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1 Research Articles

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3 Multi-scale analysis of the effect of nano-filler particle diameter on the physical

4	properties	of CAD/CAM	composite i	resin blocks
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1 ABSTRACT

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The objective of this study was to assess the effect of silica nano-filler particle diameters 3 in a computer-aided design/manufacturing (CAD/CAM) composite resin (CR) block on 4 physical properties at the multi-scale in silico. CAD/CAM CR blocks were modeled, $\mathbf{5}$ consisting of silica nano-filler particles (20, 40, 60, 80, and 100 nm) and matrix (Bis-6 GMA/TEGDMA), with filler volume contents of 55.161%. Calculation of Young's $\overline{7}$ moduli and Poisson's ratios for the block at macro-scale were analyzed by 8 homogenization. Macro-scale CAD/CAM CR blocks (3×3×3 mm) were modeled and 9 compressive strengths were defined when the fracture loads exceeded 6075 N. MPS 10values of the nano-scale models were compared by localization analysis. As the filler size 11 decreased, Young's moduli and compressive strength increased, while Poisson's ratios and 12MPS decreased. All parameters were significantly correlated with the diameters of the 13filler particles (Pearson's correlation test, r = -0.949, 0.943, -0.951, 0.976, p < 0.05). The 14in silico multi-scale model established in this study demonstrates that the Young's moduli, 15Poisson's ratios, and compressive strengths of CAD/CAM CR blocks can be enhanced by 1617loading silica nanofiller particles of smaller diameter. CAD/CAM CR blocks by using smaller silica nano-filler particles have a potential to increase fracture resistance. 18

1 KEYWORDS

- 2 Composite resins; CAD/CAM; multi-scale analysis; compressive strength; maximum
- 3 principal strain
- 4

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1 1 INTRODUCTION

2	Computer-aided design/computer-aided manufacturing (CAD/CAM) composite
3	resin (CR) blocks containing a high density of nano-filler particles (Nguyen et al. 2012,
4	Nguyen et al. 2013, Okada et al. 2014) have been available for posterior restorations
5	(Lauvahutanon et al. 2014, Shembish et al. 2016). In comparison with conventional CRs
6	(Ferracane 2011, Ilie and Hickel 2009), CAD/CAM CR blocks exhibit superior
7	mechanical properties, such as flexural strength, Vickers hardness, and fracture toughness
8	in terms of their clinical wear performance (Thomaidis et al. 2013). Although Vickers
9	hardness values are lower than those of ceramics, flexural strength is comparable to that
10	of a ceramic block and within the acceptable range (>100 MPa) for single restorations
11	described in the ISO standard for ceramic dental implants (Lauvahutanon, Takahashi,
12	Shiozawa, Iwasaki, Asakawa, Oki, Finger and Arksornnukit 2014). In addition, the effect
13	of compressive cyclic loading on fatigue resistance of CAD/CAM CR crowns has been
14	investigated and there is no greater incidence of catastrophic failure and lower
15	microleakage compared with ceramic crowns (Kassem et al. 2012). These results suggest
16	that greater compressive strength possibly improves the fatigue resistance of CAD/CAM
17	CR crowns.

18

In conventional CRs, the Young's moduli measured by the nanoindentation test

1	increases as the average diameter of filler particles increases at constant volume content
2	(Masouras et al. 2008). To further improve the mechanical properties of CAD/CAM CR
3	blocks and achieve excellent clinical wear performance, optimization of the size of nano-
4	filler particles is required. However, the influence of nano-filler size on the compressive
5	strength of CAD/CAM CR blocks cannot be rigorously evaluated through in vitro testing,
6	because the average diameter of nano-fillers is not controllable in experimental CRs, and
7	crack initiations at the nano-scale during fatigue testing cannot be followed even by
8	observation with scanning electron microscopy (Shembish, Tong, Kaizer, Janal,
9	Thompson, Opdam and Zhang 2016).
10	In silico analysis combined with three-dimensional finite element analysis and
11	composite theory has been useful for evaluating specific differences, such as the volume
12	content of fillers in conventional CRs (Li et al. 2012). However, the controllable
13	parameter for in silico analysis has been limited to date to the volume content, and
14	excludes the size of the fillers.
15	<i>In silico</i> multi-scale analysis (Fig. 1) is a coupled method for analysis of physical

properties or behaviors between different scales that has been used to predict mechanical properties of composite materials (Guedes and Kikuchi 1990) and has been applied to the prediction of mechanical strength of porous ceramics (Takano et al. 2003) and titanium

1	(Takano et al. 2010). This approach consists of homogenization analysis and localization
2	analysis. Homogenization analysis is a method for obtaining physical properties at the
3	macro-scale from knowledge of nano- or micro-scale structures. Nano- or micro-scale
4	stress and strain distribution for each component material in composite materials can be
5	visualized by localization analysis. This multi-scale approach enables the investigation of
6	the influence of nano-filler particle size on the physical properties at the macro-scale.
7	The objective of this study was to assess the effect of silica nano-filler particle
8	diameter in CAD/CAM CR blocks on the Young's modulus, Poisson's ratio, and
9	compressive strength at the macro-scale, and the maximum principal strain (MPS) at the
10	nano-scale through in silico multi-scale analysis.

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1 2. MATERIALS AND METHODS

2 2.1. In silico models

3	Nano-scale CAD/CAM CR block models were designed using CAD software
4	(Solidworks Simulation 2011, Dassault Systèmes SoldWorks Corporation, Massachusetts
5	USA), consisting of silica nano-filler particles (20, 40, 60, 80, and 100 nm) and matrix
6	(Bis-GMA/TEGDMA) (45×45×45, 90×90×90, 135×135×135, 180×180×180 and
7	225×225×225 nm), with filler volume contents of 55.161% (Fig. 2). The material
8	properties of silica and Bis-GMA/TEGDMA are shown in Table 1.

9

10 **2.2.** Homogenization analysis

A macro-scale CAD/CAM CR block model (3×3×3 mm) was designed by 11 Solidworks Simulation 2011 (Fig. 3) for compressive analysis. Calculation of Young's 12moduli and Poisson's ratios for the block were conducted by homogenization analysis in 13computer-aided engineering software (VOXELCON2015, Quint Corporation., Fuchu, 14JAPAN). Compressive strengths were defined as the values when the fracture loads 15exceeded 6075 N. The MPS distribution in the macro-scale model was observed and 1617normal strains in x-, y-, and z-axes and shear strains in yz-, zx-, and xy-planes were recorded at the location of the maximum MPS value. 18

1

2	2.3. Localization analysis
3	The MPS distribution in nano-scale models at the location where the maximum
4	MPS values were obtained in the macro-scale model was examined by localization
5	analysis (VOXELCON2015). The maximum MPS values at the nano-scale, related to the
6	threshold of fracture initiation, were compared.
7	
8	2.4. Statistical analysis
9	All parameters were statistically analyzed using Pearson's correlation test (PASW
10	Statistics 18, IBM, New York, USA).
11	

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1 3. RESULTS

3.1. Homogenization analysis

3	Young's moduli, Poisson's ratios, and compressive strengths at the macro-scale are
4	shown in Table 2. The results of macro-scale analyses show that the maximum MPS
5	values were observed at the vertex of the top surface of the cube (Fig. 4). As the filler
6	diameter decreased, Young's moduli and compressive strengths increased (Fig 5a and 5c),
7	while Poisson's ratios decreased (Fig 5b).
8	
9	3.2. Localization analysis
10	Maximum MPS values at the nano-scale containing 20-, 40-, 60-, 80-, and 100-nm
11	filler particles are shown in Table 2. As the filler diameter decreased, the maximum MPS
12	values decreased (Fig. 5d). The maximum MPS values were observed in the matrix resins
13	at the nano-filler particle spacing (Fig. 6). The nano-scale model containing 20-nm filler
14	particles showed the lowest concentration of MPS values compared with the other models
15	(Fig. 6a).
16	

3.3. Statistical analysis

All parameters were significantly correlated with the diameters of filler particles
(Pearson's correlation test, r = -0.949, 0.943, -0.951, 0.976, *p* < 0.05).

1 4. DISCUSSION

By comparing the maximum MPS values amongst the nano-scale models at the $\mathbf{2}$ location where the maximum MPS values were obtained at the macro-scale, the model 3 4 with the greatest compressive strength was identified under the assumption of the following MPS theory (Hearn 1997). MPS has been used as a failure criterion for the $\mathbf{5}$ matrix in fiber-reinforced composites during in silico analysis, resulting in good 6 agreement with experimental results (Hoover et al. 1997, Xia et al. 2000). Our results are $\overline{7}$ the first report describing the investigation of MPS propagation in nano-scale filler 8 particles and the resin matrix to elucidate the detailed mechanism behind compressive 9 strength. The in silico multi-scale analysis used in this study is useful for understanding 10 detailed stress and strain distributions at the nano-scale, which is not possible through in 11 vitro testing, and is applicable to the investigation of the influence of other controllable 12parameters such as geometric shape and the spatial layout of nano-filler particles. 13With regard to the homogenization analyses, Young's moduli showed a 28.131% 14increase of the nano-scale model containing 100-nm filler particles compared with that 15containing 20-nm filler particles, while Poisson's ratios decreased 10.619% at the same 1617volume content. The relationship between Young's moduli and Poisson's ratio was comparable with that for conventional CRs (Masouras, Akhtar, Watts and Silikas 2008). 18

1	Young's moduli of conventional CRs increase, while their Poisson's ratios decrease, as
2	the filler fraction increases (Li, Li, Fok and Watts 2012). However, the preparation of CRs
3	with high filler fractions is difficult because of theoretical limitations. In the case of face-
4	centered cubic lattice and hexagonal close packing structures using uniformly sized
5	sphere filler particles, the theoretical maximum content of particles is 74 vol% (Sloane
6	2003). The maximum filler content found for 72 commercial composites was 70 vol%
7	(Ilie and Hickel 2009). Higher filler contents induce lower fatigue resistance (Htang et al.
8	1995). Our results suggest that CRs with small, spherical nano-filler particles have the
9	potential to improve the Young's modulus and Poisson's ratio even at lower filler fractions
10	such as 55.161%, used in this study. Compressive strengths increased by 31.010% in a
11	nano-scale model containing 20-nm filler particles from that containing 100-nm filler
12	particles, while the maximum MPS values were decreased by 71.413%. Under the same
13	amount of loading, lower values of the maximum MPS definitely showed greater
14	compressive strength because the maximum MPS is defined as the threshold required to
15	initiate compressive fracture. These results suggest that the compressive strength of
16	CAD/CAM CR blocks with constant volume content can be improved by using smaller
17	filler particles.

18

With regard to the localization analysis, the model containing 20-nm filler particles

1	showed the lowest maximum MPS at the nano-scale and must have the greatest total
2	surface area of filler particles at the same volume content compared with the other models.
3	This phenomenon, called the "nano-effect", induces an enormous interfacial area per unit
4	volume (Fiedler et al. 2006, Wichmann et al. 2006) and the silane coupling rate increases
5	in association with the increase in the total surface area of filler particles. Although there
6	is a lack of simulation for the silane coupling ratio between silica nano-filler particles and
7	matrix resins (Tanimoto et al. 2006), the physical properties of CAD/CAM CR blocks
8	obtained in this study were identified, with the exception of the relative decrease in
9	absolute values associated with failure rates of the silane coupling. In addition, uniformly
10	dispersed, smaller nano-filler particles contribute to energy absorption and dissipation
11	through the nano-effect (Opelt et al. 2015). The propagation of elastic waves on human
12	tooth enamel has been analyzed by the time-dependent finite element method and moving
13	particle simulation (Yamaguchi et al. 2014). This approach is applicable to evaluate the
14	performance of energy absorption and dissipation in CAD/CAM CR blocks, while a static
15	analysis was used in this study.

Further fatigue testing to evaluate the long-term performance of CAD/CAM CR crowns has also been required (Harada et al. 2015). High fracture resistance of a mandibular first molar crown fabricated by Lava Ultimate then IPS Empress CAD has



1 5. CONCLUSIONS

The *in silico* multi-scale model established in this study demonstrated that the Young's moduli, Poisson's ratios, and compressive strengths of CAD/CAM CR blocks can be enhanced by loading silica nano-filler particles of smaller diameter. CAD/CAM CR blocks by using smaller silica nano-filler particles have a potential to increase fracture resistance.

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1 Tables

- 2 **Table 1**. Material properties of silica and Bis-GMA/TEGDMA used for homogenization
- 3 analysis

	Young's modulus (MPa)	Poisson's ratio	~
Silica	72000	0.16	RY
Bis-GMA/TEGDMA	2000	0.45	87
		S	

5 **Table 2**. Physical properties resulting from homogenization and localization analyses

Size of nano-filler particles (nm)	20	40	60	80	100
Young's moduli (GPa)	16.739	15.110	13.814	13.524	13.064
Poisson's ratios	0.303	0.320	0.332	0.335	0.339
Compressive strengths (MPa)	752	675	611	597	574
Maximum MPS	2.652e-3	4.943e-3	7.081e-3	7.340e-3	9.277e-3

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 $\mathbf{4}$

1 Figure Legends

- $\mathbf{2}$
- 3 Figure 1. Schematic of multi-scale analysis, consisting of homogenization and
- 4 localization analyses.
- $\mathbf{5}$
- 6 Figure 2. Nano-scale models containing (a) 20-, (b) 40-, (c) 90-, (d) 135- and (e) 180-nm
- 7 filler particles.
- 8
- 9 Figure 3. Macro-scale model.
- 10
- 11 Figure 4. Maximum principal strain distribution in macro-scale models containing (a) 20-,
- 12 (b) 40-, (c) 90-, (d) 135- and (e) 180-nm filler particles.
- 13

Figure 5. Relationship between nano-filler diameters and physical properties resulting
from homogenization and localization analysis. (a) Young's modulus, (b) Poisson's ratio,
(c) compressive strength, and (d) maximum value of maximum principal strain in nanoscale models.

- 1 Figure 6. Maximum principal strain distribution in nano-scale models containing (a) 20-,
- 2 (b) 40-, (c) 90-, (d) 135- and (e) 180-nm filler particles. Red arrows indicate the locations
- 3 of maximum values of maximum principal strain observed.
- 4
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1 Figure 5

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