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The Influence of Resin Matrix on The Water Sorption of Fiber-Reinforced Composites for DENTAL Use

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Abstract: Bis-GMA as resin matrix of FRC is widely used in dentistry is reported as cytotoxic. The resin matrix studied, 1,6 hexanediol dimethacrylate (HDDMA) has similar reactive groups than bis-GMA is not known as carcinogens. Clinically, resin matrix absorbs water and lower the material's strength. This study investigated the water sorption of FRC based on HDDMA monomer compared to bis-GMA. Fifteen specimens (2.0 mmx 2.0×25.0 mm) were divided into (Group-1): 78.4 wt-% HDDMA+19.6 wt-% MMA+1.0 wt-% CQ+1.0 wt-% CEMA, (group-2): 49.0 wt-% HDDMA+49.0 wt-% MMA+1.0 w-% CQ+1.0 wt-% CEMA and (group-3): 78.4 wt% bis-GMA+19.6 wt-% MMA+1.0 wt-% CQ+1.0 wt-% CEMA. Each specimen contained two E-glass fiber rovings. The specimens were light-cured and immersed in distilled water for 21 days. The differences in weights, before-after immersion were recorded. Group-1 proved the lowest average (0.004%), followed by group-3 (0.003%) and group-2 (0.01%). There was a significant difference among all groups ($p<0.05$) while LSD showed a significant difference between groups 1 and 2 ($p<0.05$) but no significant difference between groups 1 and 3 ($p>0.05$). It is concluded that FRC based on HDDMA matrix (group-1) is comparable to bis-GMA (group-3) for its water sorption.

Key words: Fiber-reinforced composite, bis-GMA, HDDMA, water sorption, comparable, monomer

INTRODUCTION

The loss of teeth that might cause functional disabilities needs the construction of a prosthesis. One of commonly used permanent treatment options is a crown or a bridge, usually made of Porcelain-Fused-to-Metal (PFM). The advantage of such prosthesis is the relatively natural appearance and good mechanical properties. Unfortunately, because porcelain used in the construction relatively brittle, it easily cracks and fracture (Hobkirk *et al*., 2003). Another disadvantage that might happen is corrosion in the metallic part (Freilich, 2000). The development of Fiber-Reinforced Composite (FRC) has provided the dentists the possibility of fabricating resin-bonded restorations with aesthetically good appearance (Zhang and Matinlinna, 2012). In addition, they are metal-free tooth restorations for single and multiple teeth replacements (Garoushi *et al*., 2011). Now a days, FRC is gaining growing popularity as a dental material of choice (Schutt *et al*., 2004). Fiber reinforced composites consist a resin matrix using either E-glass or carbon fibers for reinforcement (Zhang and Matinlinna, 2012). Fiber-reinforced composite materials employ fine thin fibers as a reinforcement which provides excellent tensile strength, crack formation stopping and high flexural modulus (Mallick, 2007). The superiority of FRCs compared to resin composites with fillers is its strength (Van Noort, 2007). Basically, FRCs have at least two distinct constituents, the reinforcing component fibers which give high strength and stiffness while the surrounding resin matrix supports reinforcement (Freilich, 2000). It is stated that E-glass fibers have high tensile strength, good impact and compression resistance properties which make it more desired as a reinforcing material. E-glass fibers need silanization and sizing to bond with the resin matrices (Lung and Matinlinna, 2012).

The structure of FRC is based on an Interpenetrating Polymer Network (IPN) structure, whereas the matrix usually consists of a cross-linking polymer, a linear polymer and a photo-initiator to facilitate polymerization (Zhang and Matinlinna, 2011). The mechanical strength of FRC depends among others on the impregnation of fibers within the resin matrix and adhesion of fibers to the matrix system (Vallittu, 1998, 1999; Vallittu and Sevelius, 2000; Soderholm and Mariotti, 1999) one of the most commonly used resin matrix which forms highly cross-linking polymer structures is bis-phenol-A-diglycidylmetha-crylate (bis-GMA) (Zhang and Matinlinna, 2011). Methyl Methacrylate (MMA) is a

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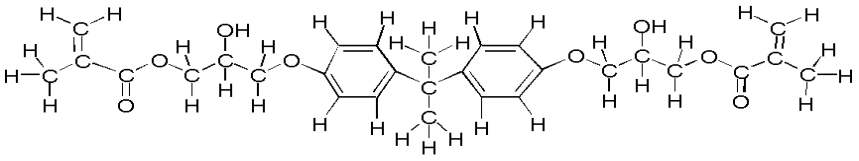

Fig. 1: The structure of bis-phenol-A-glycidylmethacrylate (bis-GMA)

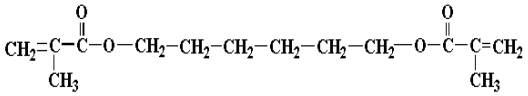

Fig. 2: The structure of 1,6-Hexanediol Dimethacrylate (HDDMA)

linear polymer (Zhang and Matinlinna, 2011) and the polymerization can be initiated using light, heat or chemical compounds (Anusavice *et al*., 2012). The photo-initiator system includes a photosensitizer and a reducing agent. Camphoroquinone (CQ) and N-N-Cyanoethyl Methylaniline (CEMA) are the commonly used photosensitizer and reducing agents (Hobkirk *et al*., 2003). Some released compounds from resin composites might cause biological reactions (16) and a case of allergic contact caused by bis-GMA was reported (Stoeva *et al*., 2008). Bis-GMA is also reported as the most cytotoxic monomer among dental resin composite monomers (Moharamzadeh *et al*., 2009). There are also some concerns that the use of bis-GMA is considered to be relatively hazardous. This is why nowadays an option of having other matrixes is gaining more and more interest. Next to this, there is a need to replace bis-GMA which is relatively hydrophilic by another hydrophobic matrix which exhibits lower water uptake (Sideridou *et al*., 2004). The structure of bis-GMA (Fig. 1) shows the hydrophilic hydroxyl groups (-OH).

A novel resin matrix consisting of 1.6 Hexanediol Dimethacrylate (HDDMA) has similar reactive groups (Fig. 2) as bis-GMA. However this resin monomer has low viscosity and it is a fast curing monomer with low volatility, hydrophobic backbone and it can be set byfree radical polymerization. The structure of HDDMA is as Figured below (Vallittu and Sevelius, 2000).

The HDDMA features water repellency property, i.e., it is hydrophobic. It is used as a functional monomer for polymers in technology and as a cross-linking agent between molecular chains of polymers. The applications of HDDMA include adhesives and sealants, coatings, elastomer, photopolymers electronics, improved adhesion, hardness, abrasion and heat resistance. It is reported that HDDMA does not produce mutagenic, embryotoxic, teratogenic or reproductive effects in humans. Related to its potential carcinogenicity, it has been reported that none of HDDMA precursor components are listed by IARC, NTP, OSHA or ACGIH as carcinogens. A preliminary result has suggested that E-glass FRCs with 78.4% HDDMA showed good flexural strength and hardness (Siswomihardjo *et al*., 2012).

Dental resin composites are extensively used in dentistry due to their aesthetic and adequate physical and mechanical properties (Rahim *et al*., 2012). The oral environment is very moist due to the presence of water in saliva and other fluids of the mouth and this may cause the hydrolytic degradation of resin composites. This condition will effect in the swelling of the material due to the sorption of water into the resin matrix and in turn, it will plasticize the resin composite (Eliades *et al*., 2005). It might also increase the material solubility, causes leakage of fillers which in turn breaks the bond between filler and matrix. This condition may cause a weakening of the material (Lassila *et al*., 2002) and compromise the mechanical and physical properties (Keyf and Yalcin, 2005). A long term aging, for about 2 year of resin composites in water has proved significantly to reduce the material fracture toughness (Drummond, 2008). Another issue of resin composites in the oral environment is the release of unreacted monomers from the material which may stimulate the growth of bacteria and promote allergic reactions (Sideridou *et al*., 2004). The immersion time for most resin composites, under which they normally will saturate is within 7-60 day (Rahim *et al*., 2012). Some other acrylic resin materials may require only a period of 17 days to become fully saturated with water (Anusavice *et al*., 2012). A study conducted by Zhang and Xu (2008) showed that the highest elution of monomer decreased on 21 days of water storage. The objective of this laboratory research was to measure the weight difference, before-after water immersion and study the water sorption of a novel resin matrix system of FRC based on HDDMA.

MATERIALS AND METHODS

Experimental: The flow of research is as figured in Fig. 3.

Specimen preparation: Two bundles of 25 mm long fibers reinforcement were placed along the long axis of

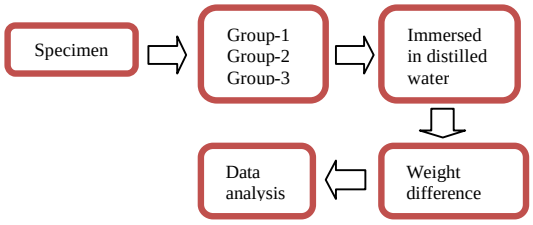

Fig. 3: Block diagram of the research method

Table 1: Matrix composition (in wt %) of the three groups

	Components (%)				
Groups	MMA	Bis-GMA	HDDMA	CQ	CEMA
1	19.6	- 7	>8.4	1.0	1.0
2	49.0	-	49.0	1.0	1.0
3	19.6	78.4	-	1.0	1.0

specimen into a custom-made brass mold and embedded into the resin matrix with different compositions as shown in Table 1 (Zhang and Matinlinna, 2011). Each study group of various matrix composition consisted of 5 specimens. Totally fifteen specimens with the dimension of (2.0× 2.0×25.0 mm) were prepared (Mallick, 2007). Next, all specimens were light-cured on both visible sides with a light curing unit (3×40 sec) to ensure polymerization and setting. After light-curing, all specimens were carefully polished using polishing paper of 360 grit to remove any extra edges (Matinlinn *et al*., 2009). The specimens were then immersed in distilled water for 24 h and kept at 37°C before the testing. Group-3 acted as the control group.

Specimens immersion in distilled water: Each specimen was tied up with thread and immersed it in a tube filled with 15 mL distilled water. All tubes with the specimen in it were kept in the incubator at 37°C for 21 days. The weight difference of the specimens, between before and after immersion were recorded.

RESULTS AND DISCUSSION

Based on the issue that bis-GMA might consider to be relatively cytotoxic and allergenic (Zhang and Matinlinna, 2011), the idea of this research was to suggest a monomer, HDDMA, to replace bis-GMA as matrix component in a novel FRC material. It has been proved that HDDMA has shown only moderate toxicity to mouse fibroblasts (Thonemann *et al*., 2002). A recent research proposed that HDDMA with the concentration of 78.4% produced comparable and good mechanical properties (Siswomihardjo *et al*., 2012).

The weights of all fifteen specimens before and after 21 day immersion were measured and the weight differences were calculated. Group-1 showed the lowest average (0.004%), followed by group-3 (0.003%) and group-2 showed the highest average (0.010%).

Table 2: The ANOVA of weight difference

Weight	Sum of squares	df	Mean square	F-value	Sig.
Between groups	0.002	2	0.001	32.667	0.00001
Within groups	0.012	12	0.001		
Total	0.022	14			

These results suggest that there was a process of water absorbtion and it is also stated by another study (Zhang and Matinlinna, 2011). In addition, in their study a novel E-glass FRC continuously have absorbed water from the moment of immersion. After polymerization composites are not stable and they will constantly be interacting with the oral environment and its humidity (Khalil, 2005). The problem associated with restorative materials is that water absorption is a fact because such acrylic materials are continuously rinsed with saliva. This said water absorption may induce weakening of the resin matrix (Biradar and Biradar, 2012). Water absorption for many dental materials is inevitable as restorative materials are continually in a wet and humid environment (Cabe and Walls, 2013). It is known that water diffuses into the resin matrix causing two opposing phenomena to take place. Firstly, water will leach out free unreacted monomers and ions which will contribute to a shrinkage and loss in weight of the material. Secondly, hygroscopic absorption of water will result in a swelling and increase in weight of the material. At last, water sorption may effect composite resin by reducing the mechanical properties and wear resistance (Khalil, 2005;Cabe and Walls, 2013).

In the current study, statistical analysis was performed with the ANOVA and the results showed (Table 2) that the matrix significantly influenced the weight difference of E-glass FRCs, before and after the immersion. This result is related to the finding that hydrophilicity of the polymer matrix is a factor that will influence the process of water sorption in composite resin (Eliades *et al*., 2005). Water sorption of resin composite is highly dependent upon the chemical structure of the resin monomers (Ferracane, 2006). If the monomers are hydrophilic related to the presence of polar groups in their structure they tend to be attracted to water molecules to form hydrogen bonding (Rahim *et al*., 2012; Matinlinn *et al*., 2013). Hydrophilic resins absorb more water and expand to a greater extent than hydrophobic resins. The volume of water absorbed by a material is determined by the content of the hydrophilic monomers (Khalil, 2005).

The posthoc analysis was performed using the Least Significance Difference (LSD) test. There was a significant difference observed between group 1 and 2 in the weight difference. This result might be related to the observation

that a novel FRC with 78.4wt-% of HDDMA showed good mechanical properties (Siswomihardjo *et al*., 2012). As for group 2 and 3, there was also a significant difference. It can be related to the fact that group-3 contained a hydrophilic resin (bis-GMA) which absorbs more water than HDDMA (Thonemann *et al*., 2002; Ferracane, 2006). The nature of hydrophilic resin means that such a resin has the ability to enhance water sorption (Yiu *et al*., 2004), HDDMA is more hydrophobic than bis-GMA (Ling *et al*., 2009). On the other hand, between group 1 and 3 no significant difference was observed, as it was supposed that there could be a difference. These results from this pilot research showed that group-3 showed a higher average in the weight difference than group 1, although statistically this difference was not significant. However, based on this result, it can be explained that although bis-GMA has different properties than HDDMA, but with the same concentration of bis-GMA and HDDMA the result may be comparable for water sorption. However, to gain the big picture and an optimal novel E-glass FRC composition, a more experimental study is necessary.

CONCLUSION

The effect of different resin matrixes and concentrations on water sorption has been studied. It was shown that HDDMA as a hydrophobic resin matrix component has a higher average of weight difference than bis-GMA. Finally, it can be concluded that a fiber reinforced composite based on 78.4wt-% HDDMA matrix system (group-1) is comparable to 78.4wt-% bis-GMA matrix system (group-3) on its water absorption and thus HDDMA might substitute bis-GMA as an FRC matrix component.

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