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A novel high-strength lithium disilicate glass-ceramic featuring a highly intertwined microstructure

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ABSTRACT

To achieve long-term clinical performance and wider application of glass-ceramic dental restorations, it is urged to enhance the mechanical properties of glass-ceramic materials. In this study, a high-strength lithium disilicate glass-ceramic was developed in a SiO_2–Li_2O–Al_2O_3–MgO–P_2O_5–ZrO_2 related glass system, which demonstrates a high flexural strength of 562 ± 107 MPa. In this high-strength glass-ceramic, the microstructure features highly intertwined colonies of lithium disilicate. This novel microstructure effectively contributes to the improvement of flexural strength. The minor crystalline phases (β-quartz, $MgAl_2Si_4O_{12}$, and Li_3PO_4) embedded within the $Li_2Si_2O_5$ (LS_2) crystal colonies and residual glass matrix could further strengthen the glass-ceramic. The development process of such a novel microstructure and its possible formation mechanism are proposed. This material could be an excellent candidate for restorative dental applications up to three-unit posterior bridges.

1. Introduction

Glass-ceramics have been envisioned for dental use since 1968 [1]. The ceramic materials for dental restorative applications need to fulfil the requirements on biocompatibility, physical and chemical properties as per the standards ISO 6872 [2]. There are several types of glass-ceramics for restorative dentistry, including mica-based glass-ceramics (e.g., DICOR®), leucite-based glass-ceramics (e.g., IPS Empress®), and lithium disilicate-based glass-ceramics (e.g., IPS Empress® 2) [3,4]. As a promising dental restorative material, lithium disilicate (LS_2) glass-ceramics have been well known for their good mechanical properties and excellent translucency [5]. The commercial dental products of this type, such as IPS Empress®2, have a typical flexural strength of 400 ± 40 MPa and toughness of 3.3 ± 0.3 MPa $m^{1/2}$ [5,6]. Nevertheless, the current products are not yet made suitable for applications of multi-unit restorations involving a molar tooth [7]. For the application of posterior three-unit dental bridges, the flexural strength and fracture toughness of dental materials should be sufficiently high, i.e. no less than 500 MPa and 3.5 MPa $m^{1/2}$, respectively [2], to withstand fracture under any masticatory stress induced on the restoration [8].

In order to enhance the reliability and achieve longer service life of glass-ceramic dental products, many efforts have been made by improving the mechanical properties of lithium disilicate glass-ceramics [9,10] Extensive research has been done on high-strength LS_2 glass ceramics for dental applications. In the last decade, Apel et al. reported a lithium disilicate glass-ceramic with high biaxial flexural strength (>700 MPa) in a SiO_2–Li_2O–Al_2O_3–K_2O–P_2O_5–ZrO_2 glass system. [10]. However, they did not report flexural strength data by a conventional three-point bending method. Instead, glass-ceramics derived from a similar glass composition demonstrated a three-point flexural strength of only 307–394 MPa [11]. Microstructural tailoring is a very important method for strengthening and/or toughening ceramic materials. A plausible approach is introducing zirconia, which is best known for its transformation toughening effect [12,13] into the glass composition to reinforce lithium disilicate glass-ceramics [10,14]. However, such a toughening effect might not be realised if the effective tetragonal zirconia polycrystals (TZP) do not form in the glass-ceramics [9,11,14]. Alternatively, one can engineer a highly-crystalline interlocking microstructure through a novel compositional design of base glass and an optimized thermal annealing process [6,9,15–17]. While, the microstructure of elongated crystals have been widely deemed as an "interlocking" feature, while the LS_2 crystals are not necessarily chemically interlocked, but physically/mechanically only.

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Table 1
Patents related to lithium disilicate glass-ceramics for dental applications.

Group	Patent Title	Patent No.	Year Issued	Inventors	Company	Info
A	Lithium disilicate- containing glass-ceramics some of which are self-glazing	US5219799	Jun 1993	Beall, G. H. Echeverria, L.M. Morrissey, J. Pierson, J.	Corning Inc. @NY, USA	Certain compositions within covered ranges are self-glazing.
B	Process to prepare moulded shaped translucent lithium-disilicate glass ceramic products	EP916625 US6420288 JP11314938	May 1999 Jul 2002 Nov 1998	Schweiger, M. von Clausbruch, S. C. Höland, W. Rheinberger, V.	Ivoclar Vivadent AG @Schaan, Liechtenstein	A process for shaped translucent lithium disilicate glass ceramic products which may be processed by pressing in the plastic state or milling to finished glass ceramic products as dental restorations.
C	Lithium disilicate glass ceramics dental product/Sinterable lithium disilicate glass ceramic	US6342458/ US6514893 US6606884 US5968856	Jan 2002 Feb 2003 Aug 2003 Oct 1999	Schweiger, M. Frank, M. Rheinberger, V. Höland, W.	above	High-strength sinterable lithium disilicate glass ceramics are described which can be further processed in particular by pressing in the viscous state to shaped dental products.
D	Lithium silicate materials	EP2284133 EP2305614 EP2269960 EP1505041 US8047021 US8042358 US7316740 JP2010168278	Feb 2011 Apr 2011 Jan 2011 Feb 2005 Nov 2011 Oct 2011 Jan 2008 Aug 2010	Schweiger, M. Rheinberger, V. Bürke, H. Höland, W.	above	Lithium silicate materials can be easily processed by machining to dental products without undue wear of the tools and which subsequently can be converted into lithium silicate products showing high strength. (compositions are ZnO-containing)
E	Lithium silicate glass ceramic	US8940651 US8162664 US7993137 US7871948 US7867933 US7867931 US7867930 US7452836 EP2261184 EP1688397	Jan 2015 Apr 2012 Sep 2011 Jan 2011 Jan 2011 Jan 2011 Jan 2011 Nov 2008 Dec 2010 Aug 2006	Apel, E. Höland, W. Schweiger, M. Ritzberger C. Burke, H. Rheinberger V.	above	Same as the "D" category above, but compositions are ZnO-free.
F	Lithium silicate glass ceramic and glass with ZrO2 content	US9249048 US8778075 US8759237 US8557150 US8536078 DE202011110342 EP2377830 EP2377831 EP2913314 EP2407439 EP2662343 EP2664594 EP2662342	Feb 2016 Jul 2014 Jun 2014 Oct 2013 Sep 2013 Jul 2013 Apr 2016 Apr 2016 Sep 2015 May 2015 Aug 2015 Aug 2015 Nov 2013	Ritzberger, C. Höland, W. Schweiger, M. Rheinberger, V.	above	The materials can advantageously be applied to zirconium oxide ceramics in particular by pressing-on in the viscous state and form a solid bond with these.
G	Pressable lithium disilicate glass ceramics	EP1149058 WO00034196 US6455451	Oct 2001 Jun 2000 Sep 2002	Brodkin, D. Panzera, C. Panzera, P.	Jeneric/Pentron, Inc. @CT, USA	Glass-ceramic dental restorations made by heat pressing into moulds produced using lost wax techniques.
H	Lithium disilicate glass-ceramics	US6802894 US6517623	Oct 2004 Feb 2003	above	above	above
I	High-strength dental restorations	EP1797015 WO06042046	Jun 2007 Apr 2006	Brodkin, D. Panzera, C. et al.	Pentron Laboratory Technologies, LLC @CT,USA	The glass-ceramics have good pressability.
J	Castable glass-ceramic composition useful as dental restorative	US4515634	May 1985	Wu, J. M. Cannon, W. R. Panzera, C.	Johnson & Johnson Dental Products Company @NJ, USA	Expired
K	Lithium silicate glass ceramic for fabrication of dental appliances	US20090258778	Oct 2009	Castillo, R.	James R., Glidewell Dental Ceramics, Inc. @CA, USA	With 8%–10% of GeO_2 in the final composition. The material has improved castability.
L	Lithium disilicate glass ceramics, method for the production thereof and use thereof	US8956987	Feb 2015	Durschang, B. Probst, J. Thiel, N. Bibus, J. Vollmann, M. Schusser, U.	Fraunhofer Ges Forschung, etc. @Munich, Germany	Lithium disilicate glass ceramics can be easily mechanically machined in an intermediate stage of the crystallization and represent high-strength, highly translucent and chemically stable glass ceramics after complete crystallization.

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