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Nano-mechanical behaviour of lithium metasilicate glass–ceramic

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ABSTRACT

This paper reports the first study on the mechanical behavior of lithium metasilicate glass–ceramic using nanoindentation and in situ scanning probe imaging techniques. Indentation contact hardness, H_c, and Young's modulus, E, were measured at 10 mN peak load and 0.1–2 mN/s loading rates to understand the loading rate effect on its properties. Indentation imprints were analysed with the in situ scanning probe imaging to understand indentation mechanisms. The average contact hardness increased by 112% with the loading rate (ANOVA, $p<0.05$) while the Young's modulus showed the loading rate independence (ANOVA, $p>0.05$). A strain rate sensitivity model was applied to determine the intrinsic contact hardness. Extensive discontinuities and largest maximum, contact and final depths were also observed at the lowest loading rate. These phenomena corresponded to inhomogeneous shear-band flow and densification leading to the material strain softening. The in situ scanning probe images of indentation imprints showed plastic deformation at all loading rates and shear band-induced pileups at the lowest loading rate. With the increase in loading rate, the induced pile-ups decreased. The continuum model predicted the largest densified shear zone at the lowest loading rate. Finally, these results provide scientific insights into the abrasive machining responses of lithium metasilicate glass–ceramic during dental CAD/CAM processes using sharp diamond abrasives.

1. Introduction

Lithium metasilicate (Li_2SiO_3) glass–ceramic (LMGC) is a metastable material used in dental computer-aided-design and computer-aided-manufacturing (CAD/CAM) processes for all-ceramic lithium disilicate ($Li_2Si_2O_5$) glass ceramic (LDGC) restorations (Guess et al., 2010; Yin and Stoll, 2014). LMGC is obtained by a controlled nucleation and crystallization of the base glass (SiO_2–ZnO–K_2O–Li_2O) through heat treatments with little or no porosity (Beall, 1992; Cramer von Clausbruch et al., 2000). It is milled directly to generate complex inlay/onlay, single crown and three-unit bridge profiles using chair-side or laboratory dental CAD/CAM systems equipped with diamond abrasive tools (Guess et al., 2010; Silva et al., 2011). After milling, LMGC prostheses subsequently undergo heat treatments which lead to the formation of the stable, high strength and highly translucent lithium disilicate reinforced glass ceramic composite (LDGC) (Goharian et al., 2010; Höland et al., 2006; Iqbal et al., 1998; Kracek, 1930; Soares et al., 2003; Yuan et al., 2013; Zheng et al., 2008). The interlocked lithium disilicate microstructure and layered crystals in LDGC contribute to strengthening the ceramic composite (Apel et al., 2008; Denry and Holloway, 2010). The mismatch in thermal expansion coefficients and elastic moduli between lithium disilicate crystals and glassy matrix

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forms tangential compressive stresses, which deflect advancing cracks (Apel et al., 2008; Denry and Holloway, 2010; Serbena and Zanotto, 2012). All these result in 360–400 MPa strength of LDGC, in comparison with other glass ceramics of <100 MPa strength (Denry and Holloway, 2010). In addition, LDGC has excellent chemical durability (Anusavice and Zhang, 1997; ElBatal et al., 2009) and aesthetic appearance matching natural teeth (Guess et al., 2010). Among all glass–ceramics currently available as aesthetic and restorative materials, LDGC is the best, possessing excellent properties enabling fabrications of crowns and multiple unit bridges in large stress-bearing areas (Höland et al., 2000).

The high strength of LDGC and the brittleness of its glassy phase make it difficult to machine using the existing chair-side or laboratory dental CAD/CAM technology (Denry and Holloway, 2010). Instead, diamond abrasive machining of LMGC of approximately 100 MPa strength, the intermediate ceramic composite containing mostly lithium metasilicate crystals (Denry and Holloway, 2010), becomes practical. However, abrasive machining of LMGC induces surface and subsurface damages due to numerous simultaneous indentations and scratches made by hard diamond abrasives (Rekow and Thompson, 2005; Rekow et al., 2011; Xu et al., 1996; Yin, 2012). These machining-induced damages in LMGC may not be healed in the subsequent thermal treatment, leading to the strength reduction in the lifetime of the LDGC restorations (Denry, 2013; Rekow et al., 2011). Therefore, the understanding of the mechanical behaviour of LMGC is critical in determining its machinability and machining mechanisms, which eventually affect the machining quality in CAD/CAM abrasive machining. The material behavior in indentation using diamond indenters mimics machining responses of the material in diamond abrasive machining with respect to the material deformation and fracture (Lawn and Cook, 2012). Thus, the indentation approach lays a foundation mechanics for abrasive machining (Komanduri et al., 1997; Malkin and Hwang, 1996). Particularly, machining forces, cutting speeds and abrasive geometries can be simulated by indentation loads, loading rates and indenter geometries, respectively (Yan et al., 2006).

However, little is known about the mechanical properties of LMGC although efforts have been made towards the mechanical behaviour of LDGC. For instance, micro-indentation of hot-pressed LDGC was conducted, in which fractures were observed (Albakry et al., 2003; Apel et al., 2008; Guazzato et al., 2004). This indicates that the critical contact loads to initiate fracture were exceeded. Nanoindentation techniques were also used to characterize the mechanical behavior of LDGC (Buchner et al., 2011; Soares and Lepienski, 2004). In those studies, fractures also occurred at several hundreds of milli-Newtons. Plastic deformation in LDGC was observed in nanoindentation when a milli-Newton (10 mN) peak load was used (Smith et al., 2014). This confirms that the fracture-dominated mode can be changed to the plastic deformation mode by progressively diminishing contact loads (Lawn and Evans, 1977).

In abrasive machining, the plastically deformed material removal can produce damage-free surfaces when machining normal forces can be progressively diminished to milli-Newtons per diamond grit (Bifano et al., 1991; Ma et al., 2003; Schmidt and Weigl, 2000; Yin et al., 2004). For brittle materials to be machined in the ductile or plastic regime, their plastic deformation behaviour becomes very important, which can be studied using nanoindentation technique which also probes the contact hardness, H_c, and the Young's modulus, E (Oliver and Pharr, 1992, 2004). Combined with in situ scanning probe imaging, nanoindentation imprints can be imaged and analysed to discern various deformation modes underneath the indenter (Schuh, 2006). In nanoindentation, loading rates mimic the dynamic feature in abrasive machining, which affect the mechanical behaviour of materials, such as bulk metallic glasses (Burgess et al., 2008; Golovin et al., 2001; Greer et al., 2004; Li et al., 2006; Schuh and Nieh, 2003) and silica glasses (Dey et al., 2011; Limbach et al., 2014). The loading rate effect is manifest on the force–displacement curves as discrete discontinuities and continuous deformation. These can be correlated to several phenomena including shear banding, dislocation activity, fracture and phase transformation (Schuh, 2006). The use of in situ scanning probe imaging plays a complementary role for the correct interpretation of the force–displacement responses.

This paper aimed to study the mechanical behaviour of LMGC at different loading rates using nanoindentation and in situ scanning probe imaging. The contact hardness and Young's modulus were measured as a function of loading rates in the range of 0.1–2 mN/s at the peak load of 10 mN. The intrinsic contact hardness was determined by the strain rate sensitivity model. The nanoindentation-induced mechanical behaviour was characterized by discontinuities in force–displacement curves and in situ scanning probe imaging. An analysis was conducted to understand the nanoindentation-induced mechanical behaviour of LMGC in relation to its machining responses in dental CAD/CAM milling processes with sharp diamond abrasives.

2. Experimental procedure

2.1. Materials

The material investigated was LMGC (IPS e.max CAD, Ivoclar Vivadent) blocks which can be directly milled using the chair-side or laboratory dental CAD/CAM systems to generate profiles of crowns, bridges, veneers and inlays. It consists of 57–80 wt% silica (SiO_2) and 11–19 wt% lithia (Li_2O) with some additives including 0–13 wt% potassia (K_2O), 0–11 wt% phosphorus pentoxide (P_2O_5), 0–8 wt% zirconia (ZrO_2), 0–8 wt% zinc oxide (ZnO) and other colouring oxides to form a $Li_2O–SiO_2–K_2O–P_2O_5–ZrO_2–ZnO$ system (Bühler-Zemp and Völkel, 2005). Li_2O and K_2O act as an oxide modifier while ZnO acts as a network forming agent (Beall, 1992; ElBatal et al., 2009). ZrO_2 is added to hamper the crystal growth, refine crystals and consequently to improve the material strength (Apel et al., 2007). The addition of ZrO_2 to the base glass also increases the glass transition temperature, viscosity, and crystallization temperature leading to a reduction in the crystal growth (Thieme and Rüssel, 2015). P_2O_5 is the nucleating agent which reduces the nucleating energy enabling a reduction in the crystallization temperature and a faster attainment of the lithium metasilicate phase (Wen et al., 2007; Zheng et al., 2008).

LMGC can be obtained by melting a base glass consisting of 69.3 wt% SiO_2, 15.4 wt% Li_2O, 6.05 wt% K_2O, 5.28 wt% ZnO and 3.84 wt% P_2O_5 at 1450 °C and annealing at 450 °C (El-Meliegy and