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Fei Zhang, Helen Reveron, Benedikt Spies, Bart van Meerbeek, Jérôme Chevalier. Trade-off between fracture resistance and translucency of zirconia and lithium-disilicate glass ceramics for monolithic restorations. *Acta Biomaterialia*, 2019, 91, pp.24-34. 10.1016/j.actbio.2019.04.043 . hal-02395508

HAL Id: hal-02395508

<https://hal.science/hal-02395508>

Submitted on 22 Oct 2021

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Trade-off between fracture resistance and translucency of zirconia and lithium-disilicate glass ceramics for monolithic restorations

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Abstract

High strength and translucency are generally not coincident in one restorative material and there is still a continuous development for a better balance between these two properties. Zirconia and lithium-disilicate glass-ceramics are currently the most popular alternatives for monolithic restorations. In this work, the mechanical properties and more important, the slow crack growth (SCG) resistance, which rules long-term durability, were thoroughly studied for three zirconia ceramics stabilized by 3, 4 and 5 mol% yttria in comparison to lithium-disilicate glass-ceramic. Translucency versus strength maps revealed that the more translucent zirconia compositions (i.e. with higher yttria contents) fill the gap between the standard 3 mol% yttria stabilized zirconia (3Y-TZP) and lithium-disilicate. Moreover, increasing yttria content did not always result in lower

strength, as values for 3 mol% and 4 mol% yttria were the same. Independent on the yttria contents, all zirconia showed similar relative **susceptibility** to SCG under static and cyclic conditions and were significantly more SCG-resistant than lithium-disilicate glass ceramic. A concern with higher yttria contents (5 and 4 mol%) however could lie in the higher sensitivity to defects, resulting in a larger **scatter** in strength.

Graphical abstract

Keywords: Yttria-stabilized zirconia; Monolithic restorations; Slow crack growth; Mechanical properties; Translucency.

1. Introduction

All-ceramic restorations are popularly applied in dentistry, thanks to their excellent biocompatibility and natural appearance as compared to conventional porcelain-fused-to-metal (PFM) crowns and bridges, this together with the benefit of CAD/CAM for faster fabrication. Ceramic materials are inherently brittle and vulnerable to crack propagation, which may limit their indication. Zirconia ceramics, in particular 3 mol% yttria stabilized tetragonal zirconia (3Y-TZP), can however exhibit high strength (~1000 MPa), which allows the fabrication of long-span fixed dental prosthesis (FDP) in load-bearing areas [1, 2]. On the other side, most 3Y-TZP are insufficiently translucent and need to be veneered by a quite thick layer of feldspatic porcelain in order to meet the patient's individual tooth characteristics [3]. More recently, there is a clear trend toward monolithic ceramic restorations, avoiding difficulties with the veneering ceramics, i.e. the clinically rather regularly encountered chipping of bi-layered zirconia ceramics, and enabling a less invasive tooth preparation. Monolithic restorations were reported to have become the first choice for posterior teeth in the US since their introduction in 2014 [4]. In order to use ceramics as monolithic restorations with or without a glaze layer, the optical properties of the material itself have to approach that of natural teeth with high translucency being emphasized as one of the primary parameters in controlling optical appearance [5].

3Y-TZP ceramics in general allow less light transmission than glasses or glass-ceramics. Initially, they were even considered as an opaque material [6]. Zirconia doped with 3 mol% yttria is polycrystalline and mostly in the tetragonal crystallography (which explains the denomination often used of 3Y-TZP for 3 mol% Yttria-doped Tetragonal Zirconia Polycrystal). Thus, light scattering occurs at grain boundaries due to birefringence, as well as at residual pores even if present in low percentage. After years of development, many methods have been proposed to improve their translucency, including reduction of porosity acting as light scattering defects or the alumina content,

using nanostructured zirconia, and very recently increasing the yttria content [7, 8]. A huge amount of different zirconia ceramics (tetragonal zirconia or TZPs, and partially stabilized zirconia or PSZs) are thus introduced in the dental field and numerous studies report the properties of various high-translucent zirconia ceramics (Table S1 in supplementary document). Increasing yttria content effectively enhances the translucency of yttria-stabilized zirconia but lowers the strength (less or no phase transformation toughening) [9, 10], by which zirconia with a high yttria content can only be used in low stress-bearing areas. However, the current positioning of the newest monolithic zirconia generations in terms of mechanical and optical properties is not clearly and fully defined.

Moreover, **few studies have been focused on slow crack-growth behavior and fatigue [11-13]**, while it is especially important to assess the long-term load-bearing capacity of crack-sensitive ceramics [14]. Standards, solely requiring minimum mechanical properties that are measured under fast fracture conditions (referred to as the inert or initial mechanical strength of the materials [15-17]) should not be considered sufficient for safe use in patients, since the humid environment and cyclic loading in the oral environment can degrade the restoration's load-bearing capacity over time. Chemically-assisted slow crack growth (SCG) occurs due to the presence of water molecules (or other corrosive species, like ammonia or polar solvents), which promote breakage of the strained ionic-covalent bonds at the crack tip, i.e. stress-corrosion reaction, and initiate crack propagation at stress intensities (K_I) below the material's fracture toughness (K_{IC}) [18-20]. Cyclic loading induces fatigue in ceramics due to degradation of extrinsic toughening effects [14]. Clinic and lab reports show that dental restorations fail either due to chipping or fracture of the whole structure; such failures often occur at a load far below the initial strength owing to SCG and cyclic fatigue [12, 21]. In particular, in case radial cracks appear at the cementation surface, SCG would very likely reduce the restoration's load-bearing capability over time [22]. **When there is occlusal contact damage at the restoration's surface, cracks can be initiated by cyclic loading and propagate downwards and**

outwards driven by water-assisted SCG (hydraulic pumping), resulting in bulk fractures [21]. Therefore, investigating the fatigue behavior of these new high-translucent zirconia is highly demanded, in particular to determine the threshold stress intensity factor below which no slow crack growth is expected. The threshold parameter was also suggested to be a better predictor than the SCG n parameter for the fatigue and clinical performance of dental ceramics [13].

The aim of this work was to study the crack-growth properties (static and cyclic fatigue) of newly developed zirconia ceramics in combination with a thorough investigation of their strength, mechanical reliability (i.e. scatter in strength values), translucency and microstructures. The results are compared to those of lithium-disilicate glass-ceramic, being another type of popularly used dental ceramics for monolithic restorations. The results are then discussed in the context of the search towards the best compromise between crack resistance and translucency.

2. Materials and methods

2.1. Materials

Three high-translucent zirconia ceramics were processed from commercially available powders (Zpex, Zpex4 and ZpexSmile, all from Tosoh, Tokyo, Japan). All these powders contain a low percentage (0.05 wt%) of Al_2O_3 . The yttria stabilizer content in Zpex, Zpex4 and ZpexSmile was 3, 4 and 5 mol%, respectively. The resulting zirconia ceramics were referred to as ‘3Y’, ‘4Y’ and ‘5Y’ throughout the text. Powders were compacted by uniaxial pressing at 40 bar applied on the piston for 40 s (Nannetti SSN/EA, Faenza (RA), Italy) to disks of 18 mm in diameter and rectangular plates with surface areas of $30 \times 80 \text{ mm}^2$, followed by cold isostatically pressing at 300 MPa for 3 min. Pressed green bodies were debinded following the thermal cycle suggested by the powder supplier, pre-sintered at 1000°C for 1h and finally pressure-less sintered in air at 1450°C for 2 h using the

same sintering cycles for all zirconia samples. All materials reached full density of 6.07 ± 0.01 (SD), 6.06 ± 0.01 , and 6.06 ± 0.00 g/cm³ for 3Y, 4Y and 5Y, respectively. Sintered disks were used for translucency, microstructural and phase characterizations. Bars for testing 4-point bending strength were cut (Servocut 301-AA, Metkon, Rotterdam, Netherlands) from sintered plates of approx. $22 \times 60 \times 5$ mm³ to a size of $5 \times 4 \times 45$ mm³ and subsequently machined parallel to their final dimensions of $4 \times 3 \times 45$ mm³. The specimens for slow crack growth tests were cut from sintered zirconia plates of approx. $22 \times 60 \times 3$ mm³ down to $20 \times 40 \times 3$ mm³ and parallel ground to 2 mm in thickness.

Lithium-disilicate glass ceramic (referred to as 'LS₂') samples were machined from IPS e.max CAD blocks (IPS e.max CAD HT A2, Ivoclar Vivadent, Schaan, Liechtenstein; 'HT' = high translucency). Plate-shaped specimens were cut from as-delivered blocks of partially crystallized lithium metasilicate precursory phase using a low-speed diamond saw (Accutom-50, Struers, Ballerup, Denmark), followed by crystallization heat treatment according to the manufacturer's instructions in a Programat P300 (Ivoclar Vivadent) vacuum ceramic oven.

2.2. Material characterization

X-Ray Diffraction (XRD; 3003-TT, Seifert, Ahrensburg, Germany) was used to characterize the phase composition of sintered ceramics. XRD patterns were collected on as-sintered surfaces using Cu-K α radiation at 40 kV and 40 mA from 20-90° (2 θ) with a step size of 0.01 for 3 s. Rietveld analysis of XRD patterns was performed with Topas academic software (Bruker AXS, Karlsruhe, Germany) to quantify phase contents and lattice parameters. Yttria content in the tetragonal zirconia phase was calculated based on the *a* and *c* unit cell parameters and the formula previously reported [10, 23, 24].

The microstructures were analyzed by Scanning Electron Microscopy (SEM; Supra 40, Carl Zeiss AG, Oberkochen, Germany,) of as-polished ceramic surfaces. Additional surface treatments (thermal and chemical etching, and coating of conductive layer) were avoided in order to observe the intrinsic microstructures of all phases present (crystalline as well as glass phase). The grain size distributions on more than 1000 grains were measured on SEM micrographs according to the linear intercept method without applying any correction factor.

Four-point bending strength was measured at a loading rate of 0.5 mm/min with 40 mm outer span and 20 mm inner span. Bending bars (n=20) were sequentially polished with 7, 3, and 1 μm diamond pastes prior to the tests. Fracture surfaces obtained by four-point bending testing were examined by SEM.

Translucency was measured on polished specimens (0.5-mm and 1.0-mm thick; n = 6 per group) using a spectrophotometer (SpectroShadeTMMICRO, MHT OpticResearch, Niederhasli, Switzerland). CIELAB coordinates (L^* , a^* and b^*) were recorded against white and black calibrated background boards with a thin layer of vaseline applied in between. Contrast ratio (CR) was calculated, which is 0.0 for a transparent material and 1.0 for a totally opaque material.

2.3. Slow crack growth

The double-torsion (DT) method was used to measure crack-growth velocity (V) versus applied stress-intensity factor (K_I) curves under static and cyclic conditions at room temperature ($\sim 20 \pm 2^\circ\text{C}$) in ambient air ($\sim 50\%$ RH). A wide range of crack-growth velocities from 10^{-12} to 10^{-3} m/s were followed; the fracture toughness (K_{IC}) associated with fast fracture and threshold (K_{I0}) below which no crack growth was observed, were determined. Details of the DT loading configuration together with the double-torsion specimens (dimensions of $40 \times 20 \times 2$ mm³ for zirconia and $40 \times 18 \times 2$ mm³ for LS₂) can be found in more details elsewhere [25, 26]. The tensile surface of the specimens

was polished to facilitate crack observation. A notch of length ‘a₀’ (≈10 mm) was machined using a 0.3-mm thick diamond sawing blade (ATM GmbH, Mammelzen, Germany). Two 10kg Vickers indentations were made **at the tip of notch** to initiate a small crack **on the tensile side of the sample**. Subsequent pre-cracking was performed by slowly loading the specimens at a rate of 10 μm/min, so to induce a real sharp crack with an initial length ‘a_i’ (≈12-13 mm) and **stable crack front geometry** [25].

The K_I in the double-torsion technique was calculated according to the following expression [18, 26]:

$$K_I = \frac{W_m}{T^2} \left[\frac{3(1+\nu)}{W\psi} \right]^{1/2} P \left(\frac{a}{a_0} \right)^{6/32} \quad (1)$$

where ‘P’ is the load, ‘a’ the total crack length, ‘a₀’ the notch length, ‘W_m’ the span width, ‘T’ the thickness of the sample, ‘W’ the width of the sample, ‘ν’ the Poisson ratio, and ‘ψ’ a calibration factor [27]. **Note that (a/a₀)^{6/32} was the correction factor to conventional expression of K_I, because K_I was demonstrated to slightly depend on the crack length due to the fact that the crack front was not straight and longer on the tensile side** [25].

To obtain the crack-growth curve in full range from threshold to fast fracture, two methods were complementarily used, namely the relaxation and constant test. In the relaxation test that enables measuring the velocity range of 10⁻⁷-10⁻² m/s, pre-cracked specimens were loaded at the rate of 0.3 mm/min to a certain load with the function of load versus time being recorded. The crack velocity (V) for the relaxation test was calculated using the following equation [26, 28]:

$$V = -\frac{P_f}{P^2} \left(a_f + \frac{D}{B} \right) \left(\frac{dP}{dt} \right) \quad (2)$$

with ‘a_f’ the final crack length, ‘P_f’ the final load, ‘dP/dt’ the slope of relaxation at a given time, and ‘D/B’ the constant from the compliance calibration [25].

To obtain lower crack rates till near threshold crack rates (10^{-12} m/s), constant loading tests were used. The specimens were subjected to different loads for a prescribed duration to induce crack extension. The crack length was measured via optical microscopy with a precision of 2 μ m. The crack velocity (V) was calculated as the ratio of the crack increment (Δa) to the duration (Δt):

$$V = \frac{\Delta a}{\Delta t} \quad (3)$$

Crack propagation tests under cyclic conditions were performed under load control using a sine waveform between minimum and maximum load at a 'R' ($R = P_{\min}/P_{\max} = K_{I,\min}/K_{I,\max}$) ratio of 0.1 and a frequency of 10 Hz. The maximum load, i.e. the maximum applied stress intensity factor, ' $K_{I,\max}$ ' (and also ' $K_{I,\min}$ ' to keep $R=0.1$) was lowered until the crack propagation was no longer observed even after loading for 6 days. The crack rate was again determined using Eq. (3) and V- $K_{I,\max}$ diagrams were obtained for cyclic fatigue. The maximum load used in this work for cyclic fatigue tests was in the range of 95-40 N, bringing $P_{\min}$ of < 10 N with a negligible effect on the crack velocity.

Known that the V- K_I diagram of oxide ceramics generally presents three typical stages in air, attributed to three distinct mechanisms; each stage (Stage I, II, III) of the V- K_I diagram can be fitted separately by the form of $V=A (K_I \text{ or } K_{I,\max})^n$, with 'A' and 'n' being constants that correspond to each stage. Since stage I has a primordial practical importance of ruling the device longevity at the low crack rate, the stress corrosion coefficient/fatigue parameter (n) value from stage I was determined as the slope in the plot of $\ln (K_I)$ vs. $\ln (V)$.

In the end, the crack-propagation features were observed by SEM at the crack tip of the surface of DT specimens.

2.4. Statistical analyses

One-way analysis of variance (ANOVA) followed by Tukey's post hoc was used to investigate statistical differences in the mean values between different materials at a significance level of $\alpha = 0.05$ (Minitab®16.2.1, Minitab Inc, Pennsylvania, USA). The bending strength results were also analyzed by Weibull statistics with confidence bounds ($\alpha = 0.05$; 95% confidence interval).

3. Results

3.1. Phase characterization

XRD patterns (Fig.1) showed that all 3Y, 4Y and 5Y zirconia ceramics were composed of mixed tetragonal (*t*-ZrO₂) and cubic (*c*-ZrO₂) phases. As expected from the ZrO₂-Y₂O₃ phase diagram and previous results [29, 30], increased yttria content led to higher cubic content at the expense of tetragonal phase. The XRD peaks corresponding to cubic phases in 4Y and 5Y can be clearly observed in between the doublet peaks of tetragonal phases (Fig. 1b). Quantification of the XRD patterns by Rietveld analysis (Table 1) revealed the formation of about 40 and 60% *c*-ZrO₂ in 4Y and 5Y, respectively, as compared to about 10% *c*-ZrO₂ in 3Y. In addition, the remaining tetragonal phase in 4Y and 5Y contained higher yttria content together with a smaller tetragonality (lower *c/a* ratio), consistent with current literature [29].

3.2. Microstructure

The microstructure of the zirconia ceramics revealed polycrystalline and dense structures with only occasionally very few pores (Fig. 2a). Grain-size distributions were all unimodal (Fig. 2b). The three zirconia ceramics significantly differed in microstructure for grain size (Table 2). On the other side, LS₂ consisted of Li₂Si₂O₅ crystals (confirmed by XRD) embedded in a glass matrix. The length of the crystals was in the range of 2 μ m (Table 2) with an aspect ratio of about 4 in respect of their

diameter. Small pores could be clearly observed in the crystalline and glass phases of the LS₂ specimens.

3.3. Translucency

At the thickness of 0.5- and 1.0-mm, the translucency of 5Y > 4Y > 3Y and increasing yttria content significantly enhanced the translucency (lower CR) of zirconia ceramics (Table 2). Reducing thickness also increased the translucency of the three zirconia ceramics. Nevertheless, at the same thickness, LS₂ HT's translucency remained significantly higher than that of all zirconia ceramics investigated (Fig. 3).

3.4. Strength

The average bending strength of 3Y, 4Y and 5Y was 908, 928 and 524 MPa, respectively (Table 2). Weibull analysis (Fig. 4a) revealed their characteristic strength (σ_0) [95% C.I.] was 951 [901-1002], 998 [925-1070] and 581 [532-631] MPa, respectively. There was no statistical difference in bending strength between 3Y and 4Y (Table 2). However, a lower Weibull modulus was obtained for 4Y samples. 5Y had an even lower strength and Weibull modulus than 3Y and 4Y. Fractography of the bending bars (Fig. 4b) were examined in the area close to the fracture origin within the mirror region, since the crack initiated and rapidly accelerated in this region. 3Y and 4Y exhibited similar fracture patterns consisting of mixed intergranular and transgranular fractures with fine zirconia-grain sizes, whereas the fracture surface of 5Y was different with almost only transgranular fractures through much larger zirconia grains.

Regarding the strength of LS₂, the size of available LS₂ blocks (max. 40 mm in length) did not allow performing the 4-point bending test in this work using 40 mm outer span and also the corresponding

statistical analysis. The flexural strength of LS₂ in Table 2 was thus referred to the value reported by Wendler et al. [31] using the same methodology and the same type of LS₂ specimens but smaller dimension. Considering also the previous report showing that the biaxial strength of 5Y and LS₂ were not statistically different [32], the 4-point bending strength of LS₂ and 5Y shown here in Table 2 should be similar or marginally different.

3.5. Crack-growth resistance

Under static loading (Fig. 5a), V-K_I diagrams were recorded from fast fracture (K_{IC}) down to threshold (K_{I0}) for all ceramics investigated, including for the reference LS₂ glass ceramic. The toughness (K_{IC}) was estimated at a high crack velocity of 10⁻³ m/s and the threshold (K_{I0}) was determined from the points below which there was an abrupt drop to a crack velocity of about 10⁻¹² m/s (see Table 3). Increasing yttria content clearly shifted the V-K_I curve towards lower K_I values. However, the V-K_I curves remained parallel in a log-log scale. Similar K_{I0}/K_{IC} values (~0.5) and a stress corrosion coefficient (n) of ~30 were thus observed for all zirconia ceramics, which revealed a similar relative susceptibility to SCG. When compared to LS₂, 5Y and LS₂ showed a similar fracture toughness, but the K_{I0} of LS₂ (1.1 MPa·m^{1/2}) was lower than that of 5Y. Although the difference was only 0.4 MPa·m^{1/2}, it represented about 30% of relative difference. This finding indicated that LS₂ was more sensitive to SCG than zirconia ceramics. The n value of the V-K_I curve corresponding to LS₂ was indeed lower (~16) and a smaller K_{I0}/K_{IC} value (~0.35) was recorded.

With respect to the effect of cyclic loading, the cyclic V-K_I_{max} curves of zirconia ceramics are shown in Fig. 5b and the comparison to reference LS₂ is shown in Fig. 5c. For the three zirconia ceramics, the crack-growth rates under cyclic loading were clearly accelerated compared to static crack growth at equivalent stress intensities. One should also note that cyclic crack-growth rate was presented as

a function of $K_{I\max}$ and that the determination of the exact cyclic degradation from the direct comparison of static $V-K_I$ and cyclic $V-K_{I,\max}$ would not be correct. The crack velocity per cycle ($da/dN-K_{I\max}$) if there was no cyclic fatigue degradation can be estimated, as previously reported [18], by an integration of the $V-K_I(da/dt-K_I)$ curve over a cycle. Crack-growth rate per cycle ($da/dN-K_{I\max}$) would have been lower than for static $da/dt-K_I$, as indicated by the dash line for 3Y in Fig. 5b. Indeed, since the average integrated load (varying between $P_{\min}$ and $P_{\max}$) is lower under cyclic loading than $P_{\max}$, the crack-growth rate would be lower than at a constant maximum load, if no cyclic degradation was present. The real effect of cyclic fatigue degradation is therefore appreciated from the difference between the calculated $V-K_{I\max}$ curve (dashed lines in fig. 5b for 3Y-TZP) and the experimental one. Therefore, cyclic loading significantly degraded the crack resistance of the three zirconia ceramics. Regarding the reference LS_2 glass ceramic (Fig. 5c), similar to the zirconia ceramics, cyclic degradation was also observed for LS_2 : cyclic crack growth occurred at stress intensities below that for static crack growth; K_{I0} was shifted to a lower value and the $V-K_{I,\max}$ curve had a lower slope (Table 3).

Fig. 6 shows SEM images in the vicinity of the crack tips at the surfaces of the DT samples. Unlike metals [14] and because of no (or very localized and limited) plasticity, the crack-propagation modes and microstructures of cyclically and statically loaded specimens did not show differences, so only images obtained from statically loaded specimens were representatively shown. Along the crack path in 3Y, grains at a depth of 1-2 grains were transformed and a twining morphology of monoclinic grains were clearly observed to have arrested the crack opening (as illustrated in the schematic). 4Y revealed a similar behavior as 3Y, but less grains were transformed along the crack path. 5Y, on the other hand, exhibited no transformed grains. Crack bridging at the level of zirconia grains can be found in all zirconia ceramics with however relative straight crack propagation, if we compare with polycrystalline ceramics with larger or more elongated grains [33-35]. Crack deflection and bridging

were more pronounced for LS₂ with clearly meandered crack paths (note also the different scale bar in LS₂).

4. Discussion

4.1. Toughening and mechanical properties that are measured at fast fractures

The toughening mechanisms of the materials in general consist of intrinsic (K_0) and extrinsic toughening (ΔK_c), and the extrinsic toughening mechanism working in the wake of the crack (i.e. crack-shielding) is known to be the primary toughening sources for brittle ceramic materials [36], by which transformation toughening, crack bridging, crack deflection and/or combinations thereof are relevant toughening mechanism in the four materials investigated [36, 37]. Reduction of phase-transformation toughening from 3Y to 4Y and 5Y (Fig. 6) obviously resulted in the shifts of the V- K_I curve to lower K_I values with lower K_{IC} and K_{I0} (Table 3). In 4Y and 5Y, not only more non-transformable cubic phase was formed, the remaining tetragonal phase was also less transformable with lower tetragonality (Table 1). 4Y thus had less toughening effect from phase transformation than 3Y, whereas 5Y did not show any evidence of phase transformation toughening. Although grain bridging that generally exists in polycrystalline ceramics was present in all zirconia ceramics including 5Y (Fig. 6), this mechanism from fine (0.3-0.5 μm , Table 2 and Fig. 6) and equiaxed grains should merely be effective on small dimensions in a limited extent, unlike in the case of coarse-grained Al_2O_3 or grain-elongated Si_3N_4 [14, 33-35, 38, 39]. Due to the relative large and elongated $\text{Li}_2\text{Si}_2\text{O}_5$ crystals (Fig. 2a) [40, 41], crack deflection and crack bridging was effective in LS₂ (Fig. 6).

On the other side, different from the general expectation, 4Y, although being less tough than 3Y, did not show a lower bending strength (Table 2). The high strength of 4Y should be related to the

small processing defects and the fine microstructure (Fig. 2). Based on the strength distribution (Table 2) and toughness values (Table 3), the size of critical flaws in 4Y was calculated to be in the range of 13-9 μm , using linear-elastic fracture mechanics and the Griffith equation ($Y=1.3$) [42]. The critical flaw in 3Y and 5Y was in the range of 20-17 μm and 26-17 μm , respectively. The grain sizes/distributions (Fig. 2) and fracture patterns (Fig. 4b) of 4Y and 3Y were also very close. Another work reported a biaxial strength of higher than 1100 MPa for 4Y, similar to that of 3Y, when using the same sintering cycles [43]. In the work reporting a lower strength of 4Y [32], the grain size of 4Y however was much larger than that of 3Y. Nevertheless, despite the similar strength of 4Y and 3Y, the strength scatter of 4Y ($m=6$) was higher than for 3Y ($m=10$), followed by an even higher one for 5Y ($m=5$) (Fig. 4). Decreased mechanical reliability when increasing yttria content should be attributed to the reduced crack-growth resistance and thus higher sensitivity to cracks/defects. At the same time, lower crack resistance raises another concern in terms of surface treatment. Clinically conducted surface treatments, such as sandblasting, are able to enhance the strength of 3Y-TZP thanks to the generated compressive stresses [44], but this effect might be compromised/lost [45] in PSZ (4Y and 5Y).

When comparing to LS_2 glass ceramic, 4Y and 3Y showed better mechanical properties, but 5Y revealed only a marginally higher fracture toughness (Table 3) and flexural strength (Table 2) [32]. Importantly, a lower strength scatter, i.e. higher mechanical reliability ($m \sim 10$ [31, 46-48]) was recorded for LS_2 than 5Y. The finding that LS_2 and 5Y have a similar fracture toughness (K_{IC}) but different reliability, should probably be attributed to the different proportions of intrinsic toughness (K_0) and extrinsic toughening (ΔK_c). As explained at the beginning of the discussion, the K_{IC} of brittle materials and more specifically the crack growth resistance (K_R) commonly consists of K_0 and ΔK_c , and ΔK_c can lead to a stably rising crack resistance, i.e. R-curve behavior [36, 37]. K_0 of

LS₂ is reported to be about 1.2 MPa·m^{1/2} [40], whereas K₀ of zirconia is around 2 MPa·m^{1/2} [36, 49]. It can then be figured that ΔK_c for LS₂ is higher than for 5Y, resulting in a stronger R-curve for LS₂ with better tolerance to cracks/defects. Indeed, the most potent extrinsic toughening mechanism (i.e. phase transformation) for zirconia ceramics was absent in 5Y zirconia, while LS₂ had obviously effective extrinsic toughening effect from crack deflection and crack bridging (Fig. 6), contributing to the higher mechanical reliability of LS₂. Another similar lithium-disilicate glass-ceramic with elongated LS₂ crystals (Empress 2, Ivoclar Vivadent AG) even showed higher Weibull modulus than a 3Y-TZP ceramic [17]. The microstructure of LS₂ investigated here (Fig. 2a) also showed more defects (porosities and heterogeneous structures) than that of 5Y, despite they revealed a similar or marginally different strength.

4.2. Long-term durability: SCG and cyclic fatigue

Independent on K_{IC} and the yttria content, 3Y, 4Y and 5Y showed a similar relative susceptibility to SCG under static loading (parallel shift of V-K_I curves in a log-log scale; K_{I0,static}/K_{IC} ~0.5; similar stage-I n value of ~30), yet having a significant higher SCG resistance than LS₂ (n of ~16, K_{I0,static}/K_{IC} ~0.4) (see Fig. 5a and Table 3). It is necessary to first clarify that the V-K_I curves obtained in this work by DT should not be influenced by the R-curve behavior. Although the extrinsic toughening effects and the resulting R-curve behavior were different for the four materials investigated, the steady state conditions of the R-curve should be reached in the crack-size domain of the DT tests. The transformation zone (h) in the studied 3Y was found to be about 1 μm (1-2 grains in Fig. 6), revealing that 3Y should reach the R-curve steady status at a crack length of about 5 μm (about 5 times the transformation zone [50]). Although the R-curve of LS₂ is estimated to be stronger than that of 5Y, a steep raise over the first micrometers or tens of micrometers was also

reported because of the fine dimensions of $\text{Li}_2\text{Si}_2\text{O}_5$ crystals [13, 41]. In addition, the rising part of R-curve would result in a higher slope of the V- K_I curve [33], which is the opposite as observed for LS_2 . Therefore, the differences in observed SCG susceptibility should not be attributed to the R-curve but to the material's intrinsic susceptibility to water-assisted stress-corrosion reactions, namely chemically assisted SCG. For the three zirconia ceramics, they are free of glass phase with the same Zr-O-Zr bonds, presenting the same stress corrosion at the crack tip, which has also been observed before for 3Y-TZP with different grain sizes and for zirconia stabilized by different stabilizers [51]. LS_2 consists of 70 vol% $\text{Li}_2\text{Si}_2\text{O}_5$ crystals and 30 vol% silicate glass phase. It is difficult to quantitatively compare the water affinity of Zr-O and Li-O/Si-O bonds, but (silicate) glass materials are known to be more susceptible to hydrolysis and stress-corrosion than polycrystalline materials [12, 13, 52]. The different SCG susceptibility recorded between zirconia and LS_2 (Fig. 5c) is reflected in stage I, confirming the effect of the stress-corrosion reaction.

Regarding the cyclic SCG, all studied ceramics showed cyclic degradation with decreased $K_{I0,\text{cyclic}}$ and lower slope of V- $K_{I,\text{max}}$. Cyclic fatigue accelerates the crack growth in brittle ceramics by degrading the extent of extrinsic crack-tip shielding toughening [14] that again involves transformation toughening, bridging, deflection and/or combinations thereof (Fig. 6) [14, 36]. Since the extent of extrinsic toughening was not the same for the four studied materials, materials undergoing higher extrinsic toughening (i.e. 3Y) were expected to be more vulnerable to cyclic degradation. However, surprisingly, the extent of cyclic degradation (~5% loss in K_{I0}) was rather similar between the different samples. This similar susceptibility to cyclic fatigue in the three zirconia ceramics points out that the transformation toughening (in 3Y and 4Y) is not degraded by cyclic fatigue, since transformation toughening acts as inelastic zones surrounding the crack tip but not behind the crack tip like bridging [38]. The observed cyclic degradation can be attributed to the grain bridging mechanism, i.e. elastic or frictional bridging of contact shielding that can be degraded

by loading/unloading [14, 38, 39, 53]. Even though, as discussed above that grain bridging for the studied zirconia ceramics is only effective in a limited extent and thus cyclic effects solely contribute to a very small proportion of overall degradation as compared to chemically-assisted SCG. From the aspect of threshold, chemically-assisted SCG presented about 50% reduction of the fracture toughness for zirconia ceramics ($K_{I0,static}/K_{IC} \sim 0.5$), but the cyclic loading gave rise to $\leq 5\%$ degradation of K_I ($K_{I0,cyclic}$ was $\leq 5\%$ smaller than for $K_{I0,static}$). The LS_2 glass-ceramic, having elongated crystals in the order of $2\ \mu m$, suffers more from the cyclic fatigue effect due to the higher dependence of its bridging mechanism on the friction effect at also a larger scale (Fig. 6). Furthermore, the volume fraction of these elongated crystals together with their size, aspect ratio, orientation are critical for the fracture resistance of the LS_2 glass-ceramic [41, 54], raising the question about the influences of these factors on the SCG and cyclic fatigue of LS_2 glass-ceramic for further investigations as well.

Above all, the three zirconia ceramics showed a similar SCG relative susceptibility under both static and cyclic loadings, but were more resistant to SCG than LS_2 due to the higher susceptibility of the glass phase to the water-assisted stress-corrosion reaction. Previous studies using indirect methods (dynamic and cyclic fatigue tests) also showed that LS_2 glass ceramic would degrade faster over time with smaller n values than zirconia [13, 55] as well as in aqueous environment [11]. In fact, glass-containing ceramics, including glass-infiltrated ceramics, generally exhibited a higher susceptibility to SCG than polycrystalline ceramics (zirconia, alumina) [13, 55-57]. One exception was reported for a leucite-reinforced glass ceramic that contained even 50-60% aluminosilicate glass (IPS Empress, Ivoclar Vivadent) [13]. Different from LS_2 glass ceramic, the enhanced SCG resistance in IPS Empress was attributed to the compressive residual stress developed around the leucite crystals [13, 40].

It is necessary to point out that the current work was performed in ambient air conditions (~50% RH) but not in water or body fluid, because water molecules in air were enough for SCG to proceed and only one stage (stage I) would be present in water with the same stress-corrosion coefficient (i.e. the same SCG **susceptibility**) and with the same threshold stress intensity factor K_{I0} (only different crack growth rate) [20, 58]. Nevertheless, the oral cavity is a complex environment with a fluctuating temperature, high acidic environment (low pH), bacteria and so on. This environment can especially be important for LS₂, because LS₂ shows a relatively high dissolution in an acidic environment (**Fig. S3** in the supplementary document) and the V-K_I diagram in an environment where the materials undergo some dissolution, could be substantially different [59]. Although zirconia ceramics (3Y, 4Y and 5Y) are quite resistant to acid etching (**Fig. S3** in supplementary documents), considering the hydrothermal aging behavior, it is also paramount to study the crack-growth behavior of zirconia ceramics in a more hostile environment more closely resembling the oral cavity, or after a prolonged duration in such media.

4.3. Material selection based on strength and translucency

Considering the numerous zirconia ceramics introduced in the dental field and the fact that the mechanical and optical properties of different zirconia ceramics are highly variable, as discussed above, it is interesting to plot a flexural strength versus translucency map (the main parameters commonly employed as guidance to select the proper material to design and fabricate FDPs). **Fig. 7** displays a scatter plot of flexural strength versus translucency for various materials that can be processed by CAD-CAM. More detail of the material classification and consulted data sources can be found in the supplementary Fig. S1 and Table S1. **Admittedly, the strength is influenced by process conditions (pressing, pre- and sintering) with different flaw sizes and distributions, surface treatments (ground, polished or as-sintered) [60], specimen dimensions and methodologies of**

bending tests with different effective stressed volume [61]. Size limitations of many commercial CAD/CAM materials also lead to the fact that no single sample dimension and test method are dominantly used. The influence of surface treatments especially for zirconia ceramics is also a topic of ongoing investigation [60]. Therefore, in addition to the property of materials themselves, all above mentioned experimental and sampling uncertainties could contribute to increase the scatter of strength data.

Zirconia, both TZP and PSZ, remain in general less translucent than lithium-(di)silicate glass ceramic. TZP here refers to 3Y-TZP in order to be consistent with the commonly used terminology and PSZ refers to zirconia containing yttria contents higher than 3Y. In fact, it should be kept in mind, and this is again shown in the experimental results provided that all yttria-doped zirconia polycrystalline ceramics, with 3-6 mol% yttria are in fact a mixture of *t*- and *c*-ZrO₂ (Table 1) [29], thus are all PSZ materials. The evolution of the different zirconia generations from various ‘TZPs’ to so-called ‘PSZ’ (sometimes even called not correctly ‘cubic zirconia’) clearly shows the significant improvement in zirconia’s translucency (Fig. S2).

In spite of the reflection at the surfaces, the main factor limiting the light transmission of polycrystalline ceramics is scattering by birefringence at the grain boundary and by pores or second phases [8]. The first generation of dental 3Y-TZP in general contains 0.25 wt% alumina, allowing some light transmission but as previously introduced, the formation of alumina particles limits their translucency [10]. Then, lower alumina additions (up to as low as 0.05 wt%) to zirconia have been suggested. The alumina addition of less than 0.2 wt% is within the solubility limit of alumina in zirconia and can thus be fully dissolved and segregated as Al³⁺ at the zirconia grain boundaries [62, 63]. In this way, the zirconia ceramics can still be easily fully-dense sintered with the aid of Al³⁺ [64] while the formation of secondary alumina particles is avoided (Fig. 2). Furthermore, as shown in Fig. S2b and by previous works [10], decreasing alumina does not significantly compromise

strength, but slightly lowers hydrothermal aging resistance [9, 10]. More recently, zirconia is being stabilized with higher yttria contents to obtain even higher translucency (Fig. 3) by decreasing the grain boundary birefringence (higher content of optically isotropic cubic phase and lower birefringence of the remaining tetragonal phase with smaller c/a tetragonality, Table 1) [9, 10, 65]. Some of the new so-called ‘high-translucent’ PSZ already present light transmission similar to that of LS₂ with a low translucency (‘LT’) or medium opacity (‘MO’). A part of the window in the map is indeed also filled by these new zirconia ceramics (Fig. 7, Fig.S2 and Fig. S1). Therefore, with the development of high-translucent zirconia, more indications in fixed prosthodontics can be covered by monolithic restorations.

Conclusions

Despite the common understanding that increasing yttria content decreases toughening in yttria-stabilized zirconia ceramics and thus reduces K_{IC} and K_{I0} , 5Y and 4Y PSZs with a higher translucency are promising materials to be used for monolithic restorations. In view of strength versus translucency map, these new translucent zirconia ceramics can fill a part of the material’s window between conventional 3Y-TZP and LS₂ glass ceramic, even if the lower Weibull moduli measured require attention in terms of reliability issues.

All investigated zirconia ceramics showed a similar relative susceptibility to static and cyclic SCG with a parallel shift of V- K_I curves. Cyclic fatigue degradation was significant, but modest for the four ceramics investigated and was not influenced by the transformation toughening effect. LS₂ glass-ceramic was less SCG-resistant than zirconia ceramics, due to its susceptibility to chemically-assisted stress corrosion. However, LS₂ with pronounced extrinsic toughening mechanism from crack bridging exhibited a higher Weibull modulus than 5Y, despite they showed a similar fracture toughness.

Acknowledgements

A large portion of the work was performed in the framework of the SISCERA project funded by the European Union's Horizon 2020 Research and Innovation Framework Programme - Fast Track to Innovation Pilot (FTIPilot2016-1) - under Grant Agreement No. 737954. F. Zhang thanks the support of the Research Foundation Flanders (FWO Vlaanderen), who awarded her a post-doctoral fellowship (12S8418N) during the time the manuscript was written. We thank Ivoclar Vivadent for providing the IPS e.max CAD samples (HT, A2/B40 L), PhD student Evita Willems and Prof. Jef Vleugels (both from KU Leuven) for preparing some zirconia samples, and Prof. Marleen Peumans (KU Leuven) for the translucency measurements.

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(a)

(b)

Contrast ratio/CR

0.0 0.2 0.4 0.6

Tables:

Table 1. Phase composition and lattice parameters using XRD and Rietveld analysis.

		3Y	4Y	5Y	
XRD		fraction (%)	87 ± 2	63 ± 4	42 ± 4
	Tetragonal	<i>c/a</i>	1.0151 ± 0.0002	1.0147 ± 0.0000	1.0126 ± 0.0007
		Y ₂ O ₃ (mol%)	2.7 ± 0.06	2.9 ± 0.01	3.7 ± 0.30
	Cubic	fraction (%)	13 ± 2	37 ± 4	58 ± 4
		<i>c</i> (Å)	5.1379 ± 0.0005	5.1360 ± 0.0007	5.1348 ± 0.0001

Table 2. Grain size, translucency and strength of all ceramics investigated. Mean values are reported with (±) 95% confidence interval.

		Translucency (CR)		4-point bending strength	
Grain size (μm)		1.0 mm	0.5 mm	Average (MPa)	Weibull modulus
3Y	0.30 ± 0.01 ^a	0.65 ± 0.02 ^a	0.54 ± 0.02 ^c	908 ± 44 ^a	9.2
4Y	0.36 ± 0.02 ^b	0.59 ± 0.01 ^b	0.47 ± 0.01 ^d	928 ± 82 ^a	6.3
5Y	0.53 ± 0.03 ^c	0.45 ± 0.00 ^d	0.36 ± 0.01 ^e	534 ± 56 ^b	4.9
LS ₂	1.72 ± 0.12 (L) 4.06 ± 0.36 (aspect ratio)	-	0.28 ± 0.01 ^{f*}	462 ± 15 [#]	10.0 [#]

Different superscripts (for each variable) indicate statistically different results (significance level $\alpha=0.05$); *LS₂ is HT, high translucency grade; [#] Value reported by Wendler *et al.* (2017) using the same 4-point bending test [31].

Table 3. SCG parameter values.

		SCG Static			SCG cyclic (10 Hz, R=0.1)		
K _{IC} (MPa·m ^{1/2})		K _{I0} (MPa·m ^{1/2})	K _{I0} /K _{IC}	n, stage I	K _{I0} (MPa·m ^{1/2})	K _{I0} /K _{IC}	n, stage I
3Y	5.0 ± 0.3	2.7 ± 0.1	0.54	30.8 ± 7.2	2.5	0.49	20.9
4Y	4.0 ± 0.2	2.2 ± 0.1	0.55	27.7 ± 5.5	2.1	0.52	17.9
5Y	3.3 ± 0.2	1.5 ± 0.1	0.46	27.2 ± 3.2	1.4	0.43	23.7
LS ₂	3.1 ± 0.2	1.1 ± 0.1	0.35	15.8 ± 5.5	1.0	0.32	10.5