# Effects of filler addition to bonding agents on shear bond strength

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ABSTRACT

## FILLER함량이 BONDING AGENT의 전단접착강도에 미치는 영향

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**목 적**

최근 개발된 bonding agent 중 일부는 다양한 함량의 filler를 포함하고 있으며 filler의 첨가는 bonding agent의 기계적인 물성을 향상시킴으로써 접착력의 향상에 기여한다는 주장이 있다.

본 연구에서는 다양한 함량의 filler를 포함한 adhesive를 실험적으로 만들어, filler의 함량이 전단접착강도에 미치는 영향을 살펴보고 임상적으로 가장 적절한 filler의 함량을 알아보고자 하였다. 또 adhesive의 간접인장강도를 측정하여 adhesive의 기계적인 물성과 접착력과의 상관관계를 알아보았다.

**방 법**

발거된 건전한 70개의 대구치를 투명 레진에 매몰하고 상아질면을 노출시켰다. 3M사의 Scotchbond Multipurpose의 etchant와 primer를 제조사의 지시대로 적용하고 1㎛크기의 barium glass filler를 0, 5, 10, 15, 20, 30, 45wt% 포함하도록 실험적으로 제작한 adhesive를 도포한 후 레진을 충전하여 시편을 완성하였다. Instron으로 0.5mm/min의 속도에서 전단접착강도를 측정하고 그 단면을 입체현미경으로 관찰하여 파절의 양상을 확인 하였다.

Filler함량에 따른 adhesive의 후경을 측정하기 위해 상기한 방법으로 시편을 제작하여 주사 전자현미경으로 관찰한 후 Sigmascan을 이용하여 그 후경을 측정하였다.

또, 지름 4mm 높이 6mm의 원통형 시편을 제작하여 Instron로 간접인장강도의 측정을 시행하였다. 얻어진 결과는 Kruskal-Wallis test와 Mann-Whitney test를 시행하여 분석하였으며, 상관관계를 분석을 위해 Pearson Product Moment Correlation Coefficient를 구하였다.

**결 과**

1) Filler함유량에 따라 전단접착강도는 유의할 만한 차이를 보였다($p<0.05$).
2) Filler함량의 증가에 따라 전단접착강도는 유의하게 증가하여 15% 수준에서 가장 높은 값(19.9±1.38Mpa)을 보였으며 20% 이상의 수준에서는 유의하게 감소하였다($p<0.05$).
3) Adhesive의 간접인장강도는 20% 수준까지는 증가하는 양상을 보였으나 통계적 유의성은 없었으며($p>0.05$), 30% 이상에서는 유의할 만한 감소를 보였다($p<0.05$).
4) Adhesive의 후경은 0% 수준에서 5.97±1.23㎛부터 45%수준에서 73.37±11.7㎛까지 유의하게 증가하였다($p<0.05$).
5) Filler함량에 따른 Adhesive의 간접인장강도와 전단접착강도는 상관관계가 없었다.

**주요어** : Bonding agent, Filler, 전단접착강도, 간접인장강도

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## Ⅰ. Introduction

In 1955, a new era in composite resin restorative dentistry began when Buonocore devised a manner to attain a predictable acid induced micromechanical bond of resin to enamel[1]. The successful bond to enamel induced further investigation into bonding systems. Since fourth generation bonding systems have been introduced, the clinical reliability of adhesive technique was greatly improved. The bonding agent used in fourth generation bonding systems forms an intermediate layer between dentin surface treated with dentin primer and the resin restoration, and chemically reacts with the dentin primer to provide micromechanical retention to tooth structure[2]. It is reported that in vitro bond strengths of resin composite to enamel exceed 20Mpa using these multi-step adhesive systems. This is sufficient to resist the shrinkage stress that accompanies the polymerization of resin composites[3]. However, the development of predictable bonding to dentin is still problematic because of its compositional and structural differences to enamel[4]. Only recently developed dentin adhesive systems produced laboratory results that approach those for enamel bonding[5-7].

In further investigations to dentin bonding system, one of the recent developments is the introduction of filled adhesives[8]. These bonding agents are loaded with varying proportions of fillers, which may be silica or glass of varying size. When using these newly developed dentin bonding systems, increased bond strengths were reported.

Fanning **et al.**[9] compared the mean shear bond strengths of filled and unfilled adhesive modalities of the All Bond 2 and Amalgambond plus and Optibond systems and concluded that the addition of filler particles in the adhesive agent resulted in an increase in the dentin bond strength.

Masashi **et al.**[10] reported that the maximum bond strength was obtained when 10% filler was added to the bonding agent.

One possible explanation for this phenomenon is the concept of an "elastic cavity wall"[11,12]. The intermediate layer, together with the resin impregnated dentin interface act as an elastic buffer, which offers the resin-dentin interface a sufficient strain capacity to dissipate the composite polymerization[13].

Enhancement of the physical properties of the bonding agent is another possible explanation for this phenomenon. Fanning **et al.**[9] suggested that the incorporation of filler particles into a system's adhesive could increase the potential shear bond strength by improving the mechanical properties. Improvements noted in such adhesive liner include greater strength, lower polymerization shrinkage, and lower linear coefficient of thermal expansion. But, a few studies have been done regarding the correlation between the dentin bond strength and filler level of bonding agent.

In this study, shear bond strengths of experimental filled adhesives with varying filler levels were tested to determine the optimal filler level. The diametral tensile strengths and thickness of each experimental adhesive were also tested to evaluate if there is a relation between shear bond strength and mechanical properties of adhesive.

## Ⅱ. Materials and methods

### A. Shear bond strength measurement

Seventy mandibular and maxillary human molars, not more than six months after extraction, were used for this study[13]. The teeth used for bond strength measurement were caries-free and unrestored. Teeth were placed immediately after extraction in a 0.5% chloramine solution for a week and thereafter stored in distilled water in a refrigerator at a nominal 4℃.

After the tooth was mounted in a holder by means of epoxy resin (Struers, Copenhagen, Denmark), the occlusal tooth surface was grinded until dentin surface was exposed. Automatic polishing machine (Pedemax-2, Struers, Copenhagen, Denmark) against silicon carbide abrasive paper with Grade 1000 was used for polishing the surface under running water.

Then the specimens were randomly assigned to seven groups. A Self-adhesive PVC tape with a

4mm diameter hole was placed over the dentin surface for confining the area of application. According to the manufacturer's instructions, etchant (3M, Dental products Inc., St. Paul. U.S.A.) was applied to dentin for 15 seconds and rinsed with water for 10 seconds. After removing excess water with an air syringe, Scotchbond multipurpose primer was applied. The dentin surface was gently dried for 5 seconds. Thereafter, each experimental adhesive (Vericom, Anyang, Korea) was applied to dentin and light cured for 10 seconds. The composition of experimental adhesive agents is shown in table 1. The average size of 1㎛m barium glass was used for the production of experimental adhesives. The particle size distribution of used filler is shown in Figure 1. 0, 5, 10, 15, 20, 30 and 45wt% filler was added to bonding agent. Then Z-100 (3M, Dental products Inc., St. Paul. U.S.A.)was applied in a bulk having 2mm thickness and light cured for 20 seconds. All the specimens were stored for 24 hours in distilled water at 37℃. Specimen preparation procedures are illustrated in Figure 2.

All the prepared specimens were mounted in a universal testing machine (model 4466. Instron corp.). A chisel-shaped rod attached to the compression load cell and traveling at a cross speed of 0.5mm/min was applied to each specimen until failure occurred[15]. The maximum load was divided by the cross sectional area of bonded composite to determine the shear bond strength in Mpa.

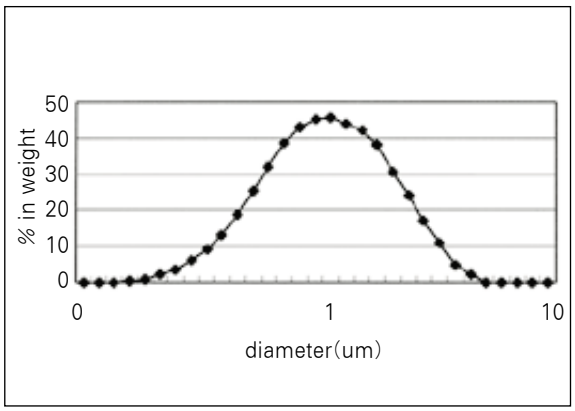


**Fig. 1.** Filler size distribution

**Table 1.** Composition of experimental adhesives (%)

| | Chemical | Ⅰ | Ⅱ | Ⅲ | Ⅳ | Ⅴ | Ⅵ | Ⅶ |
|---|---|---|---|---|---|---|---|---|
| Monomer/ Additive | Bis-GMA | 61 | 57.9 | 55 | 51.9 | 48.8 | 42.7 | 33.6 |
| | HEMA | 37.4 | 35.5 | 33.7 | 31.8 | 29.9 | 26.2 | 20.6 |
| | Additive | 1.6 | 1.5 | 1.4 | 1.3 | 1.3 | 1.1 | 0.8 |
| Filler | Barium glass | 0 | 5 | 10 | 15 | 20 | 30 | 45 |

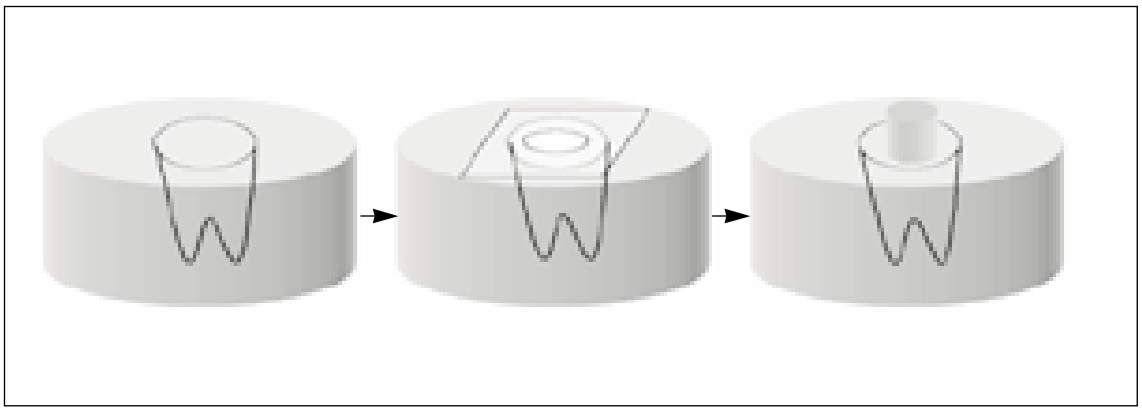

**Fig. 2.** Specimen preparation procedure

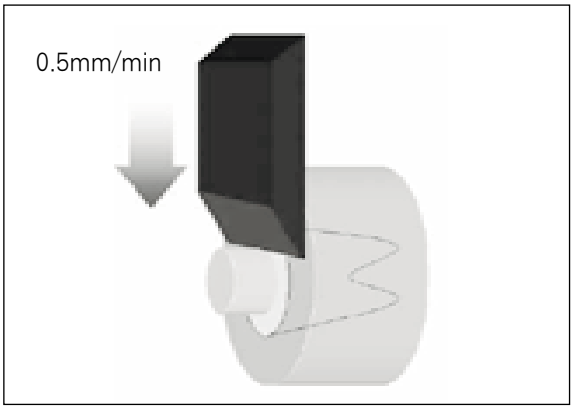


**Fig. 3.** Shear bond strength measurement.

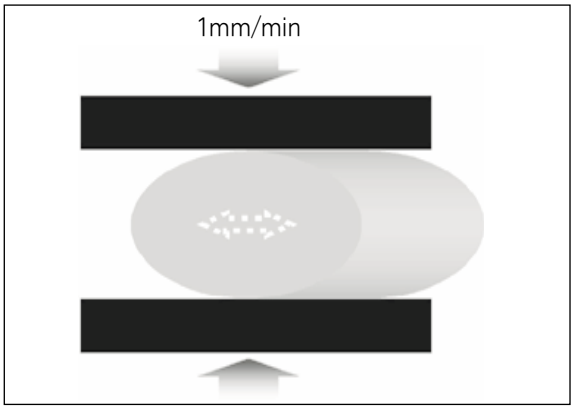


**Fig. 4.** Diametral tensile strength measurement

All the specimens were examined under optical microscope of 40x to define whether the failure was cohesive (within dentin) or adhesive (between the composite and dentin). The schematic representation of shear bond strength measurement is shown in Figure 3.

The results were analyzed by Kruskal-Wallis test, and then subjected to the Mann-Whitney test to make comparison among groups. All the statistical tests were processed with SPSS soft ware package.

### B. Diametral tensile strength measurement

Each test group was composed of ten specimens. Cylinder-shaped specimens with 4mm diameter and 6mm height were formed by addition of experimental adhesives to mold in approximately 2mm thick increments. Each increment was cured for 20 seconds prior to addition of the next. After the specimens were pulled out, the formed specimens were additionally cured in two opposite perpendicular directions for 20 seconds.

Then all the specimens were immersed in distilled water at 37℃ for seven days. For diametral tensile strength measurement, they were loaded in compression to failure in a universal testing machine (Model 4466. Instron corp.)with a crosshead speed of 1mm/min. The results were analyzed by Kruskal-Wallis test, and then subjected to the Mann-Whitney test to make comparison among groups. The schematic representation of diametral tensile strength measurement is shown in Figure 4.

### C. SEM examination

Five Specimens in each group were prepared in a manner similar to that described in shear bond strength tests. Then the specimens were molded in epoxy resin (Struers, Copenhagen, Denmark) and cross-sectioned perpendicular to the dentin surface with an Isomet low speed saw (Buehler Ltd., Lake Bluff IL). After the specimens were polished under with automatic polishing machine (Pedemax-2, Struers, Copenhagen, Denmark), the specimens were mounted on aluminum stabs, sputter-coated with gold and observed under scanning electron microscope (JSM-840A JEOL Ltd, Japan). 1000x,or 1500x observations were recorded and examined in photoshop 5.0 (Adobe system inc., moutina view, Ca, U.S.A). The thickness of adhesive layer was measured ten times at random position in each specimen using Sigma scan image version 2.0(Jandel Scientific software, San Rafel, Ca, U.S.A). The results were analyzed by turkey' test followed by T-test.

## Ⅲ. Results

The mean shear bond strengths and standard deviations for each group are shown in table 2.

Results showed that filler level had a statistically significant influence on bond strength ($p<0.05$).

In Mann-Whitney test, significant differences between 5, 10, 15, 20% filled groups are found. The maximum shear bond strength (19.9±1.38 Mpa) was obtained when 15% filler was added to the bonding agent. When fillers are added more than 20wt%, bond strengths were gradually decreased with increasing filler level.

The failure patterns of shear bond strength specimens are mostly cohesive. Fifty-one of seventy samples showed cohesive failures. Only nineteen of seventy samples showed adhesive failures. Ten of those adhesive failures are occurred in 30, 45% filled groups. The failure patterns are shown in figure 14, 15.

The diametral tensile strengths for each experimental adhesive are shown in table 3. The diametral tensile strengths for the experimental adhesives slightly increased with increasing filler levels in 0, 5, 10, 15, 20% filled groups but, showed no significant differences ($p>0.05$). But, the diametral tensile strengths were significantly decreased in experimental adhesives with over 30% filler levels ($p<0.05$).

**Table 2.** Shear bond strength of each group

| Group | Filler (wt%) | N | Adhesive failure | Mean±SD (Mpa) |
|---|---|---|---|---|
| 1 | 0 | 10 | 9 | 10.1±1.21 |
| 2 | 5 | 10 | 7 | 10.4±1.57 |
| 3 | 10 | 10 | 8 | 14.9±1.85 |
| 4 | 15 | 10 | 9 | 19.9±1.38 |
| 5 | 20 | 10 | 8 | 14.1±1.29 |
| 6 | 30 | 10 | 4 | 13.9±2.22 |
| 7 | 45 | 10 | 6 | 13.2±2.43 |

**Table 3.** Diametral tensile strength of each group

| Group | Filler (wt%) | N | Mean±SD (Mpa) |
|---|---|---|---|
| 1 | 0 | 10 | 175.8±10.5 |
| 2 | 5 | 10 | 182.1±21.9 |
| 3 | 10 | 10 | 186.5±6.63 |
| 4 | 15 | 10 | 183.4±12.2 |
| 5 | 20 | 10 | 181.9±11.4 |
| 6 | 30 | 10 | 165.8±12.9 |
| 7 | 45 | 10 | 142.6±15.5 |

**Table 4.** Adhesive thickness of each group

| Group | Filler (wt%) | N | Mean±SD (Mpa) |
|---|---|---|---|
| 1 | 0 | 50 | 5.97±1.23 |
| 2 | 5 | 50 | 4.16±0.94 |
| 3 | 10 | 50 | 6.42±1.58 |
| 4 | 15 | 50 | 13.1±0.93 |
| 5 | 20 | 50 | 27.7±1.49 |
| 6 | 30 | 50 | 32.2±2.36 |
| 7 | 45 | 50 | 73.4±11.7 |

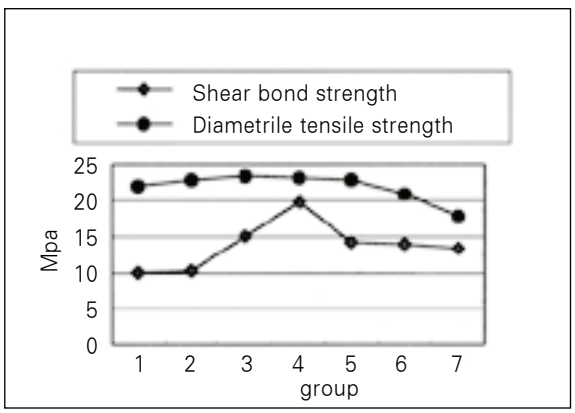


**Fig. 5.** Shear bond strength and diametral tensile strength

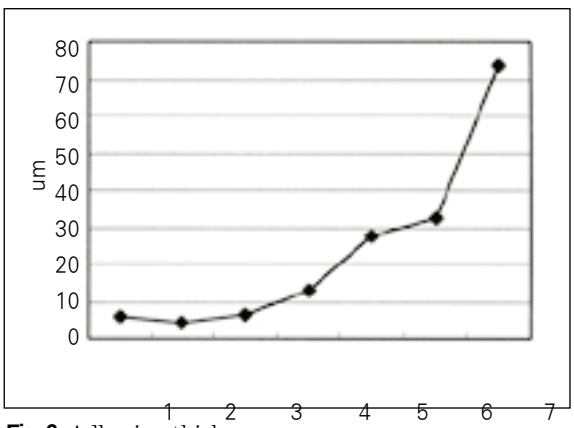


**Fig. 6.** Adhesive thickness

A relation between shear bond strength and diametral tensile strength was considered but no statistical significance was found by Pearson's correlation coefficient. The shear bond strength and diametral tensile strength are shown in figure 5.

In the SEM examination, the adhesive thickness of each group tended to significantly increase from 5.97μm at 0% filler level to 73.37μm at 45% filler level. Table 4 shows the adhesive thickness.

## Ⅳ. Discussion

In this study, the maximum shear bond strength (19.9±1.38Mpa) was obtained when 15% filler was added to the bonding agent. When filler levels were lower than 10% or higher than 20%, the shear bond strengths significantly decreased.

The result of shear bond strength in this study had a similarity to that of Masashi et al.10 In their study to investigate the shear bond strengths and temperature changes during polymerization using 0, 10, 20, 30, 40, 50, 60 and 70 % filled adhesives of average size 0.05μm, bond strengths to dentin and the temperature change were greatly affected by the filler level. They reported that maximum shear bond strength (14.3±2.3Mpa) was obtained with 10% filler level and shear bond strengths decreased with filler level higher than 30% (10.4±1.7Mpa -13.2± 2.43Mpa). They explained that the reason for the lower dentin bond strengths with higher filler level of experimental bonding agents were related to decreased penetration of adhesive monomers and to the existence of the internal voids due to the increased viscosity of experimental bonding agents.

But, in the SEM examination in our study, the experimental adhesives with highly filled groups closely adapted to primed dentin and internal void was also not found. Adversely, gaps between primed dentin and adhesive were found in 0% filled and 5% filled groups. These findings were consistent with low shear bonding strengths and diametral tensile strengths with low filler levels. The low bond strengths examined in lower filled groups might be related to oxygen inhibition. The thickness of adhesive layer in lightly filled adhesives founded to be approximately 6μm. Considering that the thickness of oxygen-inhibited layer is about 15μm[16,17], 6μm is not a sufficient thickness to be polymerized. These inadequately cured adhesives might prevent the establishment of the bond.

The major role of the adhesive resin is the stabilization of the hybrid layer and the formation of the resin extension into the dentinal tubules. It is recommended in this aspect that the adhesive resin be polymerized prior to the application of the restorative resins. In order to optimize the dentin bond strength, it is important to achieve a complete cure of bonding agent as quickly as possible after application.

The reason for decreased shear bond strengths of highly filled groups could be considered in another aspect. Diametral tensile strengths for the experimental adhesives showed no significant differences in 0, 5, 10, 15, 20% filled groups and significantly decreased in experimental adhesives with 30, 45% filled groups. In these highly filled groups, shear bond strengths are also significantly decreased. The decreased mechanical properties of highly filled groups might explain the lower shear bond strength. The thick but weak adhesive layer formed in highly filled groups might act as a site of failure. This result suggested that correlation between diametral tensile strength and shear bond strength might be exist. The increased adhesive failures in highly filled groups support this premise.

The addition of fillers reportedly improved serals of the liner's mechanical properties, which account for increase in bond strength[18-20]. Consequently increased diametral tensile strength with increasing filler level of experimental adhesives levels was also expected. But, in our experiment, diametral tensile strengths significantly decreased in experimental adhesives with higher than 30% filler levels. The unexpected result of diametral tensile strength leaved some thing to be considered. The concentration of relatively larger size of filler made the interfiller distance short

and might cause the adhesive layer more susceptible to crack propagation in these highly filled groups.

The mechanical properties of adhesives were closely related to their microstructures[21]. Factors such as the integrity of the interface between the glass particles and the polymer matrix, the particle size, and the number and size of voids have important roles in determining the mechanical properties. The interface between the filler and surrounding matrix has been thought to be a weak link.

The study of Lin CT et al.[22] support this premise. In their study of investigating the effect of silanization and filler fraction on the mechanical properties of aged dental composites, the result showed that diametral tensile strengths increased proportionally as the filler fraction of the composites increased in silanized groups. However, in the unsilanized groups, diametral tensile strength decreased as the filler fraction increased. Microscopic examination of the fractured samples showed that failure primarily occurred adjacent to the filler particles for unsilanized composites.

Dentin bonding strength and its related factor are quite complex and not fully understood. Mechanical properties of adhesive may be a key to understand the complex relation. But, mechanical property of adhesive is the result of an interaction of the various components. The diametral tensile strength and thickness of adhesive examined in this study are only a part of mechanical property of material. Other mechanical properties such as compressive strength, flexural strength and elastic modulus, degree of conversion, polymerization shrinkage are also important factors in evaluating the ultimate mechanical properties of dental materials. Further work is need to determine the effect of filler in dentin bonding agent and the various factors which may play a role in dentin bond strength.

## Ⅳ. Conclusion

According to this study we could summarize as follows:

1. The filler level showed statistically significant differences in shear bond strength ($p<0.05$).
2. Shear bond strengths increased with filler addition in bonding agent in some extent but, decreased when highly filled adhesive were used. And 15% filler level was considered as an optimal filer level for adhesives.
3. The diametral tensile strengths for the experimental adhesives significantly decreased in experimental adhesives in 30, 45 % filled groups ($p<0.05$).
4. The adhesive thickness of each group significantly increased with increasing filler level.
5. A relation between shear bond strength and diametral tensile strength was denied.

## Refernces


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## Explanation of figures

Fig. 7. SEM examination of Group1 (0% filled group)
Fig. 8. SEM examination of Group2 (5% filled group)
Fig. 9. SEM examination of Group3 (10% filled group)
Fig. 10. SEM examination of Group4 (15% filled group)
Fig. 11. SEM examination of Group5 (20% filled group)
Fig. 12. SEM examination of Group6 (30% filled group)
Fig. 13. SEM examination of Group7 (45% filled group)
Fig. 14. adhesive failure
Fig. 15. adhesive failure
Fig. 16. cohesive failure in dentin

## 사진부도 ①

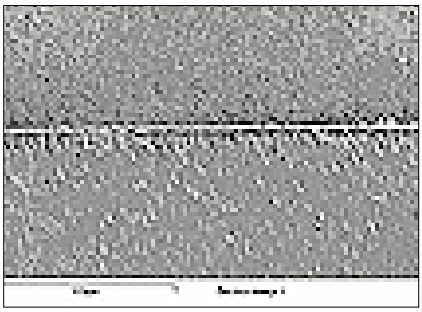

Fig. 7

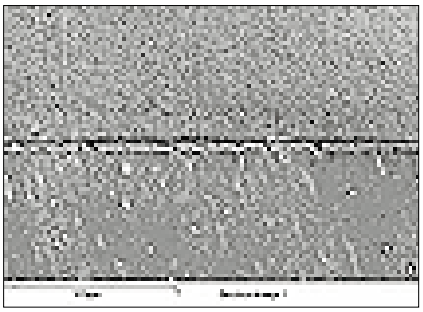

Fig. 8

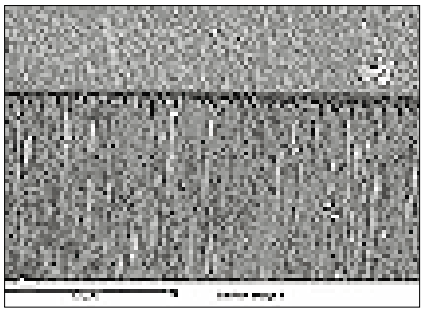

Fig. 9

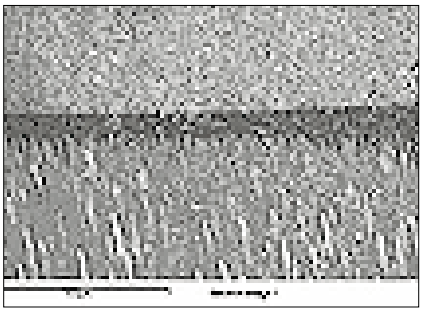

Fig. 10

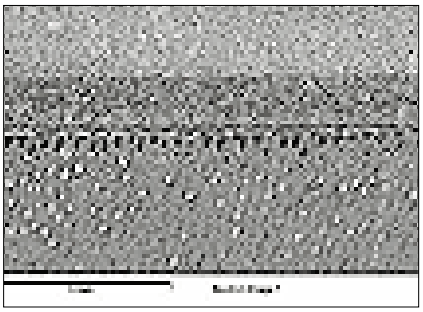

Fig. 11

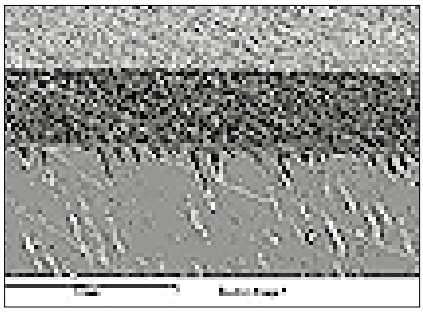

Fig. 12

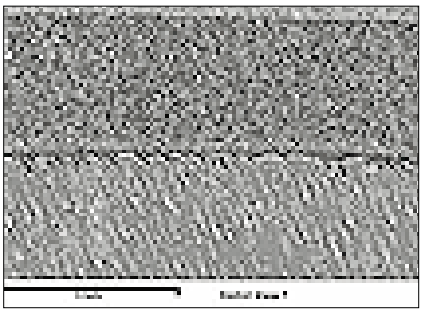

Fig. 13

Fig. 14

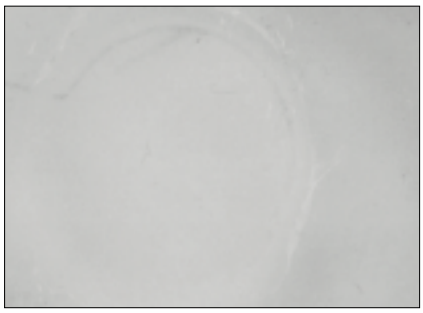

Fig. 15

Fig. 16