Contemporary Dental Ceramic Materials, A Review: Chemical Composition, Physical and Mechanical Properties, Indications for Use

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Abstract

BACKGROUND: The high esthetic expectations from the prosthodontic restorations have directed the qualitative development of the materials towards the all-ceramic materials that are capable of replacing porcelain-fused-to-metal systems.

AIM: This article reviews the literature covering the contemporary all-ceramic materials and systems with a focus on the chemical composition and materials' properties; also it provides clinical recommendations for their use.

RESULTS: The glass-matrix ceramics and polycrystalline ceramics are presented, as well as recently introduced machinable materials, all-zirconia and resin-matrix ceramics. The specific properties of zirconia, such as transformation toughening, stabilisation of the crystallographic structure, low-temperature degradation and factors affecting the zirconia's ageing, are emphasised.

CONCLUSION: The favourable properties of the resin-matrix ceramics, such as modulus of elasticity similar to dentin, shock-absorbing characteristics and high resilience and fracture resistance, are also covered in this article.

Introduction

Technological development in the dental industry, particularly in the field of ceramic materials, enabled the production of metal-free restorations made up of all-ceramic materials [1]. The qualitative improvements provided ceramic materials with many advantages over the porcelain-fused-to-metal system such as excellent esthetic appearance due to favourable optical properties (translucency and transparency), natural tooth color and chromatic stability, biocompatibility, chemical inertness and low thermal conductivity, optimal mechanical properties such as high flexural strength and fracture toughness, as well as wear resistance and low abrasive properties [2].

Consequently, all-ceramic materials can be used for manufacturing of all kind of single-tooth restorations [3] [4] such as veneers, inlays, onlays, crowns and posts; lithium disilicate ceramic can be used for production of 3-unit bridges (in the anterior and premolar region), whereas multi-unit bridges can be made up only by stabilized zirconia; ceramic materials with a resin matrix inside, are especially suitable for the production of crowns over the implants or tooth restorations in a region where high masticatory pressure is generated [5] [6].

This article reviews the current literature regarding the all-ceramic materials, represents the recently proposed classification system [7] based on the phase or phases present in materials' chemical composition; presents already established as well as recently developed and introduced materials, their

properties and clinical indications. Glass-ceramics have superior optical properties [8] [9] [10], stabilized zirconia, referred to as "ceramic steel" [11] not only because of the phenomenon called "tension expansion", but including other properties such as the highest strength, high fracture toughness and Vickers hardness [12] [13] [14], whereas so-called "hybrid ceramics" appeared to have advantages in terms of fracture resistance, high resilience and shock-absorbing properties, milling efficiency, polishability and accuracy-less marginal chipping [5] [15] [16].

The purpose of this review is to help therapists take into consideration not only the patients requirements, but material features as well, when choosing a ceramic material, so all-ceramic restorations with high reliability and long-term clinical success can be made.

Dental Ceramics – Chemical Composition

For a long time, ceramic materials have been defined as compounds of metallic and non-metallic elements consisting of oxides, nitrides, carbides, and silicates [17]. Most of the ceramics used in dentistry were primarily based on silicon that usually occurs in the form of silica (silicon dioxide), due to the silicon's high oxygen affinity or as silicates compounds [18].

The increasing use of polycrystalline ceramics (with no silicon in their composition), and the introduction of so-called 'hybrid' ceramics imposed the need for a new classification system [7]. According to this classification system, all-ceramic and ceramic-like restorative materials can be categorised into three groups: (1) glass-matrix ceramics, (2) polycrystalline ceramics, and (3) resin-matrix ceramics, depending on the phase/phases present in their chemical composition. "Glass-matrix ceramics" are nonmetallic inorganic ceramic materials that contain a glass phase, while "polycrystalline ceramics" are defined as nonmetallic inorganic ceramic materials that do not contain glass, but only a crystalline phase. In the third group - "resin-matrix ceramics" are included materials that have a polymer matrix, containing predominantly inorganic refractory compounds [7]. Different phases present in materials' chemical composition affects the sensitivity of the ceramic material to the hydrofluoric acid when etching (as a surface treatment method before adhesive luting) to achieve stronger resin-ceramic bond [19].

Glass-Matrix Ceramics

The first group, glass-matrix ceramics, is further divided into three subgroups: feldspathic ceramics, synthetic ceramics, and glass-infiltrated ceramics.

Feldspathic ceramics

The traditional type of dental ceramics are feldspar-based, composed of the significant amount of feldspar ($KAlSi_3O_8$), quartz (SiO_2), and kaolin ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$). Feldspar is a greyish crystalline mineral that can be found in rocks rich with iron and mica. Feldspar rocks are grounded, and after using strong magnets to remove iron compounds, it is milled to obtain the purest powder. Quartz or silica (SiO_2) is the matrix component (55–65%) responsible for the translucency of the restoration. As it is not a strong material, 20–25% alumina (Al_2O_3) is added as a reinforcing component. Kaolin is a hydrated aluminium silicate that is used in a limited amount (4%) as it has opaque properties, unlike the human teeth which are translucent. It is used in the composition of dental ceramics as it binds the loosely held ceramic particles together [18] [20].

VITABLOCS® from VITA Zahnfabrik are the most used feldspar-based CAD/CAM ceramics with an average grain size of 4μm and flexural strength of 154MPa. In 1985 first inlay was produced made of VITA Mark I, whereas in 1991 VITA has promoted Mark II, a monochromatic material with improved chemical composition and physical properties. To imitate the natural colours of the tooth, VITA has introduced next generations: VITABLOCS® TriLuxe (2003) and TriLuxe forte (2007). VITABLOCS® TriLuxe includes three, while TriLuxe forte four layers of different shade intensity from the cervical to the incisal edge, especially suitable for veneers, partial and full crowns in the anterior region. Further improvement in a replica of the shade gradient of natural teeth between the dentin and the edge areas, has been succeeded with VITABLOCS® RealLife (2010), multichromatic feldspar ceramic with different colour intensity in three dimensions [21].

Numerous micropores and channels of different sizes with irregular ceramic particles can be observed on the surface of VITA Mark II after surface etching with hydrofluoric acid; such a modified surface micromorphology is suitable for capturing of a composite luting cement [19] [22].

Synthetic glass-ceramics

To remain less dependent on natural resources of raw materials, the so-called "synthetic" glass-ceramics are invented, with a greater presence of crystalline phase that reduces the possibility of crack formation, or the propagation is slowed down if it has already occurred [7]. The presence of the crystals improves the mechanical properties of the ceramic.

The microstructure of glass-ceramics comprises dispersed crystalline phase (crystals)

surrounded by a translucent glassy phase (matrix). The glassy phase possesses the usual properties of glass such as translucency, brittleness and non-directional fracture pattern. The crystalline phase improves light scattering and opacity, thereby colour adaptation of transparent glassy phase to dental hard tissues (enamel and dentin), and provides the ceramic material with strength, stability during firing and resistance to stresses that occur in the mouth [18].

The crystals are "artificially" created by controlled nucleation and crystallisation. The size and distribution of the crystals are determined by the composition and processing of the base glass and the subsequent heat treatment. This process allows "tailor-made" materials to be produced, which exhibit homogeneous structure, good optical properties, appropriate wear characteristics, as well as optimal strength [23] [24].

The final mechanical properties of the synthetic glass ceramics are determined by two groups of factors: intrinsic and extrinsic. Intrinsic factors are crystal size, number and geometry, the distribution pattern of the crystals (homogeneity), as well as thermal expansion/contraction matching between the crystal phase and glassy matrix. Long-term performance of the material also depends on the extrinsic factors such as fabrication conditions and conditions of the oral environment: humidity (stress corrosion), variations of the pH level, thermo shocks, cyclic loading and peak loads that can reach extremely high levels when hard objects are accidentally encountered during mastication [25].

Leucite-reinforced, lithium disilicate, zirconia-reinforced lithium silicate, and fluorapatite-based ceramics are representatives of synthetic glass-ceramics.

Leucite-reinforced ceramic (IPS Empress CAD, Ivoclar Vivadent) consists of leucite crystals (35–45 vol%) that are homogeneously distributed into the glassy matrix. Leucite crystals are created by the controlled firing of feldspar at 1150°C. Potassium aluminium silicate is disintegrated into leucite and two molecules of silica:

$$K_2O \cdot Al_2O_3 \cdot 6SiO_2 \rightarrow K_2O \cdot Al_2O_3 \cdot 4SiO_2 + 2SiO_2 \text{ [17]}$$

The process is known as surface crystallisation, in which the crystals grow slowly along the grain boundaries towards the centre of the grain [26].

Due to the high silica content (60–65 wt%) this ceramic has improved translucency, fluorescence and opalescence, while the crystalline content is responsible for the flexural strength of 160 MPa and ability to absorb the fracture energy that results in arrested or slowed down crack propagation. The diameter of the leucite crystals ($KAlSi_2O_6$) is 1–5 µm [27] [28].

IPS Empress CAD is monochromatic ceramic,

with the need for further characterisation of the milled restoration, whereas IPS Empress CAD Multi is polychromatic, with high intensity of chroma and opacity in the cervical area and high level of translucency in the incisal region [29].

As a glass-ceramic, the best surface treatment method before adhesive luting is HF-acid etching. The acid dissolves the leucite crystals more quickly than the glassy matrix resulting in a surface with honeycomb-like appearance [22] [27] [30].

Another leucite-reinforced ceramic is Paradigm C, introduced in 2006 by 3M ESPE.

Lithium disilicate ceramic (IPS e.max CAD, Ivoclar Vivadent) is comprised of approximately 70 vol% of crystalline phase incorporated in the glassy matrix. In the process of production, ceramic is cast in transparent glass ingots that contain lithium orthosilicate. The process of partial crystallisation that follows leads to the formation of 40% platelet-shaped lithium metasilicate crystals (with the average size of 0.2–1.0 µm), Li_2SiO_3, embedded in a glassy phase. It is a so-called intermediate crystalline phase or 'blue' state, with 130 ± 30 MPa flexural strength, in which the blocks can easily be milled in CAM unit. Milled restorations are tempered at 850°C and eventually lithium disilicate crystals - $Li_2Si_2O_5$ are formed [31] [32] [33], which impart the milled restoration with the final shade and flexural strength of 360 ± 60MPa:

$$\text{Lithium orthosilicate } (Li_4SiO_4) \rightarrow \text{lithium}$$
$$\text{metasilicate } (Li_2SiO_3) \rightarrow \text{lithium disilicate } (Li_2Si_2O_5) \text{ [31]}$$
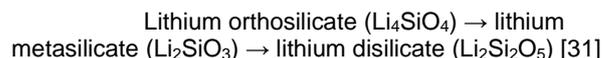
Randomly-oriented, densely distributed, elongated fine-grained lithium disilicate crystals, 1.5 µm in length, scattered with shallow irregularities become visible after dissolving the glassy matrix by etching the surface with hydrofluoric acid (HF) [19] [22] [30].

Zirconia-reinforced lithium silicate is glass-ceramic material enriched with highly dispersed zirconia, developed in close cooperation between Degudent, VITA and Fraunhofer Institute for Silicate Research [34]. After breaking up the partnership, Dentsply and VITA continued with their research which, in 2013, resulted in the introduction of Celtra Duo™ (Dentsply, Degudent) and VITA Suprinity®, (VITA Zahnfabrik).

Production of the ceramic blanks and fabrication of the restoration go through three stages similar to those of lithium disilicate ceramic. In the first stage, the melted material is moulded, and the block is in the glass state. Nucleation/precrystallisation is a subsequent thermal pretreatment in which crystals start to form and grow, and the glass features ceramic properties. In this stage, the material contains only lithium metasilicate crystals and is easy to mill in the CAM unit. The final crystallisation stage is short heat treatment at 840°C for 8 min in which the restoration exhibits its final colour and physical properties. The crystalline phase consists of 25% lithium metasilicate

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(Li$_2$SiO$_3$) and 11% lithium disilicate (Li$_2$Si$_2$O$_5$) crystals. During the last two stages, zirconia acts as a nucleating agent but remains dissolved in the glassy matrix [25] [35] [36].

Despite the high silica content of 55-65 wt%, these ceramics have improved flexural strength of 370-420 MPa after glazing, due to the lithia (15–21 wt%) and particularly zirconia (8–12 wt%) content (approximately ten times more zirconium dioxide than in traditional CAD/CAM glass-ceramic) [37].

The specific composition has a positive effect on the optical properties of the material; consequently, aesthetic appearance 'as a natural tooth' of the restoration (natural opalescence, fluorescence and pronounced chameleon effect) is achieved. The lithium silicate crystallites in Celtra with a size of 0.5-0.7 μm corresponds to the wavelength range of natural daylight thus mimicking the opalescence behaviour of the tooth enamel, and together with the high glass content are responsible for the fluorescence of the restoration [38].

Values for the fracture toughness (2.31 ± 0.17 MPa m$^{0.5}$), flexural strength (443.63 ± 38.90 MPa), elastic modulus (70.44 ± 1.97 GPa) and hardness (6.53 ± 0.49 GPa) of Vita Suprinity are significantly higher compared to lithium disilicate ceramic [39].

Dissolving of the glassy matrix using hydrofluoric acid before adhesive lutting of the restoration made up from zirconia-reinforced lithium silicate causes exposure of the homogeneously and densely arranged bean-like lithium silicate crystals [19] [22].

Fluorapatite glass-ceramics (IPS e.max Ceram and IPS e.max ZirPress, Ivoclar Vivadent) contain fluorapatite crystals Ca$_5$(PO$_4$)$_3$F in various sizes embedded into the glassy matrix; the crystals, responsible for material's opalescence, are less than 300 nm in length and 100 nm in diameter, whereas crystals along the longitudinal axis are larger, 2–5 μm in length and less than 300 nm in diameter. IPS e.max Ceram is a nano-fluorapatite layering ceramic in the form of a powder, used for the production of veneers or as veneering material for glass or oxide ceramics, whereas IPS e.max ZirPress are pressable ingots suitable for the production of veneers and veneering of zirconia substructures using press-on-zirconia technique [40] [41].

The flexural strength of IPS e.max Ceram is significantly lower than IPS e.max ZirPress due to the pores present in the material, result of an air-bubbles' incorporation during mixing the ceramic powder with the mixing liquid [42]. Having high silica content of approximately 60 wt% and very low alumina content of ≈12 wt%, the flexular strength is lower (only 90-110 MPa) compared to other all-ceramic materials, which means that they can't be used for the production of the substructure of the restorations [40] [41].

Further development of glass ceramics is aiming to fulfil the protective requirements of the dental materials. ***Fluorophlogopite glass ceramic*** seems to have similar properties to other glass ceramics regarding mechanical properties, milling efficiency and etchability, but further on, this material possesses biocompatible property such as fluoride releasing. The flexural strength is approx. 120 MPa, the Weibull modulus is higher, while the Vickers hardness is lower compared to feldspar-based, lithium disilicate and resin-matrix ceramic [43].

Glass-Infiltrated ceramics

The glass-infiltrated ceramics belong to a group of ceramic-glass interpenetrating phase composites as they have at least two interpenetrating phases intertwined throughout the material [15]. The ceramic material is fabricated utilising the slip-casting or CAD/CAM technique. A slurry of densely packed ceramic particles is sintered to a refractory dye (if using the slip-casting technique) or the ceramic powder is dry pressing into a mould and compacted to produce a ceramic block (if using CAD/CAM technique) that are subsequently milled in CAM unit. The porous ceramic skeleton is infiltrated with lanthanum glass in a second firing, thus increasing the strength of the restoration [44].

Optical properties and final strength of the ceramic depends on the chemical composition of the porous core. *VITA In-Ceram^{TM} SPINELL*, consisted of alumina and magnesia (MgAl$_2$O$_4$), had the lowest strength (400 MPa) but very high translucency, thus indicated for single crowns in the anterior region. The alumina content in *VITA In-Ceram^{TM} ALUMINA* has reached 80% obtaining optimal translucency and strength (500 MPa) of the material, therefore indicated for single crowns in the anterior and posterior region, and for production of 3-unit bridges in the anterior region. Due to the alumina core strengthened with zirconia, *VITA In-Ceram^{TM} ZIRCONIA* had highest bending strength (600 MPa) compared to other glass-infiltrated ceramics and was indicated for single crowns in posterior region and 3-unit bridges no matter of the localisation [45]. No statistically significant difference has been found in the biaxial flexural strength and fracture toughness between In-Ceram ALUMINA and In-Ceram^{TM} ZIRCONIA [46].

The acid etching with HF acid did not change the superficial microstructure of these ceramics [30] [47] [48]. The use of this class of materials is abandoned due to the complexity and sensitivity of the manufacturing process, as well as the increased popularity of lithium disilicate ceramic and zirconia.

Polycrystalline ceramics

The main characteristic of ceramics classified in this group is a fine-grain crystalline structure without

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the glassy phase. The crystals are densely arranged into regular arrays, thus reducing the crack propagation, providing the material with high strength and fracture toughness [44]. The absence of the glassy matrix is the reason for ceramics' resistance to surface etching with hydrofluoric acid [49] [50].

Alumina

Aluminium oxide (Al_2O_3) is a naturally occurring mineral (corundum, bauxite) with a high Mohs hardness of 9. It is used in engineering as an abrasive material, cutting tools, electronic substrates, whereas in medicine, due to its biocompatibility, low friction and excellent wear and corrosion resistance, is used as a bone replacement material (production of hip joint balls) [51]. For years crystalline aluminium oxide has also been used to increase the stability of dental ceramics (so-called "dispersion strengthening").

Alumina exhibits the highest resistance to hydrolysis compared to other ceramic materials, low thermal conductivity and high flexural strength (> 500 MPa). The alumina blocks (consisting of 99.5% Al_2O_3) are initially manufactured partially sintered which allows easy processing-milling. Shrinkage, which occurs during the subsequent sintering process, can be exactly calculated, so precision-fit structures are obtained. Since grinding does not cause phase transformation in the structure, restorations can be reshaped in the sintered condition, with no need of subsequent regenerative firing [52].

With an elastic modulus of 380 GPa [53], alumina is prone to bulk fractures [54]. Furthermore, the increased use of materials with improved mechanical properties, such as stabilised zirconia and its feature for transformation toughening (thus the ability for crack "repairing"), has led to a decreased use of alumina.

Procera® AllCeram from Nobel Biocare (the first fully dense polycrystalline ceramic) and In-Ceram® AL, a product of VITA Zahnfabrik, are representatives of this type of ceramic.

Zirconia

Zirconium (Zr) is a shiny silvery metal. It is relatively soft and flexible when in a highly pure form. Its most important compound is zirconium dioxide ZrO_2, chemically an oxide and technologically a ceramic material. About 0.02% of the earth crust comprises of zirconia, with the largest deposits in Brazil and South Africa as baddeleyite (monoclinic zirconia) and high proportion in Australia and India where can be found as zircon ($ZrSiO_4$) sands [55]. Zirconia was discovered by the German chemist Martin Heinrich Klaproth in 1789 [13].

Pure zirconia is a polymorphic material that occurs in three crystallographic structures depended on the material's temperature. This phenomenon is known as allotropy since different structures have the same chemical composition but a different atomic arrangement. When cooling down from the molten state, following phases can be observed: cubic (*c*) from 2680°C, the melting point, to 2370°C; tetragonal (*t*) from 2370°C to 1170°C; and monoclinic (*m*) from 1170°C to room temperature [13] [56] [57] [58]. The spontaneous transformation from the *t* phase (higher material density) to the more–stable *m* (lower material density) phase is associated with a volume increase of 3% to 5%. Occurred tension (during the cooling phase after sintering) inside of the restoration made of pure ZrO_2 results in numerous microcracks, which will eventually lead to premature failure of the restoration [59] [60].

In the distant 1929, Ruff et al., [61] [62] have discovered that the tetragonal, or even the cubic form of zirconia could be retained metastable at room temperatures by alloying pure zirconia with other cubic oxides. Since then, numerous oxygen biocompatible compounds have been proposed as a zirconia stabilizers, used to relocate the phase transformation towards lower temperatures, thus preventing the catastrophic failure of the restorations made of zirconia: MgO [63] [64] [65] CaO [66], CeO_2 [67], Al_2O_3 [68] and Y_2O_3 [66] [69].

In 1975, Hannink et al., [69], proposed that the *t–m* transformation followed by volume expansion could be used to enhance the fracture toughness of partially stabilised zirconia-based materials. In 1976, Claussen and Steeb [70] had explained this mechanism as "oriented nucleation of microcracks". When a restoration containing metastable *t*-zirconia is subject to an external source of energy, as for example in the case of a tensile stress, temperature shock, an overloading in patient with parafunction, or in contact with water at low temperatures (condition occurring over time known as "low temperature degradation" LTD) [71] [72], the cracks may occur. Zirconium oxide grains are transformed from their tetragonal to the monoclinic form accompanied by a volumetric expansion of the grains thus restricting the crack. Since this expansion is constrained by the surrounding material, the net result is compressive stress on the surfaces of the crack, which propagation is thus hindered, eventually preventing the failure of the zirconia restoration [71] [73]. This is the reason why this phenomenon is called "phase transformation toughening" [69] [70] [71] [72] [73].

As "tension expansion" is a phenomenon otherwise known only in the case of steel, zirconium oxide was also referred to as "ceramic steel" [11].

In their review paper, Lughi and Sergo [71] summarising the scientific data concluded that the main factors affecting the zirconia ageing are the stabiliser type (oxides) and its content, the grain size and the residual stress. The most appropriate stabiliser is Y_2O_3 when added between 3.5 and 8

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mol%. Theoretical density of the material should be more than 99%, with the grain size less than 0.3μm and negligible monoclinic content. Residual tensile stress should be less than 300 MPa [71].

The most widely used stabiliser is Y_2O_3 in the content of 3 mol% (corresponds to 5.1% by weight) for the stabilisation of a tetragonal and 8 mol% for the stabilisation of a cubic form of Y-TZP [71]. It is well known that Y-TZP is prone to LTD in the presence of water. To date, there are several proposed mechanisms [60] [74] [75] that explain this phenomenon, but none of them is confirmed. Anyway, some facts are accepted regarding the origin and spreading of the micro-cracks: _t–m_ grain transformation starts from the surface of the material and then proceeds inward, causing a surface uplift [76] [77] and creating microcracks [78], that enables water penetration below the surface. Propagation of the _t–m_ grain transformation into the material [79], leads to the development of major cracks [78] that eventually ends with a catastrophic failure of the restoration. Low-temperature degradation of the Y-TZP has encouraged researchers to look for other stabilisers; aluminium trioxide, Al_2O_3, seems to have a crucial role in the ageing stability of Y-TZP ceramics and is used in very low content, with alumina particles optimally distributed within the zirconia material. It can be added independently of yttria stabiliser and its content, in the amount of 0.25 wt% having a higher degradation retarding effect to Y-TZP ceramics than 2 or 5 wt% of alumina addition, which have a comparable effect. According to Zhang et al., the apparent activation energy for the degradation process is increased by adding alumina in higher content [68].

Stabilization by cerium oxide provides better thermal stability and resistance to 'low-temperature degradation' than Y-TZP. CeO_2 gives zirconia the best properties regarding phase transformation although it is needed in larger amount comparing to Y_2O_3 to maintain the same degree of stability [80]. A Ce-TZP material with 8 mol% of ceria was characterised by less than 10% monoclinic content on the surface after 360 h water storage at 80°C [81]. When containing more than 12 mol% ceria, the system is almost non-transformable [82]. Another positive effect of adding ceria to zirconia is the pseudo-plastic behaviour of this compound (Ce-TZP can bend before fracturing) that is the most expressed feature among all other ceramic materials [83]. The major drawback of Ce-TZP ceramic materials that are commercially available is the chemical instability of Ce^{4+} that can be relatively easily reduced to Ce^{3+}, which does not have the same stabilising ability toward _t_-zirconia [84]. On the other hand, CeO_2 is yellow, affecting the colour of the final Ce-TZP restoration from light yellow to almost brownish, that may even become dark grey due to the high concentration of oxygen vacancies [84] [85].

Schmauder and Schubert [86] have shown that stress plays a critical role in the _t–m_ transformation of zirconia grains as it does not occur in stress-free zones of the material, even if it contains an insufficient amount of stabiliser. As mentioned before, stress can be "applied" (tensile stress, temperature shock, an overloading in patient with parafunction), but it may also appear as a "residual stress" occurring during fabrication process of the restoration when it is fired at high temperature and then cooled down to room temperature, during sintering or veneering with materials having a different coefficient of thermal expansion (CTE) [71]. The residual stress has so-called "autocatalytic effect" that can be explained by prolonged _t–m_ transformation. When some _t_-zirconia grains transforms to _m_-phase for any reason, zirconia becomes two-phase material with _m_-zirconia in compression and the remnant _t_-zirconia in tension; it is assumed that for every 10 vol% of _t–m_ transformation, the tensile stress in the remnant _t_-zirconia increases by 250 MPa [87]. Because of this tensile stress, _t_-zirconia will be more prone to further transformation to _m_-zirconia. Experimentally, it was confirmed that the tensile stress initiated by _t–m_ transformation as a result of the material bending, could reach a value of 700 MPa [88]. The outcome is slow increasing of the monoclinic content inside the zirconia.

Another factor that influences the stability of the tetragonal phase and LTD is an average size of the zirconia grains. Reducing the grain size (GS) has a beneficial effect on the stability of the zirconia-based materials; a reduction below a certain critical value has a potential of fully inhibiting LTD [71]. When at room temperature, the grains' critical size of pure zirconia powder is in the range of 5–10 nm, whereas in 1.5 Y-TZP powders it is about 90nm [89]. Anyway, when solid, dense zirconia material is observed, "the _t_-form can be stable even if the grains are much larger than the critical GS identified above for powders"; however, zirconia GS should be less than 300 nm to prevent LTD over a period [71]. Exact calculation of optimal GS in a solid material is complex, as the effect of different stabilisers and their content, as well as strain energy, should also be taken into consideration.

Sintering process, temperature and the dwell time influence the ageing stability of zirconia as well. Hallmann et al., [90], didn't find _t–m_ phase transformation when Y-TZP has been sintered at a temperature of 1350°C. The diffusion-controlled _t–m_ phase transformation initiated at 1450°C and became remarkable when the sintering temperature was increased to 1600°C [90]. Similarly, Inokoshi et al., [91] have demonstrated that increasing the sintering temperatures and elongating the dwell time (2 or 4 hours), increased the ZrO_2 grain size and decreased yttria content in the remaining tetragonal grains, which eventually lead to a higher sensitivity to LTD of Y-TZP ceramics. The best resistance to low-temperature degradation can be achieved by sintering at 1450°C with a one h dwell time [91]. According to Hjerppe et al., the bi-axial flexural strength of zirconia was not

affected by decreasing the sintering time or thermocycling, although the amount of monoclinic phase on the surface of zirconia was increased after thermocycling and water storage [92].

The effect of increased sintering temperature (from 1350°C to 1550°C) on the LTD sensitivity can be explained by the following mechanism: it causes increasing of the average size of the tetragonal ZrO_2 grains, whereas increasing content of the cubic zirconia grains that are enriched with yttrium concomitantly results in a decreased yttrium content as a stabilizer in the remaining tetragonal grains [93].

However, decreasing the sintering temperature to obtain 3Y-TZPs resistant to LTD, leads to the creation of a material with moderate mechanical properties, i.e. lower fracture toughness. To improve surface degradation resistance, Zhang et al., [94] proposed a new way of material production, incorporating stabiliser by coating yttria in the starting powder of 3Y-TZP, a method different than co-precipitation. Optimization of the mechanical properties can be achieved by adding 0.25 wt% Al_2O_3 that allows sintering at a lower temperature to obtain a higher LTD resistance without compromising the hardness and the fracture toughness. The improved LTD resistance in such material could be attributed to the segregation of Al^{3+} at the grain boundary and the heterogeneously distributed Y^{3+} stabiliser [94].

But, not only sintering conditions influence the LTD behaviour of the zirconia materials. Surface treatments (ST) of dental zirconia conducted before adhesive luting, have a great impact on the monoclinic content and thereby ageing sensitivity, i.e. degrading susceptibility. Some monoclinic grains have been observed after sandblasting the surface, with no particular influence of the abrasive particles' size on the LTD of Y-TZP [90]. Anyway, the effect of surface treatment mostly depends on the chemical composition-oxides used as stabilisers. Inokoshi et al., have shown that ST improved the ageing resistance of Y-TZP zirconia, although the highest monoclinic volume fraction was observed in Al_2O_3 sandblasted zirconia. The LTD behaviour of Ce-TZP/Al_2O_3 zirconia (with higher initial monoclinic volume fraction after ST, but stronger ageing resistance compared to the Y-TZP zirconia) was not affected by conducted treatments. Y-TZP/alumina (Y-TZP/Al_2O_3) zirconia showed a strong ageing resistance when the surface was left untreated; insignificant degradation was observed after ST [95].

All the factors mentioned above influence the crystallographic structure of the zirconia when at room temperature. The grains' transformation from tetragonal to monoclinic form is a one-way process, meaning that "once it takes place, the crack-hindering effect cannot be exploited for limiting further fractures" [2]. Restorations made of zirconia with monoclinic state of the grains are more prone to catastrophic failure. The solution for such a negative outcome has been found in the so-called "regeneration firing" that is conducted after final adjustment of the already sintered restoration. Annealing the material in the temperature range 900–1000°C for a short time, induces the reverse phase transformation from monoclinic back to tetragonal form [96].

Beside LTD, shading of the zirconia frameworks before sintering also has a negative effect on the biaxial strength and surface microhardness. Some reductions in bi-axial flexural strength, as well as dimensional changes, are seen when colouring zirconia in green-stage form. Shaded zirconia has lost less weight after sintering than un-shaded, indicating that colourant particles have been incorporated in the crystal structure of the material. The shrinkage of zirconia during sintering is also diminished by shading, which "might have clinical effects as the marginal accuracy, fit and cement thickness of the shaded is different to the un-shaded substructures" [97].

There are different types of zirconia materials depending on the zirconia's grains phase statement (composition), but only three are used in dentistry: (1) *partially stabilized zirconia (PSZ)*, a two-phase material with the tetragonal phase (transformable *t*-zirconia grains) precipitated into the cubic matrix, where stabilization is accomplished by magnesia [44] [71]; there is only one available MgO-PSZ product for the dental market, Denzir-M® by Dentronic AB, Skellefteå, Sweden, suitable for hard machining. (2) *Zirconia toughened composites (ZTC)* - a matrix with high elastic modulus embedding transformable *t*-zirconia grains; the most used matrix is alumina, so the material is known as zirconia-toughened alumina (ZTA) (this material is mostly used for the medical prostheses fabrication) [98]. (3) *Tetragonal zirconia polycrystals (TZP)* - the whole material consists of transformable *t*-zirconia grains. Stabilized with yttria as Y-TZP, has been used in dentistry for production of orthodontic ceramic brackets [99] and endodontic posts [100] since the early 1990s, as well as fixed restorations since the later 1990s [101]. PSZ and ZTC are two-phase materials, whereas TZP is a single-phase [71].

As mentioned before, zirconia blocks for the CAD/CAM technology could be used in their pre-sintered or sintered state. For better quality of the restoration, it is better if *pre-sintered* chalk-like blocks are used (in so-called *'green' stage*), with porosity in their microstructure (50% for IPS e.max ZirCAD), so the milling process is easier, the average milling time is reduced, and the milling tools can be used longer. After milling in the CAM system, enlarged crown and bridge substructures undergo sintering process (1350–1500°C). During the sintering, shrinking of the restorations (20–25%) occurs, causing the structure densification to more than 99%, so the final properties of the material are achieved [102].

Zirconia blocks for computer-aided-

manufacturing can also be obtained from the manufacturers as industrially sintered; *hot iso-static pressed 'HIP' zirconia* (known as 'white blocks'). This material is in its final high strength and is characterised by a constant grading and better homogeneity, with no need of further sintering after milling [103]. According to Stawarczyk et al., hipped zirconia has the highest flexural strength compared to several pre-sintered zirconia materials tested after sintering [104]. Anyway, milling time is longer and wear of the milling tools is higher compared to milling of pre-sintered zirconia blocks [53]. The most important thing when milling hipped zirconia is substantial amounts of surface and structural defects caused by diamond burs, which negatively impact the permanent strength and durability of the ceramic.

Zirconia-based materials have the highest strength, fracture resistance, fracture toughness and Vickers hardness among all other ceramic materials: high fracture toughness of 6–15 $MPa \cdot m^{-0.5}$, flexural strength of more than 900 MPa, high Vickers hardness of 1200–1350 HVN, high thermal expansion of more than $10 \cdot 10^{-6}$ K^{-1}, very low thermal conductivity of less than 2 W/mK (vs. 200 W/mK for gold alloy, and 40 W/mK for base metal), and a good thermo-shock resistance of $\Delta T = 400–500°C$ [105].

Resin-Matrix Ceramics

In the last six years, a new category of dental materials has been promoted, hybrids, consisting of an organic matrix highly filled with ceramic particles [6] [7] [15] [106].

These materials can be included in a classification system of dental ceramics [7] as the 2013 version of the American Dental Association Code on Dental Procedures and Nomenclature [107] defines the term porcelain/ceramic as "pressed, fired, polished, or milled materials containing predominantly inorganic refractory compounds - including porcelains, glasses, ceramics, and glass-ceramics". The former version (2012) of the referred code, didn't allow materials with resin matrix to be classified as ceramic materials, as it has defined porcelain/ceramic as "non-metal, non-resin inorganic refractory compounds processed at high temperatures (600°C and above) and pressed, polished, or milled, including porcelains, glasses, and glass-ceramics". As these hybrid ceramics are composed predominantly (> 50% by weight) of inorganic compounds, it would be justified if they are taken into consideration while making systematisation of dental ceramic materials.

First, of them, Lava Ultimate from 3M ESPE has been marketed as *'Resin Nano Ceramic' (RNC)* as it contains nanoceramic particles (nanomer and nanocluster particles) bound in a highly cross-linked polymeric matrix. Lava Ultimate contains two types of nanomers that are monodispersed, nonaggregated and nonagglomerated: silica nanomers of 20 nm diameter and zirconia nanomers of 4 to 11 nm in diameter. The nanocluster particles (with the average size of 0.6–10.0 μm) are synthesized and composed of 20 nm silica nanomers and 4–11 nm zirconia nanomers. Nano-dimension of the particles, allows a high proportion of ceramic filler (approximately 80% by weight) to be incorporated into the resin. Both of them, nanomers and nanoclusters are treated with a silane coupling agent so that chemical bonds can be provided between ceramic particles and the resin matrix. The material is processed several hours in a special heat treatment process, which results in highly cured material, so there is no need of further firing after milling [108].

Specific composition and production technology resulted in a material with higher flexural strength (200 MPa), fracture and wear resistance than composite materials (provided by nanoclusters), and with significantly improved polishability and optical properties (because of the nanoparticles). The polymeric resin as a matrix contributes to some properties that composites have: the material is not brittle and is fracture resistant, with shock absorbing characteristics. Despite a high ceramic content, this material is not recommended for the production of crowns, but only for inlays, onlays and veneers [108].

Intact resin matrix with embedded ceramic nano-particles could be observed after HF etching of the material's surface [19] [22].

Taking into consideration a specific structure and composition of the spongy bone and dentin, consisted of relatively weak and brittle inorganic constituents and organic matrix that provides elasticity, a novel kind of interpenetrating phase material has been developed [15] [109] referred as *'Polymer Infiltrated Ceramic Network' (PICN)*.

VITA Enamic from VITA, have two 3-dimensional network structures interpenetrating one to another; the dominant fine-structure feldspar ceramic network (86% by weight or 75% by volume) is strengthened by a polymer network consisting of methacrylate polymer (14% by weight or 25% by volume). The blocks are manufactured in few steps: first, the ceramic powder is initially compressed into blocks followed by a sintering process to obtain a porous ceramic network. Next, a ceramic base structure is conditioned with a coupling agent. The conditioned porous inorganic network is finally infiltrated with a monomer mixture, followed by heat-induced polymerisation to create a polymer network. Both of the networks are interconnected through the chemical bonds obtained by the coupling agent [15] [110].

To reproduce the natural shade gradient, in 2017, VITA has started production of the VITA Enamic multiColor, blocks with six finely nuanced layers, from cervical to incisal area [111].

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Table 1: Composition, properties and clinical indications of CAD/CAM blocks as published by representative manufacturers

Ceramic type	Product name / Manufacturer	Chemical Composition mass%		Flexural strength MPa	Modulus of elasticity GPa	Clinical indications
Feldspar	VITABLOCS®, VITA Zahnfabrik: Mark I (1985) Mark II (1991) VITA TriLuxe (2003) VITA TriLuxe forte (2007) VITA RealLife (2010)	SiO₂ Al₂O₃ Na₂O K₂O CaO TiO₂ pigments	56-64 20-23 6-9 6-8 0.3-0.6 0.0-0.1 <0.1	154	45	veneers, inlays, onlays, partial crowns, anterior and posterior crowns, as a veneering CAD/CAM material for multi-unit bridge substructure made of oxide ceramic
Leucite	IPS Empress CAD, Ivoclar Vivadent (2006) IPS Empress CAD Multi	SiO₂ Al₂O₃ K₂O Na₂O BaO, CaO, CeO₂. B₂O₃, TiO₂ pigments	60.0-65.0 16.0-20.0 10.0-14.0 3.5-6.5 0.5-7.0 0.2-1.0	160	62	veneers, inlays, onlays, partial crowns, anterior and posterior crowns
Lithium-disilicate	IPS e.max CAD, Ivoclar Vivadent (2006)	SiO₂ Li₂O K₂O P₂O₅ ZrO₂ ZnO Al₂O₃ MgO pigments	57.0-80.0 11.0-19.0 < 13 < 11 < 8.0 < 8.0 < 5.0 < o 5.0 < 8	360±40	95	veneers, inlays, onlays, partial crowns, anterior and posterior crowns, 3-unit bridges (anterior and premolar), hybrid abutments, hybrid abutment crowns; 3-unit posterior bridges, as a veneering CAD/CAM material of multi-unit bridge substructure made of IPS e.max ZirCAD
Lithium-silicate	Celtra Duo, Dentsply (2013)	ZrO₂	10	370		veneers, inlays, onlays, partial crowns, anterior and posterior crowns, implant-supported crown
	VITA Suprinity®, VITA Zahnfabrik (2013)	SiO₂ Li₂O ZrO₂ P₂O₅ Al₂O₃ K₂O CeO2 pigments	56-64 15-21 8-12 3-8 1-4 1-4 0-4 0-6	420	70	veneers, inlays, onlays, partial crowns, anterior and posterior crowns, implant-supported crown
Zirconia	Vita In-Ceram® YZ, VITA Zahnfabrik (2002)	ZrO₂ Y₂O₃ HfO₂ Al₂O₃ SiO₂ Na₂O	91-94 4-6 2-4 <0.1 <0.1 <0.1	>900	210 GPa	primary telescope crowns, anterior and posterior crowns, anterior and posterior bridge (up to 2 pontics), cantilever bridges
	Lava™ Frame Zirconia, 3M ESPE (2001)	ZrO₂ Y₂O₃ Al₂O₃		>1100	210	anterior and posterior crowns, splinted crowns up to 4 units, implant abutments, crowns on implant abutments, 3-unit inlay and onlay bridges, cantilever bridges, anterior adhesive bridges, 3–4 unit bridges , long-span and curved bridges
	IPS e.max ZirCAD, Ivoclar Vivadent (2006)	ZrO₂ Y₂O₃ HfO₂ Al₂O₃ other oxides	87-95 4-6 1-5 0-1	900±50		anterior and posterior crowns, primary telescope crowns, implant superstructures, 3-unit bridges (anterior, premolar and posterior), multiple-unit bridges, inlay bridge frameworks
All Zirconia	Lava™ Plus High Translucency Zirconia, 3M ESPE (2012)	ZrO₂ Y₂O₃ Al₂O₃	>90 3 0.1	>1100	210	anterior and posterior crowns, splinted crowns up to 4 units, implant abutments, crowns on implant abutments, primary crowns, 3-unit inlay and onlay bridges, cantilever bridges, anterior adhesive bridges, 3–4 unit bridges , long-span and curved bridges
	Cercon® ht True Color, Dentsply, Degudent (2015)	ZrO₂ Y₂O₃ HfO₂ Al₂O₃ SiO₂	5 <3 <1	1200	210	anterior and posterior crowns, primary telescope crowns, multi-unit bridges
	Zenostar® Full Contour Zirconia, Wieland Dental/Ivoclar Vivadent (2013)	ZrO₂ Y₂O₃ HfO₂ Al₂O₃	>99 4.5-6.0 <5 <1	1200±200	210	anterior and posterior crowns, primary telescope crowns, multi-unit bridges (as frameworks or full-contour)
Hybride	Lava™ Ultimate CAD/CAM Restorative, 3M ESPE (2011)	Ceramic: ZrO₂ SiO₂ Resin: Bis-GMA, UDMA, Bis-EMA, TEGDMA	% %	204	13	veneers, inlays, onlays
	VITA Enamic®, VITA Zahnfabrik (2013) Vita Enamic® multiColor (2017)	Ceramic network: SiO₂ Al₂O₃ Na₂O K₂O B₂O₃ CaO ZrO₂ KaO TiO₂ Polymer network: UDMA TEGDMA	wt%: 58-63 20-23 9-11 4-6 0.5-2 0.3-0.6 < 1 < 1 <0.1 66 33	150-160	30	veneers, inlays, onlays, anterior and posterior crowns, implant-supported crowns
	CERASMART™, GC (2014)	Ceramic network: SiO₂ Barium glass Monomer: Bis-MEPP UDMA DMA	71 29	231		veneers, inlays, onlays, anterior and posterior crowns, implant-supported crowns

Bis-GMA: bisphenol A diglycidylether methacrylate; Bis-MEPP: 2,2-Bis(4-methacryloxypolyethoxyphenyl)propane; UDMA: urethane dimethacrylate; TEGDMA: triethylene glycol dimethacrylate; Bis-EMA: ethoxylated bisphenol-A dimethacrylate; DMA: dimethacrylate.

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The flexural strength of this two-phase material can reach a value of about 150–160 MPa, significantly higher than that of a porous ceramic (below 30 MPa) and polymer (135 MPa) alone [112]. The superior flexural strength of the resin-infiltrated ceramic material compared to the single components implies a reinforcement mechanism of the polymer network to the dominant ceramic network [106] [112]. Elastic modulus, hardness, and fracture toughness values are as follows: 30.14 GPa, 2.59 GPa, and 1.72 MPa·m$^{-0.5}$ respectively [5], all of them between those obtained for human dentin and enamel. With the highest filler content (73.1 mass %) compared to other hybrid ceramics and composites, Vita Enamic has the highest Vickers hardness of 189.8 [113]. Moreover, VITA Enamic has similar two-body and tooth-brushing wear characteristic to natural enamel [114]. All these features, together with the possibility material to be milled very thin thus preserving the tooth structures, are in favour of using this material in patients with erosions where tooth preparation is not recommended [115].

HF etching of VITA Enamic causes dissolving of the superficial ceramic network, so that acrylic polymer network became visible with scattered irregular ceramic particles [19] [22].

The third, CERASMART™ from GC, referred to as **"Flexible Nano Ceramic"**, is composed of relatively small and uniformly distributed particles of alumina-barium-silicate embedded in a polymer matrix [116]. The flexural strength (approx. 242 MPa) is significantly higher, whereas the flexural modulus (10.0 GPa) and Vickers hardness (approx. 64.1 HV) are significantly lower than Lava Ultimate (170.5 MPa / 14.5 GPa / 97.9 HV) and VITA Enamic (140.7 MPa / 28.5 GPa /189.8HV) respectively [113]. The modulus of resilience of 3.07 ± 0.45 MPa is the highest, and the margin edge roughness of 60 ± 16 μm is the lowest compared to other ceramics or hybrid materials [117].

All hybrids have a modulus of elasticity similar to dentin, and modulus of resilience significantly higher than feldspar-based and glass-ceramics, thus significantly higher stress can be absorbed without permanent deformation or failure. That is why hybrids are the most recommended materials for fabrication of crowns over implants, where the periodontal ligament (tissue that acts as a shock absorber) is already lost.

Milling time in the CAM unit is shorter compared to other ceramic materials, with longer lifetime on the milling burs. There is no need for sintering or crystallization firing after milling; final gloss and smoothness of the restoration can be achieved by surface polishing. Hybrids are wear resistant and "gentle" to the opposite dentition [115]. Restorations can be easily repaired in the mouth, although these materials are characterized by virtually no chipping.

Clinical Indications

Feldspar-based and leucite-reinforced ceramics according to their low flexural strength (154MPa and 160MPa respectively) are indicated for single tooth restorations such as veneers, inlays, onlays, partial crowns, and anterior and posterior crowns. Beside the listed, zirconia-reinforced lithium silicate (420 MPa) can also be used for the fabrication of implant-supported crowns. Having the high crystalline content, specific microstructure and flexural strength of 360–400 MPa, lithium disilicate ceramic can be used not only for single tooth restorations but the fabrication of hybrid abutments, hybrid abutment crowns and three-unit bridges as well (up to the second premolar as the terminal abutment). Due to the polymer content, low flexural strength (150–240 MPa) and high resilience, hybrid ceramics can be used only for single tooth restorations such as veneers, inlays and onlays (Lava Ultimate), as well as for anterior and posterior crowns and implant-supported crowns (VITA Enamic and Cerasmart).

Densely sintered, high–purity alumina can be used for primary elements of conical and telescopic crowns, crowns in the anterior and posterior area, as well as for bridges only in the anterior area with no more than 1 pontic (due to the average high flexural strength of 500 MPa).

Yttria-stabilized zirconia (with high flexural strength of more than 900 MPa) is indicated for fabrication of anterior and posterior crowns, implant abutments, implant abutments crowns, primary telescope crowns, 3-unit inlay and onlay bridges, cantilever bridges with minimum two abutment teeth and maximum of one pontic of no more than one premolar width, anterior adhesive bridges, as well as multi-unit long-span (up to 14 units) and curved bridges with a maximum of four pontics next to one another in the anterior area, and a maximum of three pontics (DC-Zircon) next to one another between abutment teeth in the posterior area. Some of them (Lava™ Plus High Translucency Zirconia, 3M ESPE) are indicated for clinical situations with limited interocclusal space, as well as when the tooth-preserving preparation is needed (minimum 0.5 mm occlusal wall thickness). CAD/CAM ceramic materials, their chemical composition, some properties and clinical indications as claimed by the respective manufacturers are presented in Table 1.

Conclusion

Although it seems that current ceramic materials could "cover" every clinical situation where

single-tooth restoration and fixed partial dentures are indicated for use, there are some limitations that should be taken seriously. All-ceramic restorations are not recommended in patients with insufficient remaining natural tooth substance (short clinical crowns), sub-gingival preparations (mainly for adhesive luting), inadequate oral hygiene, and patients diagnosed with excessive masticatory functions, in particular teeth grinders and clenchers.

The future of all-ceramic materials is bright. Further improvements in chemical composition, internal structure, grain-size decreasing to nano dimensions and improved protocols for industrial production and laboratory processing, all of them will for sure lead to a material with extraordinary features fulfilling the esthetic, mechanical and biocompatible demands.

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