedral layer$[Al_2(OH_4)]^{2+}$ [20]. **Figure 1** shows the crystal structure of kaolinite. Chemical composition of kaolinite in oxide equivalent is: Al_2O_3 = 39.50 wt%, SiO_2 = 46.56 wt% and H_2O = 13.95 wt% based on its ideal chemical formula. **Tables I** and **II** present the chemical and mineralogical compositions of kaolin and kaolinitic clay.

The refractory character of clay increases with the Al_2O_3 content. However, the presence of minerals containing alkaline (K_2O, Na_2O) or alkaline-earth (MgO, CaO) oxides can significantly reduce the temperature at which the liquid phase appears without greatly affecting the Al_2O_3 content. Furthermore, the presence of certain impurities contributes substantially to the coloring of the final product. Thus, according to T. Manfredini and M. Hanuskova (2012), the contents of iron and titanium oxides are important because they have a strong effect on the color of the product after firing, the intensity of which depends on the content of the two oxides, their transformation and the sintering environment [26]. The influence of the presence of iron compounds on the thermal transformations and the mullitization of clays have been widely studied [27]. These studies have shown that the presence of iron reduces the thermal stability of kaolinite by lowering the temperature of endothermic and exothermic transformations. At high temperatures, the decomposition of iron oxide occurs according to **equation (2)**:

$$3Fe_2O_3 \rightarrow 2Fe_3O_4 + \frac{1}{2}O_2 \qquad \textbf{Equation (2)}$$

The release of oxygen gas trapped in the pores (called blistering) causes a decrease in bulk density [28]. N. Bouzidi et al. (2012) indicated that the presence of TiO_2, considered an impurity in porcelain raw materials, promotes the formation of secondary mullite [24]. Organic materials, on the other hand, affect the rheology of suspensions and the behavior of the material during forming. Organic matter and $CaCO_3$ in the raw material release carbon dioxide during sintering. This dioxide increases the porosity of the material and causes it to balloon, thus affecting the mechanical strength of the finished product.

2.2. Fluxes

The melting minerals are feldspars and feldspathoids, talc, pegmatites, Li-minerals, dolomite, glasses. Their role is to form a liquid phase at high temperature, which allows the body densification by viscous flow, and creates a vitreous phase embedding fillers [15].

2.2.1. Feldspar

Historically, potash feldspars (microcline and orthoclase) have been the most commonly used fluxes in porcelain [18]. Feldspar is a mineral with a low melting point and acts as a flux during sintering, giving rise to a liquid phase, which improves the growth of mullite crystals. Therefore, it facilitates the densification of the ceramic body through the filling of the interconnected pores [29]. Also, feldspar helps to lower the kiln temperature during firing thus saving energy and lowering the cost of production. Owing to their composition, feldspars are widely used as raw materials in glass (~60 wt%), and ceramic manufacture (~35 wt%). Feldspar raw materials are a source of alumina (Al_2O_3), alkali (Na_2O and K_2O), and silica as well as they are used as fluxing agents to form a glassy phase at lower temperatures [30]. The rate of feldspars varies and being used in many applications, such as dinnerware (17–20 wt%), sanitary wares, floor tiles (55–60 wt% of feldspar content), wall tile (0–11 wt%), high-tension electrical porcelains (25–35 wt%), dental porcelains (60–80 wt%), kitchen and ovenware (10 wt%), etc [31]. It is considered as a degreaser in raw. Its melting power is most active when it is used very finely ground [32]. Feldspar is one of the minerals commonly associated with phyllosilicates. Feldspar is a silicon aluminate of alkali earth metals ($K_xNa_{1-x}(AlSi_3)O_8$) : Orthoclase $K(AlSi_3O_8)$; Albite $Na(AlSi_3O_8)$;… [33]. **Table III** presents the chemical and mineralogical composition of some types of feldspar. Feldspars are aluminosilicates that form when in some silicon tetrahedra there is a substitution of Si^{4+} by Al^{3+}, which causes a positive charge defect that will be compensated by different alkali and alkaline-earth cations. In terms of composition, feldspars are generally described in a ternary system formed mainly by three large groups: potassium, sodium and calcium feldspars [31].

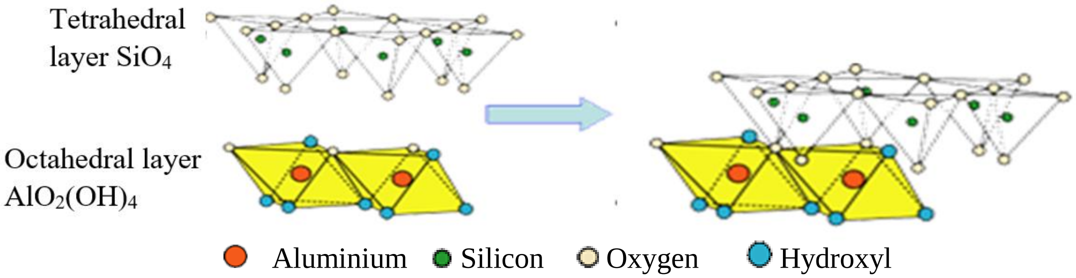

Figure 1: Crystal structure of Kaolinite [20]

Table I: Chemical composition of kaolin of different regions.

Oxides (wt%)	SiO_2	Al_2O_3	CaO	K_2O	MgO	Na_2O	TiO_2	Fe_2O_3	P_2O_5	LOI	Total
Commercial kaolin (high crystallinity) [21]	44.20	39.7	–	0.05	0.03	0.01	1.39	0.21	–	14.38	99.97
Commercial kaolin (poor crystallinity) [21]	43.9	38.5	–	0.06	0.03	–	2.08	1.13	–	14.36	100.06
Kaolinitic clay of Nongfaïrè (Burkina Faso) [11]	54.59	31.5	0.03	2.09	0.54	0.29	1.43	0.87	–	7.29	98.63
Kaolin (Ceramica Eliane, Cocal do Sul, Brazil) [22]	46.96	38.05	0.00	1.14	0.02	0.03	0.03	0.46	0.108	13.2	99.9
Kaolin of Lingshan (Chine) [23]	55.00	42.80	0.10	0.30	0.40	0.40	0.70	0.20	–	–	99.90
kaolin of Charentes (France) [24]	42.40	37.84	0.26	0.02	0.05	0.03	1.99	0.55	–	16.78	99.92
kaolin from the Troup basin (U.S.A.) [25]	58.00	28.00	0.18	0.54	0.40	0.26	1.28	1.36	–	9.66	100.01

Table II: Mineralogical composition of different kaolin

Phases (wt%)	Commercial kaolin [21]		Kaolinitic clay of Nongfaïrè (Burkina Faso) [9]	Kaolin of Charentes (France) [24]]	kaolin from the Troup basin (U.S.A.) [25]
	high crystallinity	poor crystallinity			
Kaolinite	96	97	62	91	72.0
Illite	0	0	18	0	5.5
Quartz	–	–	17	2	21.0
Rutile/anatase	1.4	2.3	1	2	1.5
Gibbsite	2.5	0	0	3	–
Total	99.9	99.3	98	98	100
Balance	0.1	0.7	2	2	–

Table III: Chemical and mineralogical of common additive components to raw materials

Oxides (wt%)	Feldspars		
	K-feldspar [34]	Na-feldspar [34]	Feldspar (provided by Compañía Europea de Arcillas, S.A.) [35]
SiO_2	72.35	70.11	69.86
Al_2O_3	13.50	17.82	16.45
CaO	2.53	1.32	0.54
K_2O	6.58	0.59	10.20
MgO	0.17	0.19	0.06
Na_2O	0.21	8.97	2.28
TiO_2	–	0.06	0.05
Fe_2O_3	1.02	0.30	0.06
P_2O_5	–	–	–
LOI	3.60	0.64	0.50
Phases	13 wt%quartz, 76 wt% Albite, 4 wt% orthoclase and 3 wt% impurity	56 wt% Albite, 22 wt% orthoclase, 22 wt% Nepheline and 1 wt% impurity	65 wt% microcline 20 wt% albite 13 wt% quartz 2 wt% others

2.2.2. Pegmatites

Very similar to feldspars in composition but containing less alkaline, pegmatites are used in a completely analogous manner. Pegmatites are silicate magmatic rocks characterized by exceptional size, from a few centimeters to more than a meter, which allows manual selection. They appear in the form of clusters or veins on the periphery of granite massifs with varied compositions. Pegmatite was defined as an igneous and granitic rock. It has crystals of coarse and variable size with skeletal growth. Pegmatite used as a raw material for the production of ceramics, is composed exclusively of potassium feldspar or albite with quartz or nepheline and small amounts of other minerals [36].

2.2.3. Talc

Talc is a magnesium hydrosilicate with the theoretical formula ($3MgO.4SiO_2.H_2O$) [37]. It is used as an auxiliary flux for electric porcelain, sanitary appliances and vitreous porcelain. The properties of talc, both structural and chemical, give it used in many industrial fields. Talc is typically used as a filler in composite materials to decrease production costs while improving the physical and chemical properties and providing new functionalities [38].

2.2.4. Other Fluxes

In addition to feldspar, pegmatites and talc, there are many other minerals used as fluxing agents. One serie is based on minerals containing lithium such as spodumene ($LiAlSi_2O_6$) which is used directly in many applications. Other flux minerals include barium sulfate $BaSO_4$, witherite $BaCO_3$, and fluorite CaF_2. Micas are also sometimes used [39]. Presently, borates, especially colemanite ($Ca_2B_6O_{11}.5H_2O$) are technologically considered as alternative important raw materials [40]. They have a glass-forming character and have been used in the ceramic industry to reduce the melting point, lower the viscosity and decrease the thermal expansion of the glass phase and therefore raise its heat-resistance, and increase the mechanical strength and chemical stability of the materials [41].

2.3. Degreaser

A degreaser is a non-plastic material added to the dough (clay, kaolin, ...) to make it a less plastic and more solid. The addition of degreasers promotes the formation of less compact texture, which facilitates the release of gases resulting from the thermal decomposition of certain associated minerals. Also, because of their large size, degreaser particles limit shrinkage, prevent deformation and constitute the inert and rigid skeleton of cooked products [15]. At high temperatures, these materials can contribute to the formation of crystallized or amorphous phases, which can influence the final properties of ceramic products, such as mechanical strength. In this regard, in the case of porcelain whose manufacture requires good mechanical strength, the nature of the degreaser plays a very important role [42]. Quartz and generally quartzites are the most refractory ones of those having this structural function [43]. Quartzite, sand and chamotte are degreasers, which are often added to raw materials of industrial ceramics. These materials are relatively inert at the decomposition temperatures of clay minerals.

Due to insulation defects observed with quartz in porcelain, it is often partially or completely replaced by soda lime glass, alumina, vermiculite and barium carbonate to increase the electrical porcelain strength [44, 45].

2.3.1. Quartz

Quartz (SiO_2) is known to prevent warping of the fired materials. It is used as a filler and reduces pyroplastic deformation and shrinkage in the baked product [46]. However, its presence can cause a detrimental effect on the mechanical behavior of ceramics due to the sudden variation in dimension ($\Delta L/L \approx -0.35\%$) associated with the reversible transformation of α quartz to β observed around 573 °C [33]. During sintering, quartz dissolves partially in the liquid phase of fluxes since its melting temperature is higher than those of kaolin and feldspar [11]. Quartz crystals undergo structural changes when heated. Ordinary or α quartz, heated to a temperature of 573 °C, is converted into β quartz, whose crystal structure and physical properties are different. However, when cooled, the high quartz returns to the low quartz state. Between 870 °C and 1470 °C, quartz exists in a form called tridymite, and above 1470 °C, the stable form is known as cristobalite. At around 1710 °C, the quartz melts. The generally accepted sequence of transformation as a function of temperature can be schematized by **Equation (3)** [47]:

$$Quartz\ (\beta) \xrightarrow{870°C} Tridymite\ \alpha \xrightarrow{1470°C} Cristobalite\ \alpha \xrightarrow{1710°C} Fusion \qquad \textbf{Equation (3)}$$

2.3.2. Chamotte

Chamotte, used in the preparation of ceramic materials, is made up of calcined clays between 1100 °C and 1400 °C. It is inert and allows raw parts to have good mechanical properties. The presence of the chamotte in the mixture also makes it possible to reduce the shrinkage of the final parts during sintering. Calcined alumina is also used as a filler in place of quartz to avoid inversion of the quartz and thus improve the mechanical properties of the finished product. Alumina dissolves extremely slowly compared to that of quartz due to the limited solubility of alumina in feldspar glass. The main disadvantage is the significantly higher cost of alumina compared to quartz [18].

2.3.3. Sand

Sand is the most common and least expensive degreaser. It is defined as a set of granular rock particles between 4.75 mm and 75 µm in size produced by natural crushing or disintegration of cemented rocks (sandstone or quartzite). It is composed of silica SiO_2 in the form of quartz but can be introduced in the form of sandy clays. Silica can also be introduced in the form of highly calcined sea pebbles before grinding. It will react during sintering with the other constituents and will contribute in particular to the possible formation of vitreous phases. Very fine, clayey sands are the degreasers par excellence in the terracotta industries. The purest sand is often used, after grinding, in porcelain or earthenware paste compositions [39].

3. Application domain of porcelains

Properties of porcelain depend on the quality of raw material, firing temperature and soaking time [48]. Porcelains are vitreous (vitrified) white-wares ceramics used extensively in tableware (dishes, cups, bowls, etc.), stoneware, sanitary ware, decorative ware, electrical insulators, abrasion resistance and dental prosthetics [49, 50]. The basic ingredients of porcelain which are kaolin or/and kaolin clay, quartz and feldspars are all characterized by small particle size. The combination of these components of porcelain in varying proportions is based on intention and the researcher's desire: the quartz of 10 to 50 wt%, feldspar from 10 to 50 wt% and kaolin from 40 to 80 wt% [8]. The percentages of components of porcelain in the mixture are variable. **Figure 2** presents the location of different ceramics in the feldspar, quartz and clay triaxial diagram. For example, A.A. Nodeh (2017) in his investigation on the influence of bone porcelain scraps on the physical characteristics and phase composition of a hard porcelain body, formulated hard porcelain with a mixture of 80 wt% clay, 7 wt% feldspar, and 13 wt% silica [48]. Hard porcelains typically generally require a composition of about 50:25:25 wt% [51]. These everyday products are an excellent example of the use of natural ressources in our living environment. There is a wide range of products, depending on the applications and characteristics sought.

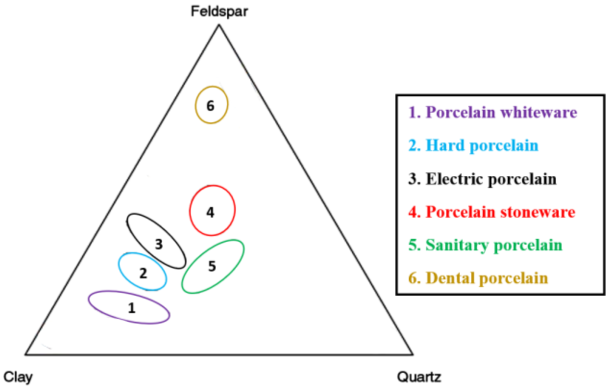

Figure 2: Location of different porcelains in the feldspar–quartz–clay triaxial diagram [11].

4. Properties of porcelain

The choice of a material depends on the requirements expected during its use. Requirements are performance criteria that the material must meet to fulfill a given function. These criteria can be determined by carrying out tests.

The categorization of a porcelain type is dependent not only upon the body composition but also upon the process methodology and firing sequences adopted. However, all types of porcelain must possess translucency to qualify for the appellation [14].

4.1. Chemical composition

Due to the raw material used, the shards of porcelain are white and translucent in thin. The porcelains are almost not open pores but may present some large porosity closed pores. Their break is bright and glassy in appearance. After glazing, the surface of the parts is remarkably smooth and shiny [52]. Most ceramics are formed with the first elements of the periodic table $(Li, C, N, O, Mg, Al, Si)$ [53]. EDS study by Eaba Beyene et al. (2022), on partial substitution of feldspar by alkaline-rich materials in the electrical porcelain insulator for reduction of processing temperature, displays the presence of $Si, O, Al, Na, K, Ca,$ and Fe elements on the fracture surface [54]. **Table IV** presents the chemical composition of some porcelain bodies. The chemical bonds which are strong bonds are of iono-covalent type. The ionic bonds and covalent bonds are very cohesive [55]. Silica SiO_2 forms the basis of many ceramics. Although it has a simple chemical formula, it is a versatile material that can exist in different forms. The basic unit of silica is $(SiO_4)^{4-}$, regular tetrahedron: an Si^{4+} ion is surrounded by four O^{2-} ions.

4.2. Mineralogical composition

Sintered porcelain contains mullite ($3Al_2O_3.2SiO_2$), undissolved crystals of quartz (SiO_2) embedded in a glassy phase. Each phase has a specific influence on the mechanical and dielectric properties of porcelain depending on its concentration and microstructural characteristics [59]. During the heat treatment, the clay materials react, giving rise to the endothermic reaction: dehydroxylation. For a higher temperature, of the order of 1000 °C, the products resulting from the dehydroxylation form different phases which result in the rapid crystallization of the mullite. The content of mullite gradually increases with temperature [60].

Mullite is the only stable binary phase in the $Al_2O_3-SiO_2$ system. Pure mullite is a solid solution with a composition $Al_2[Al_{2+2x}Si_{2-2x}]O_{10-x}$, with $0.17 \leq x \leq 0.5$ [60]. Two main types of mullites are formed at different stages of the porcelain firing. These mullites are distinguished by their different compositions and morphologies. The first type of so-called primary mullite comes from the clay component, and the second, identified as secondary mullite crystallizes from the melting of feldspar. The formation of the primary mullite occurs via the transformation of the spinel-type structure phase formed from metakaolin, while the secondary mullite forms from the reaction between the clay and feldspar relicts at ~1200 °C [61]. Iqbal and Lee discovered in their investigation that there are two types of secondary mullites: The first one named type–MII mullite has a granular morphology, and its crystals are shorter than those of the second type–MIII mullite [62].

Due to its stable temperature, high mechanical properties, low coefficient of thermal expansion and low thermal conductivity, mullite is widely used as a thermal and electrical insulator. According to the literature, the dispersion-strengthening hypothesis and the matrix reinforcement are explaining the strength of porcelains. Especially, this hypothesis explains that the strength is directly related to the interlocking of fine mullite needles in porcelain bodies [63]. An amorphous phase is directly formed from the silica released during the decomposition of the metakaolin reactive phase and forms with the potassium and sodium feldspar a eutectic mixture [64]. This amorphous phase is responsible for closing open pores, in particular on the surface of samples [65]. Therefore, the microstructure of a standard porcelain tile consists of quartz grains, primary mullite crystals, secondary mullite crystals, a silica-rich amorphous phase and porosity [10]. **Figure 3** presents the different phases generally identified according to Youssouf S. et al. (2022) investigations on the optimization of contents of three raw clay materials in the formulation of porcelain [66]. The typical mineralogical composition of commercial porcelain, which commonly is 55 wt%–65 wt% glassy phase, 20 wt%–25 wt% quartz, and 12 wt%–16 wt% mullite [67]. But depending on the composition of the starting raw materials, some porcelain can also contain other mineral phases such as anorthite, rutile, etc. **Table V** presents a quantification of different phases made by T.K. Mukhopadhyay et al. (2009) on porcelain specimens fired at 1250 °C and 1300 °C [58].

Yaseen Iqbal and William Edward Lee (2000) found that porcelains with a high percentage of quartz and fired at over 1400 °C, may also contain cristobalite [62]. Fillers (such as quartz) are materials of high melting temperature that are chemically resistant at commercial firing temperatures (<1300 °C).

Table IV: Chemical composition of some porcelain’s bodies

Oxides (wt%)	SiO_2	Al_2O_3	CaO	K_2O	MgO	Na_2O	TiO_2	Fe_2O_3
Single-crystalline anorthite porcelain [56]	49.60	26.79	19.20	2.89	0.81	0.58	0.03	0.09
Electrotechnical porcelain [57]	68.30	25.70	0.22	3.50	0.22	1.80	–	0.25
Standard hard porcelain [58]	72.285	21.88	1.11	3.03	0.06	0.53	0.6	0.51

Table V: Phase analysis (wt%) of specimen (25 wt% China clay, 25 wt% Plastic clay, 25 wt% Feldspar and 25 wt% Quartz) fired at 1250 and 1300 °C [58].

Temperature	Mullite (wt%)	Quartz (wt%)	Glass (wt%)
1250 °C	15.5	29.4	55.1
1300 °C	16.5	21.0	62.5

They reduce the tendency of the body to warp, distort, or shrink when it is fired to temperatures resulting in substantial quantities of viscous glass. Dissolution of quartz depends on its size: i.e., coarser quartz dissolution is slower than that of fine grains. Quartz grains < 20 mm completely dissolve at ~1350 °C [62]. The use of finer quartz is favorable for porcelain strengthening, and that is also dependent on particle size distribution based on microstructure considerations [22].

During sintering, the raw materials react, and new crystalline phases are formed. W. M. Carty (2002) proposed that quartz dissolution begins at temperatures as low as 1100 °C and increases significantly above 1200 °C [68]. Suat Yilmaz. and Z. Engin Erkmen (2007) indicate that during the heat treatment of porcelain, the major component of kaolinite ($\mathrm{Al_2O_3.2SiO_2.2}H_2O$) is transformed into metakaolinite at 550–600 °C, with a dehydrated metastable amorphous structure [69]. At 950–1000 °C, it is very progressively transformed into an alumina spinel phase and amorphous silica. Just above 1000 °C, feldspar grains react with the amorphous silica and metakaolinite to form a viscous liquid. This process involves the progressive dissolution of the finest quartz grains into the liquid phase [11]. Thus, kaolinite, the minerals mica and feldspar undergo different chemical reactions; consequently, part of the quartz in the original raw materials mixture remains in the final composition and the mullite ($\mathrm{3Al_2O_3.2SiO_2}$) develops as a final product of the transformation of metakaolinite ($\mathrm{Al_2O_3.2SiO_2}$) according to the following **equation (4)** and **(5)** [69, 70, 71]:

$$\mathrm{Al_2O_3.2SiO_2.2H_2O} \xrightarrow{550\text{ to }600\text{ °C}} \mathrm{Al_2O_3.2SiO_2} + \mathrm{2H_2O} \xrightarrow{950\text{ to }1000\text{ °C}} \mathrm{Si-Al_{spinel}} + \mathrm{SiO_{2(amorphous)}} \quad \textbf{Equation (4)}$$

$$\mathrm{Si-Al_{spinel}} + \mathrm{SiO_{2(amorphous)}} \xrightarrow{1200\text{ °C}} \mathrm{3Al_2O_3.2SiO_{2(mullite)}} + \mathrm{SiO_{2(amorphous)}} \quad \textbf{Equation (5)}$$

The process begins with the formation of metakaolinite ($\mathrm{Al_2O_3.2SiO_2}$) as a result of the loss of structural water from kaolinite. It is established that the kinetics of dehydroxylation is of first order with a rate of dehydroxylation directly proportional to the specific surface of kaolinite. In addition, the dehydroxylation of kaolinite is an endothermic process accompanied by a rearrangement of aluminum ions, which changes from octahedral coordination in kaolinite, to mainly tetrahedral coordination in metakaolinite. Subsequently, amorphous silica and spinel are released and at a higher temperature, they react, causing the formation of mullite (**figure 4**). The amorphous silica released during the decomposition of metakaolinite is highly reactive, possibly assisted by the formation of a eutectic melt at 990 °C. As for the exact structure of the spinel, it is currently the subject of debate. Depending on the micro-region from which they develop, mullite crystals have varying shapes and sizes in the baked microstructure [67]. The dissolution of quartz depends on its particle size and also on the type of feldspar used [72].

4.3. Physico-chemical and mechanical properties

Porcelain is characterized by its technological properties such as low water absorption, strong shrinkage, high bending strength, high chemical and staining resistance and an exceptional aesthetic appearance. The porcelain shards are white and translucent at low thickness [17, 73].

4.3.1. Linear shrinkage

Shrinkage is a reduction in the dimensions of the material after drying or under the action of heat. The dimensions of the material are measured before and after sintering (heat treatment). The difference between these dimensions of the material gives the linear shrinkage [66].

In our previous work, we noticed that the shrinkage increases sharply from 1200 to 1250 °C, and up to 1300 °C, it increases slightly. In this work, the maximum linear shrinkage is 14.4 l% [11]. Other authors have observed the same phenomenon with different maximum values of linear shrinkage at different temperatures. According to Sonia Conte et al. (2023), the densification process begins at 1000°C. Linear shrinkage increases sharply after this temperature. This author noticed that maximum shrinkage (≈10 %) is reached around 1280°C and beyond this temperature an inversion of the shrinkage curve is observed due to the expansion of the bodies [74]. C. Zanelli et al., (2011) found in their investigations, a maximum shrinkage value of 7.8 % at a temperature of 1220°C [75]. At 1200°C, Khaled Belhouchet et al. (2019), obtained a maximum shrinkage value of 18 % [76].

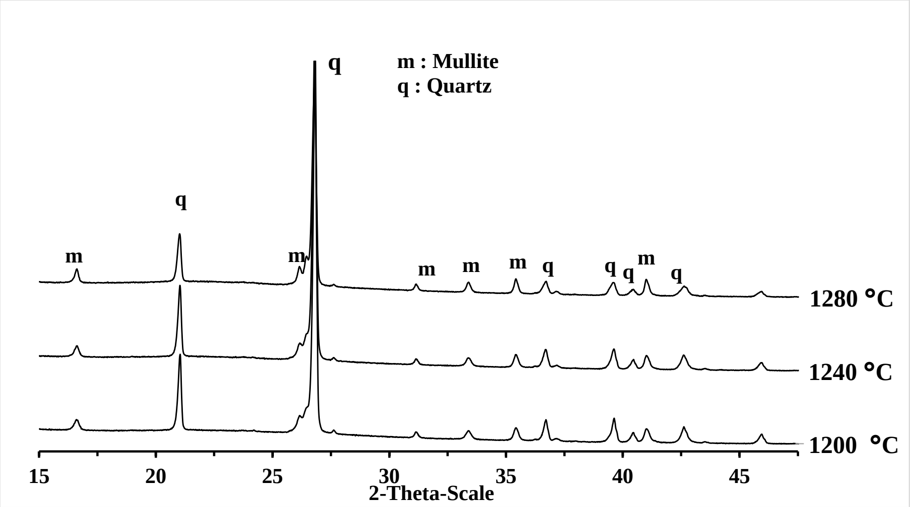

Figure 3: Diffractograms of fired porcelain specimens formulated with 55 wt% kaolinite clay, 25 wt% pegmatite and 20 wt% sand [66]

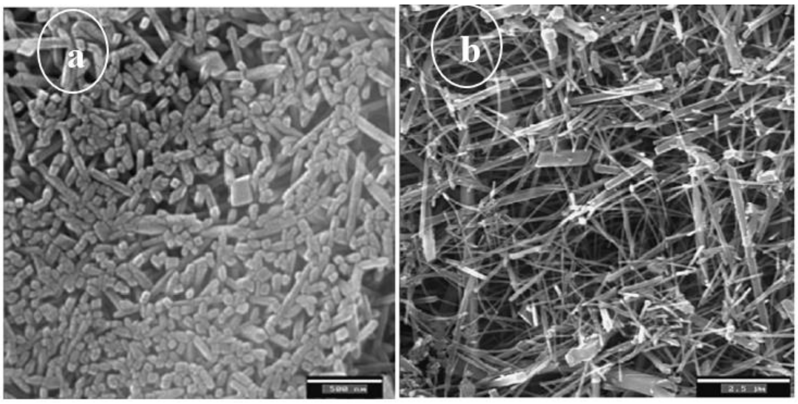

Figure 4: High magnification view of mullite crystals in alumina porcelain: (a) primary (bar = 500 nm); (b) secondary of types II and III (bar = 2.5µm) [61].

4.3.2. Water absorption

Water absorption is the amount of water absorbed by a composite material when immersed in water for a stipulated period [77]. All researchers who have investigated porcelain have shown that water absorption decreases with the sintering temperature, reaching the value prescribed for vitrified products in correspondence with the temperature of maximum densification. According to ISO 10545-3 (2018) water absorption of porcelains products must be less than 0.5 wt% [78]. S.Kr. Das et al. (2003) indicate that the porcelain body achieved vitrification (%WA ~ 0.07) at 1220 ◦C [79]. Hamdan Yahya et al. (2018), indicate that with 9 wt.% dolomite in raw materials, a value of water absorption 0.5 wt% can be reached at a sintering temperature of 1150 °C [80]. Blasius Ngayakamo et al. (2019) in replacement of quartz with 20 wt% of vermiculite, show that the water absorption values of porcelain samples are less than 0.5 wt% (respectively 0.41 wt% and 0.21 wt% at 1200 °C and 1250 °C) [44]. Olupot et al. (2006) while investigating ceramic raw materials for electrical porcelain production, observed a decrease of water absorption in the electrical porcelain sample with a composition of 30 wt% of kaolin, 20 wt% of Ball clay, 20 wt% of feldspar, and 30 wt% of flint from 2.5 wt% at 1200 °C to 0.5 wt% at 1250 °C [81].

4.3.3. Apparent density

The bulk density of specimens can be determined by the ISO 10545-3 (2018) [78]. The density increases with increased consolidation with temperature [43]. Studies of W. Lerdprom, et al. (2016) indicates that the changes in heating rate have minimal influence on apparent bulk density values after sintering at 1150 °C and 1175 °C [10]. However, after sintering at 1200 °C, there is a significant change in apparent bulk density values. This change is due to bloating. It is a not heating rate effect. In other words, the bloating effect has much more effect than the heating rate when samples are sintered beyond optimal temperature. In evaluating the effect of alumina on the mechanical and electrical properties of electrical porcelain, with a composition of 35 wt% of kaolin, 25 wt% of Ball clay, 15 wt% of feldspar, 5 wt% of alumina and 20 wt% of silica, Noori et al. (2007) showed that, bulk density changed from 2.43 $\mathrm{g.cm^{-3}}$ at 1250 °C to 2.46 $\mathrm{g.cm^{-3}}$ at 1300 °C and 2.48 $\mathrm{g.cm^{-3}}$ at 1350 °C [45]. Kitouni et al. (2011) utilizing local raw materials within fractions of 37 wt% kaolin, 35 wt% feldspar and 28 wt% quartz, indicated that porcelains sintered at 1200 °C for 2 h with a heating rate of 5 $\mathrm{°C.mn^{-1}}$, have a bulk density of about 2.50 $\mathrm{g.cm^{-3}}$ [42]. Sonia Conte et al. (2023) by investigating porcelain versus porcelain stoneware, indicated that porcelain bodies reach a maximum bulk density of 2.443 $\mathrm{g.cm^{-3}}$ for traditional hard porcelain (50 wt% kaolin, 25 wt% quartz and 25 wt% feldspar) and firing at 1280 °C [74].

4.4. Flexural strength

The influence of raw materials composition on the flexural strength of porcelain bodies has been experimentally studied by many researchers [43]. The flexural strength increases both with the increase in the bulk density of the porcelain bodies and with the firing temperature [82]. In our previous studies (optimization of contents of three raw clay materials in the formulation of porcelain), we have formulated porcelains with high flexural strengths (53.54 MPa) from 55 wt% of clay, 25 wt% of pegmatite and 20 wt% of sand at the temperature of 1240 °C. Ç. Öztürk et al. (2021), studying on Effect of calcined colemanite addition on properties of porcelain tile, indicate that 50.02 MPa of flexural strength value can be reached with 36 wt% of clay, 11 wt% of kaolin, 11 wt% of quartz and 42 wt% of Sodium feldspar. This author showed that with 0.50 wt% of calcined-colemanite addition in raw composition, a higher bending strength (66.04 MPa) is reached [75]. According to the International Organization for Standardization EN ISO 10545–4 (2019), the average value of bending strength of porcelain bodies is 35 MPa [83].

4.5. Dielectric permeability

The electrical parameter characterizing the dielectric properties of porcelain is its dielectric permittivity, which depends on the capacity of the cations of the glassy and crystalline phases to polarize in an electric field [84]. The two phases, quartz and the glassy phase are insulating phases at room temperature. The mullite phase is considered to be a conductive or semi-conductive phase due to the presence of vacancies of O^{2-} in its structure. The main phase is the glassy phase which has a dominant influence on the dielectric properties of sintered ceramics [85]. Porcelain is an electrical insulator having an electrical resistivity between $10^{12} - 10^{14}$ ohm-cm at room temperature [86]. Ceramics in general are excellent electrical insulators with dielectric strength between 6.1 and 13 $\mathrm{kV.mm^{-1}}$ [59, 87] and can be used to support elements of electrical circuits. It is they, in particular, which constitute the insulators of high-voltage lines. Andualem Merga et al. (2019) are observed that the dielectric strength of porcelain insulators increases with increasing firing temperature and reaches a maximum of 8 $\mathrm{kV.mm^{-1}}$ at a firing temperature of 1300 °C, with the composition of 45 wt % clay, 45 wt % feldspar 10 wt % quartz in raw materials [87]. Blasius Ngayakamo et al. (2019) while evaluating Kalalani Vermiculite for Production of High Strength Porcelain Insulators, are showed that with 20 wt% of vermiculite the dielectric strength increased from 50.8 $\mathrm{kV.mm^{-1}}$ at 1200 °C to 61.3 $\mathrm{kV.mm^{-1}}$ at 1250 °C [44]. Boussouf et al. (2018), showed that the dielectric strength values of porcelain sintered at 1275 °C increased and reached at maximum 36.27 kV/mm, when the percentage of quartz decreased and quartz milled to a very fine size [88].

The brightness of vitrified porcelain is one of the important technological requirements. The whiteness of vitrified porcelain is sometimes affected due to the increased amount of $\mathrm{Fe_2O_3}$ and $\mathrm{TiO_2}$ impurities present in it [89].

4.6. Microstructure of porcelain body

According to M. Romero and J. M. Pérez (2015), after sintering, standard porcelains are deprived of open porosity, but they constantly contain a considerable number of closed pores (6–10 vol.%) because of the high viscosity and the low mobility of atoms in the large amount of fused aluminosilicates [67]. The microstructure of porcelain is characterized by the presence of small, strongly imbricated crystals of mullite embedded in an amorphous vitreous phase. Porcelains lack open porosity (<0.5 vol.%), but may have some (2–10 vol.%) large closed pores (dimpling) [17, 40, 67, 90]. During the sintering process, the development of the liquid phase gradually closes the capillaries which constituted the open porosity and a fine closed porosity develops, which consists of small closed pores (<5 µm) distributed throughout the clay matrix. In addition, larger isolated pores of spherical shape (> 10 µm) form the coarse closed porosity (**Figure 5**). It is directly linked to the melting of feldspar grains. Finally, the interparticle porosity is composed of pores of irregular morphology located at the limits of the quartz and feldspar grains with the vitreous matrix [67]. According to Tulyaganov, D.U. (2006), closed pores cannot be eliminated (remain between 2 and 8 vol.%), and both small spherical pores and irregularly shaped coarse pores remain in the microstructure [91]. Sanchez, E., et al. (2010) attempted to explain the effect of sintering on the microstructure of porcelain by saying that at high temperatures (> 1200 °C), the gas pressure of the occluded pores is high and counteracts capillary pressure, causing the bloating of the tile body. In the final stage of firing, the coarsening and solubility of gases filling the closed pores become the most important phenomena affecting the microstructure [92]

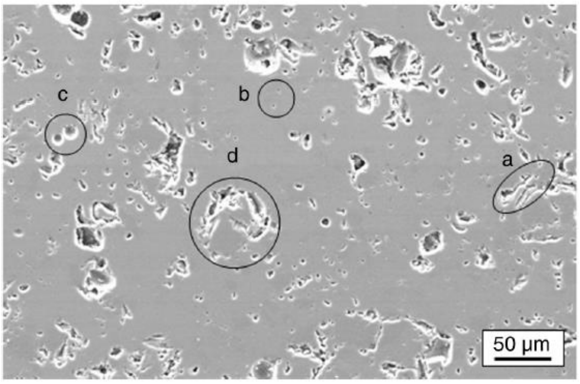

Figure 5: Different types of porosity existing in porcelain tiles: open porosity (a); fine closed porosity (b); coarse closed porosity (c); and interparticle porosity (d) [67].

Recently, William Ochen et al. (2019), reported that the isolated pores prevent the growth of any crack, and the interconnected pores facilitate crack propagation [93].

5. Discussion

Clay can be very plastic and too sticky to use on its own. Indeed, although easy to shape, the paste is often too deformable to allow manipulation without damage to the preform. Moreover, the colloidal character of the clay particles and their strong propensity to flocculate do not make it possible to obtain pastes containing a high-volume fraction of phyllosilicates. The drying and sintering of such a preform should be accompanied by a strong contraction which would make it difficult to control the shape and dimensions of the final part. To limit all of these effects, non-plastic products, called degreasers, must be present in the initial mineral mixture. They form a skeleton which helps to improve the mechanical strength of the preform and to limit sintering shrinkage. Degreasers are generally made up of relatively large grains (>10 µm) to generate the interconnected network of large capillaries necessary for drying. The more the degreaser contains free quartz, the more its melting power decreases. A. Arib et al. (2007), in the study of the influence of the feldspar source demonstrated that the fluxing power is more active when the material is used very finely ground [32].

The properties of a ceramic product are highly dependent on the characteristics of the raw materials and their subsequent modification during processing. Silica (SiO_2) is a major component of raw materials that affect refractory properties during the sintering process. Alumina (Al_2O_3) both free and bound is an important component of ceramic raw materials. P. Sinuhaji et al. (2020) assert in this regard that alumina has high thermal and electrical resistance [94]. Sodium oxide (Na_2O), potassium oxide (K_2O) and calcium oxide (CaO) present in the flux sample in appreciable amounts will serve to reduce melting temperatures. These oxides directly affect the sintering parameters, ie the rate of densification, the eutectic point and the rate of crystallization. Calcium oxide CaO is an active flux for high melting temperature glazes and promotes crystallization. Magnesium oxide MgO has a behavior similar to that of CaO, in small quantities, it presents itself as an active flux, promotes the elasticity of the glaze and ensures better hardness. Potassium oxide is chemically very active and acts in glazes as a powerful flux at both the lowest and the highest temperatures. The fluxing power of Na_2O is substantially equivalent to that of K_2O, but its coefficient of expansion is lower. The viscosity will be significantly decreased with the use of Na_2O compared to K_2O [95].

The temperature, time and atmosphere in the kiln affect the chemical reactions and microstructural development of porcelain and, therefore, play an important role in the firing properties of porcelain. In the same direction, F. F. Lange (1984) indicated that these parameters directly influence the density, the grain size and the homogeneity of the composition of the material [96]. The thermal evolution of the raw materials involved in the manufacture of porcelain is carried out according to several well-known phenomena. First, dehydration during which the water physically adsorbed on the surface of the particles is desorbed, at atmospheric pressure, at temperatures generally below 100°C while more strongly bound residual water can be released at temperatures between 100 and 300°C. Then comes the combustion of organic matter, which concerns the organic products introduced for manufacturing, such as petroleum and mold release oils. It takes place between 300°C and 500°C. Beyond 500°C, the mineral dehydroxylation reaction occurs which can be broken down into two main stages: the dissociation of the OH groups into O^{2-} and H^+ which combine to form water molecules (dehydroxylation in the strict sense) followed by the removal of water from the crystal lattice. According to Pampuch (1971) [97], the dehydroxylation mechanism is homogeneous if there are adjacent OH- of different acidities. It depends on the presence of different cations or hydroxyls in the octahedral layer. The last phenomenon before cooling is recrystallization. It does not involve a significant reorganization of the crystal lattice. The different transformations during the sintering of a ceramic material can be represented in **figure 6**. The stability of materials, their chemical reactivity, their opacity, their fluidity and their resistance are affected by the size and characteristics of the particles that compose them.

The maximum densification is achieved with little residual porosity and this phenomenon is driven by the progressive melting of feldspars and to a minor extent of quartz [75].

Porcelain is characterized by a strong shrinkage (12 to 14 l%). Its withdrawal is linked to two parameters:

- the water leaving when the dough dries, when the physical state of the kaolin silicates changes.
- the vitrification and crystallization of raw materials in the form of mullite crystals.

The water absorption by a ceramic body is influenced by the granulometric distribution of a raw material or ceramic paste, the chemical and mineralogical composition of the clay minerals, crystalline structure and also the porosity after drying and firing [3].

According to standards on ceramics (ASTM C 373-88 (2006), ISO 10545-3 (2018), ISO 10545-4 (2019)), the conditions shrinkage < 12 l%, water absorption < 0.5 wt% and flexural strength > 35 MPa are necessary for a longer cleanliness of the final product [78, 83, 98]. The apparent density of typical porcelain bodies is between 2.3 and 2.5 $g.cm^{-3}$ [99].

Maria Gorea et al. (2007), indicate that the porcelain's electric and mechanical characteristics depend on both the properties of the crystalline phases (crystal size and distribution, some isomorphous substitutions in the crystalline network), and those of the vitreous phase (presence of some ions, especially Na^+ and K^+ from the feldspar melt) and of the pores (shape, size, distribution) [100]. The dielectric strength of electrical insulating materials depends on the thickness of the sample [54].

According to Yaseen Iqbal (2008), the formation of about 40 to 60 vol.% of glass leads to a reduction in porosity and enhanced densification of the porcelain body [101].

The mechanical properties of porcelain are improved when replacing quartz with other materials such as rice husk ash and silica fume, others are kyanite, bauxite, fly ash, sericitic pyrophyllite and sillimanite sand [102]. All authors investigated the effect of perlite addition (Kayacı. (2020)) the effect of spodumene addition (Aydın et al. (2014)), the using nepheline-syenite, as a fluxing agent (Esposito et al. (2005)), pointed out the porcelain tiles had high bending strength (46-85 MPa) [103-105].

The flexural strength of porcelain tiles is affected by the presence of crystalline phases (mullite, quartz, cristobalite) and amorphous phase in the final product and the particle size of raw materials. The increase of flexural strength was explained by two reasons: the higher formation of crystalline phases and the high liquid phase formed by the rich alkali oxides in the composition [106].

The presence of pores in the material has two key aspects. Firstly, they reduce the overall load-bearing area, therefore flexural strength will be dependent on the minimum contact area. Secondly, pores lead to stress concentration, thereby facilitating crack growth hence affecting flexural strength [93]. The maximum flexural strength develops in a porcelain

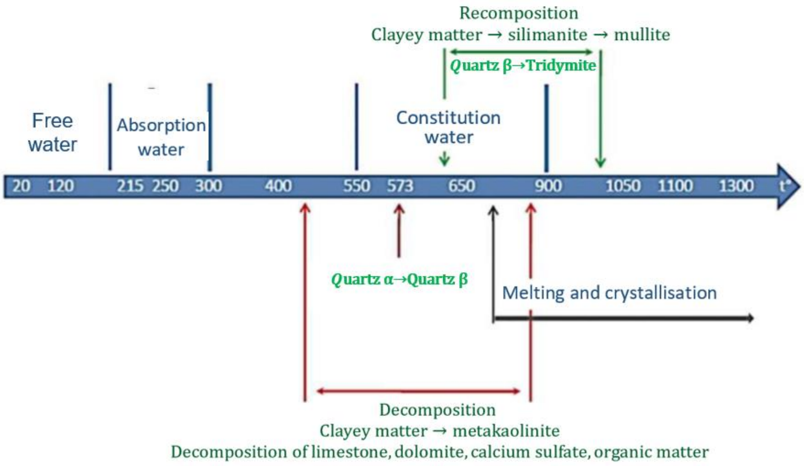

Figure 6: Departure of water and physico-chemical transformations

body when apparent porosity decreases to zero [80]. Boussouf et al. (2018) indicated that the dielectric properties of electrical porcelain samples are determined by the concentration and mobility of K^+, Al^{3+} and Na^+ ions in the glassy phase. This author explains that when there is a large amount of glassy phase present in the structure of samples, these ions find an easy path to move and hence increase the conductivity [88].

Indeed, during firing, the melting of the clay constituents and additions at the sintering temperature creates the liquid phase which, during its flow, ensures the densification of the material. The presence of this liquid phase makes it possible to initiate the rearrangement of the grains by capillarity, which causes an increase in the compactness of the system. Once the grain rearrangement step is completed, part of the quartz dissolves in the liquid phase. The grains tend to approach each other in order to densify the material. The best vitrification range is obtained when the open porosity reaches a minimum value, close to zero, and simultaneously the linear shrinkage is maximum.

6. Conclusion

Composed primarily of white clay and plastic, fluxe, and degreaser, porcelains are heat-treated to form a mixture of glass and crystalline phases. In addition to chemical nature of compound, it is the microstructure of the material (size, grain shape, rate and types of porosity, phase distribution) that controls their properties. In many porcelain applications, the requisites for the finished product (nearly zero water absorption, very high values of flexural strength) are affected by both the choice of the raw materials and the processing conditions during the basic phases of batching, grinding, pressing, drying and firing. The fine and translucent porcelains are used generally to produce tableware, household kitchen utensils, sanitary ware, laboratory materials, electric insulators, sparkplugs for combustion engines, and biomedical organs (such as teeth, disc, or body).

7. References

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