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Microstructural study of microwave sintered zirconia for dental applications

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Abstract

Conventional sintering techniques for zirconia-based materials, which are commonly used in dental reconstruction, may not provide uniform heating, with the consequent generation of microstructural flaws in the final component. A sintering system using microwave heating may represent a viable alternative. The purpose of this study was to compare the dimensional variation and physical and microstructural characteristics of commercial zirconia (Y-TZP), used as a dental restoration material, sintered in conventional and microwave furnaces. A physical-mineralogical-microstructural characterisation was carried out to evaluate the level of densification and the presence of flaws in the sintered specimens. Use of the microwave systems allowed the length of the sintering cycle to be reduced to a few minutes, compared with the several hours necessary with a 'traditional' heating system. Additionally, the maximum temperature used to reach the required density decreased from 1450–1480 °C with the electric furnace to 1200 °C in the microwave furnace. An important clinical implication is that the reduced sintering time could allow the introduction of zirconia in chair-side treatments, if used as a monolithic material.

Keywords: Microwave sintering; Y-TZP; Dental ceramic; Microstructure

1. Introduction

A well-known review on zirconia for dental applications [1] noted that in the last 20 years, the diffusion of metal-free restorations in dental practice has increased considerably due to the growing demand for highly aesthetic and natural-appearing components. Bioceramics [2] are particularly suitable for the use in prosthodontics as possible metal substitutes because of their wear resistance, superior mechanical properties, high biocompatibility, and excellent aesthetic appearance.

Particular attention has been paid to "yttria-tetragonal zirconia polycrystalline ceramics" (Y-TZPs), which have been used as framework materials for dental crowns and fixed partial dentures (FDPs), because their aesthetic appearance is similar to that of natural teeth and their mechanical characteristics are good: indeed, the highest reported for any dental ceramic [3]. Both the chemistry and processing of these materials allow obtaining a fully dense polycrystalline zirconia, in a tetragonal phase, with a homogeneous distribution of submicron zirconia grains, giving a translucent aspect, which meets the requirements for natural teeth-looking restorations [2]. Furthermore, compared with other ceramics, 3 mol% yttria-stabilised tetragonal zirconia polycrystals (3Y-TZP) are characterised by high fracture toughness and flexural strength, caused by a stress-induced phase transformation, from a tetragonal (t) phase to a monoclinic (m) phase, which increases its crack-propagation resistance [4].

The combination of these particular mechanical and physical characteristics has allowed Y-TZP to become the core material of choice for many categories of dental ceramic restorations. Additionally, CAD/CAM system technology has solved the difficulties of processing this hard material by working it when partially sintered or presintered [5]. In the usual practice of dental laboratories, pre-sintered Y-TZP blocks are rapidly

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CAD/CAM-converted into restoration components that require a final firing step both to reach a higher density and to eliminate any stress induced by the strong surface working actions [6].

This final sintering is currently performed in large, electrically heated ovens to a high temperature, followed by slow cooling to prevent cracking, which is a time-consuming and expensive procedure. The thermal cycles typically indicated by the manufacturers are characterised by maximum temperatures of 1350–1550 °C, at which the ceramic components remain for almost 60 min. Due to the slow cooling step, the total sintering cycle is generally 6–10 h.

Many ceramic powders can be sintered using microwaves at lower temperatures and for shorter times than with conventional electrical heating sources, allowing finer and more uniform microstructures to be obtained. Microwave irradiation, also known as dielectric heating, as applied to the sintering of advanced ceramic components, has become an important topic of scientific research [7–10].

In conventional sintering, heat is transferred from the radiant elements of the furnace to the surface of the ceramic component, reaching the core of the component through conduction mechanisms. In microwave sintering, the heat is produced as a consequence of an interaction between the ceramic sample and the electromagnetic waves and involves the whole sample volume; in this way, the heating is more rapid and uniform [11].

The extent of the energy transfer from the electromagnetic field to the matter depends strongly on the dielectric properties of the material, the temperature, and the radiation frequency [12–15]. The microwave sintering of yttria-doped zirconia has been investigated extensively using several types of microwave furnace. Due to its poor coupling with microwaves below 400 °C and moderate coupling at higher temperatures, it is necessary to use susceptors that absorb microwaves at room temperature and act as heating elements to increase the initial temperature of the material to the critical value at which it starts to absorb more effectively. For this purpose, several studies have reported the use of silicon carbide susceptors, because they absorb microwave energy and subsequently transfer it, in the form of heat, to the material, via conduction. This approach, often referred to in the literature as ‘hybrid microwave heating’, uses hybrid heating devices such as multi-mode cavities with SiC rods [16] or SiC powder [17] acting as a susceptor, or microwave/conventional hybrid furnaces [18].

When samples are heated in an electric furnace or a microwave furnace, two methods can be used to control the temperature: 1) intermittent powering of the magnetron at a fixed power output (on/off control method or time-control method), or 2) continuous powering of the magnetron with a variable power output (power-control method). The first method involves the use of the magnetron at its highest output power as typically programmed in domestic ovens, while the second is commonly used in industrial processes, where continuous adjustments of output power are necessary to follow the desired heating profile. It has been pointed out that there is no difference between the two methods in terms of grain growth or sample densification level, but the power-control method gives more precise control of temperature versus the on/off control method [19].

The multiplicity of experimental procedures reported makes obtaining a clear view of the behaviour of zirconia powder during microwave sintering difficult, despite the useful review in [20]. Regarding the sintering of nano Y-TZP, hybrid conventional-microwave heating sintering allowed obtaining $>99\%$ TD dense ceramics with an average grain size of less than 100 nm [18], but also near-theoretical density values for 3Y-TZP using a multi-mode microwave sintering furnace at 2.45 GHz [21]. Microwave sintering has been also proposed to overcome the problem of grain growth associated with conventional heating in nanocrystalline 3Y-TZP, by obtaining more homogenous microstructures with higher mechanical characteristics [22]. Further improvements in the physical and mechanical properties of Y_2O_3-ZrO_2 ceramics have been achieved through the use of nanopowders and the use of microwave sintering [23].

The results of these studies underlined that, compared with conventional sintering, the use of microwaves provides several advantages, such as rapid and volumetric heating, lower heating temperatures, enhancements in densification, grain growth limitations, and cost savings.

The aim of this work was to assess the possibility of using hybrid microwave sintering with a 3Y-TZP pre-sintered material for dental applications. Two microwave methods, multi- and single-mode, were used for the tests, both of which were fed with continuous variable power to better control the process temperature. The results indicated that the density of the microwave-fired samples depended strongly on the firing changes, which, once optimised, allowed the generation of highly dense zirconia samples with a firing time of only a few minutes.

2. Material and methods

A 3% Y-TZP pre-sintered commercial material (Biotech Srl, Milano, Italy, one of the most utilised in Italian Dentistry and in some countries of Europe), suitable for shaping using CAD-CAM technology, was used for the sintering tests. From the commercial supplies, provided in the form of cylinders, rectangular specimens, of about $20\times10\times14\ \mathrm{mm}^3$, were cut with an high-precision electric saw (Isomet 1000 Precision Saw, Buehler Ltd., Düsseldorf, Germany) and subjected to three heating treatments: conventional, multi-mode, and single-mode microwave sintering.

Conventional sintering was conducted in an electric furnace, using the following sintering cycle: 12 °C/min up to 300 °C, 5 °C/min up to the maximum temperature, holding time 60 min, with natural cooling. After several preliminary trials, two maximum temperatures were used: 1450 and 1480 °C. These sintering cycles required ~10 h at either maximum temperature.

Microwave sintering was performed with a commercial CEM-MAS 7000 multi-mode applicator (CEM Corporation, Matthews, NC) at 2.45 GHz (950 W, nominal power) and on a TE10 n single-mode applicator (0.5–3 kW output power), connected to a 2.45–GHz TM030 microwave generator (Alter Power System, Long Beach CA). The multi-mode applicator