Nano - analysis Model of Curing and Adhesion in Dental Biomaterials

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Abstract
This purpose was to propose nano-analysis model to calculate cure performance and adhesive strength in new dental biomaterials. The experiment was done by micro tensile test, using micro tensile samples already defined and used[1,2]. Micro test samples had bonding area with three different interfaces and four layers in this study. Namely, the interfaces within the samples between resin composite and dentine and the layer were defined for the test samples : hybrid layer/dentine interface (interface I), adhesive resin layer/hybrid layer interface (II) and resin composite/adhesive resin layer interface (III). As previously reported, new dental adhesive in dental bonding systems was one - step one - bottled system (etching/priming/bonding agents ; single bottle adhesive). Such bonding area was cured by three different curing systems as TBB-H_2O-O_2 system, chemical cure and visible-light cure[3]. So, for this micro test, three types of bonding area were proposed. The following results were summarized. The models showed different curing point and shrinkage direction within the bonding area, and also the formation of microcracks within the layers and the interfaces. The calculation models to obtain adhesive strength in micro test were given for these cases of different bonding area.

Introduction

A new dental biomaterial as one - step one - bottled bonding system was bonded to human dentine, and thus a new method to measure adhesive strength was needed to evaluate *real* bond strength between resin and dentine. On the contrary, the *apparent* strength value was obtained by old type of two - step or three - step bond system with an acidic primer, which removed smear layer on mineralized dentine. For a new dental biomaterial as one - step one - bottled system (single bottle including dentine conditioner, primer and bonding agents) the idea to obtain *real* adhesive strength value was considered.

Thus, dental bonding biomaterial using adhesion method to give interface strength to bonded test specimen was discussed about adhesive strength using micro tensile test. This method was done with very small sample cut from one human dentine, compared with conventional cylindrical test sample. Conventionally two - step or three - step system bonded to human dentine was used using macro tensile test with cylindrical sample. Such different diameter and height as 1 to 6 mm diameter and 1 to 6 mm height was carried out using ten pieces of different human dentine in conventional macro test.

The adhesive samples were changing from the natural dentine (apatite and collagen fibres) to a new composite of adhesive resin and collagen fibres, which was named as the hybrid layer, or resin - impregnated interdiffusion zone. Such layers were composed by resin composite, adhesive resin layer, hybrid layer and dentine within a bonding area. In the definition of the interfaces, interface I, interface II, and interface III were used as described in abstract. Previous studies showed that *apparent* adhesive strength was measured without defining the bonding area. Thus, the dentine as a biomaterial was considered as a natural composite, including the organic matrix which is made primarily of apatite crystallites and collagen fibres[1, 2, 4-15]. The adhesive specimen at micro test (1 mm width and 1 mm thickness) was made for a new adhesive system as dental biomaterials.

Previous methods were defined as *first generation* for conventional tensile test (Nakabayashi)[2], and *second generation* for micro - tensile bond test (Sano et al)[4]. In 2002, four layers and three interfaces were defined within adhesive samples in case of a newly - defined micro test (Wakasa et al)[8]. Such test samples were given as *third generation* of adhesive bonding test[10,11]. Also, a new micro

test to obtain adhesive shear strength has been proposed first as *fourth generation* (Wakasa, 2007)[13].

As *third* or *fourth generation, real* adhesive strength was calculated perpendicular to tensile direction or along the resin composite/dentine interface. Adhesive strength was given theoretically as interfacial strength of specimen samples with, or without hybrid layer, using Eshelby's inclusion model[10-13,15].

The present study was thus to propose the calculation model to clarify the relationship between cure performance and adhesive strength in dental biomaterial.

Materials and Methods

1. Materials

The micro test sample, which was the same test dimension as Shono study[14], was used. The occlusal enamel was removed and the portion of dentine was cut perpendicular to the long axis of the tooth.

Recent experimental work was applied to the present study to analyze adhesive strength in case of one-step one-bottled system as a new dental biomaterial[6,7]. The visible light - curing unit was used for adhesive resin to make hybrid layer after demineralization. As the bonded area of the samples was usually 0.25 to 12.25 mm^2, the area tested was 1 mm^2 (1 mm X 1mm) during the micro test, to compare with test samples by three different curing systems.

2. Models

2.1 Cure performance

In these cases, the model of curing systems was proposed for specimen samples between tooth and resin[3], which was cured by different curing performances (three different curing systems as TBB-H_2O-O_2 system, chemical cure and visible-light cure : Figure 1 (● ; an initiation point of curing, an arrow ; direction of curing shrinkage)). Thus, the bonding area including the interface II between adhesive resin layer and hybrid layer has to be considered between tooth and dentine.

2.2 Micro test

(1) The formation of microcracks

To calculate adhesive bonding strength of dental biomaterials, the model shown in Figure 2 was newly proposed to consider the bonding area including the interface II between adhesive resin layer and hybrid layer. Here, the interface was indicated as a dotted line for chemical cure (left) and visible-light cure (right). The formation of microcracks along the interface will be observed perpendicular to the direction of curing shrinkage, showing that both curing systems had different formation along different interfaces.

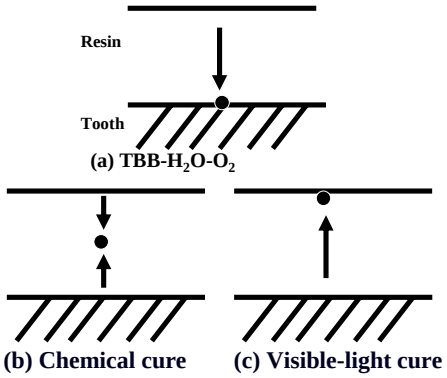

Figure 1 Curing systems

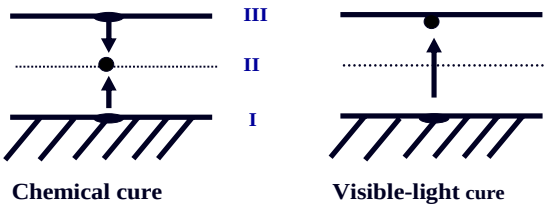

Figure 2 The formation of microcracks

(2) Sample dimension

Figure 3 was given as micro tensile test sample with four layers and three interface, and the micro tensile bond test was done (bonded area of 1 mm^2). Previous reports about micro test showed that the strength value was not *apparent* adhesive strength, but *real* adhesive strength[1,9-13,15].

Thus, this model showed that *real* adhesive tensile strength was given as that perpendicular

to the interface for the micro test.

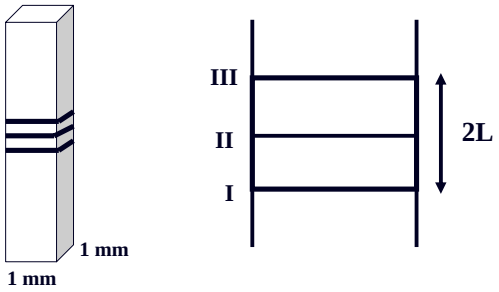

Figure 3 Specimen dimension

(3) The mechanism of fracture

Figure 4 shows the model of fracture mechanism within the bonding area to calculate fracture strength (adhesive strength) of new dental biomaterial in this study, when two types of curing performance, respectively, chemical cure (left) and visible-light cure (right), were used to cure bonding area of micro tensile test samples. The microcracks formed along the 45-degree to a tensile direction of the test sample.

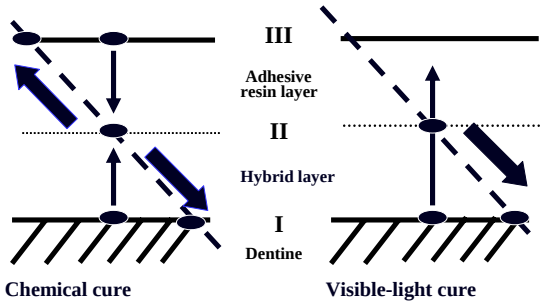

Figure 4 The mechanism of fracture

Four layers and three interfaces in Figures 2, 3 and 4, were used, and *real* adhesive interfacial strength was discussed as an interfacial strength of interface II (adhesive resin layer/hybrid layer interface) with a micro test (1mm X 1mm) as a model of *third generation* sample[11,12]. This model gave adhesive strength of interface II in case of thin hybrid layer, or normal thickness of hybrid layer, because the procedure to calculate adhesive strength with the bonding area was theoretically proposed[8-13,15].

2.3 Calculation models

(1) Fracture strength and elasticity of bonding area

Fracture strength values were measured, using test models of Figures 2 to 4. The values were then related to such elasticity value of each layer within the test samples as follows, as previously reported[9-12]. In this calculation, the elasticity values of hybrid layer and adhesive resin layer, measured by nano - indentation method, were used, as the load/deflection curves of hybrid layer or adhesive resin layer during loading/unloading at load = 1 gf was obtained. In this case the thickness of hybrid layer is 10 μm, and average indentation depth was about 3 μm. Elastic property was related to fracture strength analysis.

(2) Adhesive strength along the interface

Using fracture analysis based on the concept of fracture strength and critical stress, these values when crack formed along the interface were obtained using Eshelby's inclusion model[9-13,15]. Thus, adhesive strength of interface II was obtained as a *real* value, both considering fracture strength at the interfaces or within adhesive resin layer, and examining fracture mode during a micro test.

Results and Discussion

1.Adhesive strength along the interface

The values of cohesive strength with cohesive failure were measured for test samples. As fracture strength perpendicular to the interface, adhesive strength was estimated. Previous report clarified that, if the resin matrix was used as either bis - GMA - based or UDMA - based one, plastic deformation at UDMA - based one was larger than that of bis - GMA - based one with larger (Elasticity/Hardness) ratio, showing that the normalized values by their values, that is (b/2a), changed increasingly with increasing (E/H) ratio, using b (plastic deformation size), 2a (triangular indenter size) and E (Elasticity). It was thus

clarified that the elastic properties affected to adhesive strength as fracture strength along the interface II.

2. Curing system and adhesive strength

The effect of microcracks fraction on adhesive strength, as indicated in Figure 4, was observed to obtain the value of adhesive strength within the bonding area cured by chemical cure or visible-light cure (Figures 5 and 6).

In Figure 5, adhesive strength in case of chemical cure was calculated, considering the interaction energy between a main crack along the interface II and microcracks within the layer.

The value changed with a wide range of shrinkage strain at each applied strain.

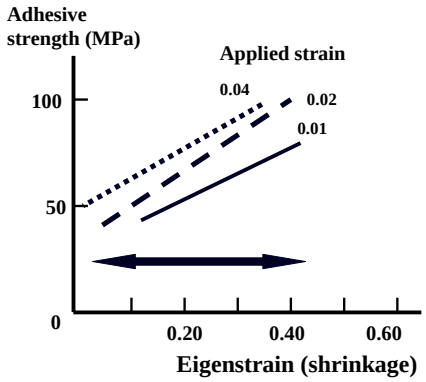

Figure 5 Adhesive strength in chemical cured sample

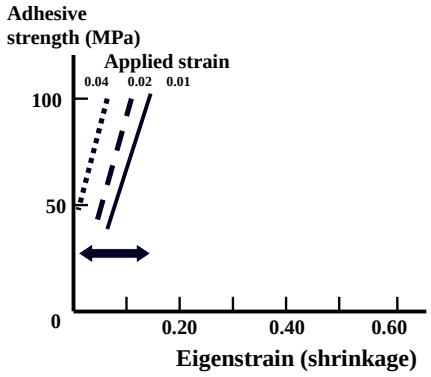

Figure 6 Adhesive strength in visible-light cured sample

In Figure 6, adhesive strength in case of visible-light cure was also calculated. The value changed with a very narrow range of shrinkage strain, compared with test samples of chemically-cured bonding area.

Thus, it was clarified that the calculated values depended on the interfacial properties. This method is appropriate to calculate the relation between curing performance and adhesive strength parallel to the interface in dental bonding biomaterials.

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References

1)K.Wakasa et al, *ECCM CTS-4*, IOM Com, London, (1998) 253-261.
2)N.Nakabayashi, *J. Japan Dent. Mater.* 1(1982)78-81.
3)N.Nakabayashi, M.Nakamura, N.Yasuda, *J. Esthet Dent.* 3(1981)133-138.
4)H.Sano et al, *Dent.Mater.* 10 (1994) 236 - 240.
5)K.Wakasa et al, *J.Dent.Res.* 78 (1999) Abst. No. 1430.
6)S.Uno et al, *Nihon Shika Rikou Gakkai Zasshi* 23 (2004) 412.
7)E.Yoshida and S.Uno et al, *Nihon Shika Rikou Gakka*i *Zasshi* 25 (2006) 107.
8)K.Wakasa et al, *The Proceeding of ICCE/9*, San Diego, USA, (2002) 827 - 828 .
9)K.Wakasa, *The Proceeding of ICCE/10.* New Orleans, USA , (2003) 929 - 932.
10)K.Wakasa, *The Proceeding of ICCE/11,* (CD - R) , Hilton - Head, USA, (2004) .
11)K.Wakasa, *The Proceeding of ICCE/12,* (CD - R) , Canary Island, Spain, (2005) .
12)K.Wakasa, *The Proceeding of ICCE/14,* (CD - R) , Boulder , USA, (2006) .
13)K.Wakasa, *The Proceeding of ICCE/15,* (CD - R) , Kaikou Island, China, (2007) .
14)Y.Shono et al, *The Proceeding of ICCE/7.* New Orleans, USA , (2000) 803 - 804.
15)K.Wakasa, *The Proceeding of ICCE/16,* (CD - R) , Kunming, China, (2008) .