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Effect of conditioning and 1 year aging on the bond strength and interfacial morphology of glass-ionomer cement bonded to dentin

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1 INTRODUCTION

2 With the help of numerous research and clinical evidence, we are now able to accomplish
3 tooth adhesion to enamel and dentin to a satisfactory level by means of dental restorative
4 materials such as resin-based composites and glass-ionomer cement (GIC). Contemporary
5 focus is on ensuring materials are bioactive, tougher and self-reparable. The concept of
6 biocompatibility has evolved to bioactivity, which is now a big trend in restorative
7 dentistry[1].Dental restorative materials should be called “bioactive” only when they actively
8 stimulate or direct tissue responses, and they can control interactions with microbiological
9 species besides their primary function of restoring or replacing missing tooth structure [2].In
10 this sense, bioactivity has two major aspects, which are remineralization and anti-microbial
11 properties. Regarding remineralization, bioactive materials containing calcium phosphate [3],
12 hydroxyapatite [4,5], calcium silicate [6,7] etc, were reported to have remineralization ability.
13 Regarding the anti-microbial property, the release of compounds with antibiotic-like efficacy
14 added to dental restorative materials such as quaternary ammonium compounds [8], zinc
15 oxide nanoparticles [9] etc, were used to inhibit oral bacteria and biofilm.

16 GIC is one example of a dental bioactive material. It has both remineralization and anti-
17 microbial ability [10-13] and has been used for dental restoration and the Atraumatic
18 Restorative Treatment (ART) technique reliably for a long time [14,15]. Although resin
19 composite is the major dental restorative material used nowadays, GICs are often used in
20 clinical situations because of their technique simplicity, cost effectiveness and relative
21 tolerance in the moist oral environment. Additionally, having no conversion shrinkage is an
22 advantage compared with resin composite, and for relatively deep cavities it is still an ideal
23 material for use [16,17]. Moreover, Peumans *et al* reported the lowest annual failure rate
24 scores for GIC in vivo [18]. Although the bond strength of GIC may be much weaker
25 compared with resin-based materials, the means by which GICs obtain such clinically
26 satisfactory results is still not fully understood.

27 Some laboratory studies have reported improvement of the adhesion of GICs to tooth
28 structure in terms of bond strength when surface pre-treatment is carried out [19,20]. In
29 contrast, some other studies have reported certain GICs adhere to tooth structure without
30 pre-treatment [21,22].

31 The purpose of this study was to assess the tooth-GIC adhesion by means of bond strength
32 and interfacial morphology after 1 week and 1 year of aging, with and without surface pre-
33 treatment. The null hypothesis tested in this study was that pre-treatment of dentin using a
34 polyalkenoic acid conditioner did not affect the long-term durability of a conventional GIC.

35

36 **2.MATERIALS AND METHODS**

37 **2.1. Micro-tensile bond strength (μ TBS)**

38 The bond strength to dentin was determined using a standard micro-tensile bond strength
39 test [23].The materials used in this study are shown in Table 1. Six human molars, stored in
40 a 0.5% chloramine T solution, were used within 1 month of extraction. The protocol of this
41 research was approved by the Commission for Medical Ethics of Hokkaido University.The
42 extracted molars were sectioned at the mid-coronal portion to create a flat dentin surface by
43 using a low-speed diamond saw (Isomet 1000, Buehler, Lake Bluff, IL, USA). A standard
44 smear layer was produced using #600 grit silicon carbide paper. The teeth were randomly
45 divided into two groups of three teeth each. Prior to the application of the GIC, the dentin
46 surface of the specimens in one group was pre-treated with a polyalkenoic acid conditioner
47 (Cavity Conditioner, GC, Tokyo, Japan). This contains 3% Aluminum chloride as well as
48 20% polyalkenoic acid. The specimens in the other group did not receive any pre-treatment.
49 The dentin surface was subsequently built up free-hand and in bulk with a conventional GIC
50 (Fuji IX GP Extra, GC, Tokyo, Japan) to a height of 5-6 mm, followed by application of a
51 surface sealer (GC Fuji Coat LC, GC, Tokyo, Japan) which was light-cured for 10 seconds.

52 After 1 week of storage in distilled water at 37°C, the specimens were sectioned
53 perpendicular to the bonding surface, to obtain 1-mm² stick-shaped micro-specimens using

54 an Isomet saw. The specimens were then randomly assigned to four groups (10 specimens
55 each) according to age/storage time: 1 week and 1 year, *i.e.* the 1 week specimens were
56 tested after sectioning while the rest continued in storage to 1 year. This is based on the
57 following power calculation: if the specimen is used as the statistical unit, an absolute 3 teeth
58 per experimental group with appropriate consideration of tooth dependency are required [24].
59 At the relevant time period, the micro-specimens were fixed to a jig with cyanoacrylate glue
60 (Model Repair II Blue, Dentsply-Sankin, Ohtawara, Japan) and stressed in a testing device
61 (EZ-test, Shimadzu, Kyoto, Japan) at a crosshead speed of 1 mm/min until failure
62 occurred. The μ TBS was calculated in MPa, derived by dividing the force applied (in N) at the
63 time of fracture by the bonded area (in mm²). Statistical analysis was performed using one-
64 way ANOVA ($\alpha=0.05$) and *post hoc* Tukey-Kramer multiple comparisons tests. The mode of
65 failure was determined by examining the fractured surface at a magnification of 50x using a
66 stereo-microscope (Wild M5A, Heerbrugg, Switzerland).

67

68 **2.2. TEM interface characterization**

69 Additional GIC specimens were prepared for examination using TEM (H-800, Hitachi, Tokyo,
70 Japan). For this, a further four teeth were randomly divided into two groups of two teeth
71 each; the dentin was pre-treated with polyalkenoic acid conditioner in one group but not in
72 the other. The procedure of bonding the GIC to dentin was the same as previously described
73 in the μ TBS test, before storage in distilled water for 1 week and 1 year at 37°C. The GIC-
74 bonded dentin specimens were sectioned perpendicular to the GIC/dentin interface using an
75 Isomet diamond saw. From each tooth, seven or eight rectangular sections, of approximately
76 1 mm thickness each, were obtained. After storage for each time period, TEM sample
77 preparation was performed in accordance with common procedures used for ultra-structural
78 TEM examination of biological tissues [25]. This involved specimen fixation overnight in 2.5%
79 glutaraldehyde in 0.1 M sodium cacodylate buffer at pH 7.4 and 4°C, followed by rinsing in
80 0.1 M sodium cacodylate buffer for 1 min with 3 changes. Dehydration was carried out in
81 ascending grades of ethanol (50%, 75%, 95%, 100%) for 10 min each, with 2 changes. This

82 was followed by immersion in 1:1 absolute ethanol-epoxy embedding resin for 30 min, and
83 then resin infiltration in 100% epoxy embedding resin for another 4hrs. Finally, embedding of
84 the resin-infiltrated samples in molds with 100% epoxy resin was carried out. Before being
85 embedded, the specimens were oriented in the molds so that ultra-thin sections through the
86 GIC/dentin interface could be cut from the dentin part from each original tooth. The epoxy
87 blocks were polymerized in an oven at 60°C for a minimum of 48 hrs. Subsequently, non-
88 demineralized, 70–90 nm thin sections were cut using a diamond knife (Diatome, Bienne,
89 Switzerland) in an ultramicrotome (Ultracut UCT; Leica, Vienna, Austria). The GIC/dentin
90 interface in each section was observed by TEM.

91

92 **2.3. Chemical element analysis**

93 To analyze the chemical elements of the GIC/dentin interface, a Field Emission Transmission
94 Electron Microscope (FE-TEM) (JEM-2010F, JEOL, Tokyo, Japan) was used. The same
95 specimens prepared for TEM observation were used for the FE-TEM observation as well.
96 Images were captured and analyzed by STEM mode at 200kV.

97

98 **3. RESULTS**

99 **3.1. Micro-tensile bond strength (μ TBS)**

100 The mean μ TBSs are presented in Figure 1. No pre-testing failures (ptfs) were found in this
101 study.

102 There was no significant difference in μ TBS when Cavity Conditioner was used at each time
103 period ($p>0.05$). In addition, 1 year water storage did not show significant difference between
104 conditioned and non-conditioned dentin in terms of μ TBS results.

105

106 **3.2. SEM failure analysis**

107 At 1 week, the failure patterns were generally of a 'mixed' nature, involving areas that failed

108 at the interface and areas that failed cohesively within the GIC, for both the conditioned and
109 non-conditioned groups. At 1 year, while the failure was still of a mixed nature, there was a
110 tendency for more areas of cohesive failure. It appeared that aging of both conditioned and
111 non-conditioned specimens caused them to fail slightly more frequently cohesively within the
112 GIC.

113

114 **3.3. TEM interface characterization**

115 Representative TEM photomicrographs of unstained, non-demineralized sections of the
116 GIC/dentin interface with polyalkenoic acid conditioning using Cavity Conditioner stored for 1
117 week and 1 year are shown in (Figures 2 a&b), while GIC/dentin with non-conditioned
118 interface for 1 week and 1 year are shown in Figures 3 a&b).

119 A shallow partially demineralized dentin layer (De) of about 0.5-1 μ m was seen at the dentin-
120 conditioned interface (Figures 2 a&b). Hydroxyapatite (HAp) remained within this partially
121 demineralized layer. On top of this layer, a seemingly matrix-rich layer (ML) was seen; this
122 appeared to be of a few hundred nanometers for the 1 week specimens and of about 100
123 nanometers for the 1 year specimens (Figures 2a&b). On top of the matrix-rich layer (ML), an
124 intermediate layer (IL) of a few hundred nanometers was noted, and this zone was typically
125 demarcated from the rest of the GIC matrix by small electron-lucent globules (Figures 2 a&b).
126 Representative TEM photomicrographs of unstained, non-demineralized sections of the
127 GIC/dentin interface without polyalkenoic acid conditioning stored for 1 week and 1 year are
128 shown in Figures 3 a&b. The GIC was closely attached to the dentin surface without any
129 intervening layers detected (Figures 3 a&b). No clear signs of bond degradation were
130 observed after 1 year of water storage.

131

132 **3.4. Chemical element analysis**

133 The image of GIC/dentin interface with polyalkenoic acid conditioning for 1 week storage as
134 captured by FE-TEM is shown in Figure 4. There were 3 plots made in this analysis. Plot 1
135 indicated the GIC area, Plot 2 indicated the IL area and Plot 3 indicated the ML area.

136 Chemical compositions analyzed by STEM mode are shown in Figure 5. Plots 1 and 2
137 showed the various components of GIC such as Si, Sr, Al. Plots 1, 2 and 3 showed almost
138 the same tendency although Plot 3 showed more Ca content.

139

140 **4. DISCUSSION**

141 The use of Cavity Conditioner did not make a significant difference to the μ TBS. As cohesive
142 failure within the GIC tends to occur over time, this may be the reason why there was no
143 significant difference in μ TBS. The fact is that there was no difference in μ TBS even when
144 polyalkenoic acid conditioning was carried out, although it does not mean that there are no
145 advantages of surface conditioning. Polyalkenoic acid probably facilitates the calcium and
146 phosphate ions from dentin for the ionic reaction with GIC because it removes the smear
147 layer, increases the contact area and facilitates wetting of the surface [26-28]. Hence, it may
148 be difficult to evaluate the quality of the interface by means of only μ TBS in this case.

149 From the TEM photomicrographs of Figure 2a and Figure 2b, in the conditioned specimens,
150 we can observe different layers moving outwards from the dentin towards the bulk of the
151 GIC: a partially demineralized layer (De), a matrix-rich layer (ML) and a further intermediate
152 layer (IL). The De layer, within which HAp remained, is immediately adjacent to the
153 unaffected dentin. Beyond this, there is a zone (ML) that is reasonably well-defined; it may
154 be a zone that arises due to interaction between the acid-affected dentin layer and the glass
155 component of the glass-ionomer. This interaction was confirmed by the presence of more Ca
156 in the ML as detected by the chemical element analysis. The ML is followed by the next layer,
157 which appears to contain more unreacted glass. The differentiation of layers at the
158 conditioned interface is likely, given the high viscosity of the setting glass-ionomer material.

159 The widths of the De and IL layers were almost the same in the 1 week and 1 year samples.

160 In contrast, the dimensions of the ML reduced over time. This phenomenon may be ascribed
161 to the maturing effect of GIC, especially as with the use of polyalkenoic acid conditioning, the

162 calcium and phosphate ions' reaction with GIC was activated and the remineralizing effect
163 may have been promoted as well. The increase of apatite formation and mechanical property
164 could be expected, but this has to be confirmed in further work. There were no signs of
165 interface degradation comparing the 1 week and 1 year (Figure 2a and Figure 2b) interfaces
166 observed.

167 From the TEM photomicrographs of un-conditioned specimens (Figures 3 a&b), we can
168 observe the GIC area and dentin area without any intervening differentiation or layers; there
169 was no significant difference in μ TBS compared with the polyalkenoic acid conditioned group.
170 It is possible that there was an ultra-thin demineralized layer at the interface which could not
171 be seen in these TEM photomicrographs. There was again no clear sign of degradation
172 between the 1 week and 1 year specimens (Figure 3a and Figure 3b).

173 From the chemical element analysis, 3 areas were chosen, which were estimated areas: De
174 (Plot 1), IL (Plot 2) and ML (Plot 3) areas. Basically, the composition of GIC includes a
175 polymeric water-soluble acid, glass, and water [29]. The glass components were either of the
176 $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaF}_2$ system or the more complex $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-P}_2\text{O}_5\text{-CaO-CaF}_2$ system, also
177 calcium has been substituted by strontium [30]. The components of the GIC material such as
178 Si, Al, Sr, Ca were detected from all 3 plots, indicating the presence of GIC components in
179 the IL and ML. The ML appears to be a mixture of GIC and dentin tissue, which has been
180 unknown until now. The IL is a reaction layer which is probably formed by polyalkenoic acid
181 and HAp. Due to the ionic exchange of fluorine and strontium, GIC has a remineralization
182 effect on demineralized tooth in terms of quantitative analysis of the mineral content of the
183 remineralized structures, and their mechanical properties were previously described [31-35].

184 Partial caries removal or incomplete caries removal is more demanding based on scientific
185 evidence [36-38]. For those situations, using the stepwise removal and selective removal
186 technique, GIC is recommended as it has similar bond strength to both normal and caries-
187 affected dentin [39,40]. GIC has superior clinical survival results for deep dentin and hyper-
188 mineralized dentin as well [18,41]. This is because of its resilience, low polymerization
189 shrinkage and good sealing ability.

190 From the results of the μ TBS test, pre-treatment of dentin using a polyalkenoic acid
191 conditioner did not affect the long-term durability of a conventional GIC; hence, the null
192 hypothesis should be accepted.

193 Further research will provide an understanding of the remineralizing effect of GIC on caries-
194 affected dentin using polyalkenoic acid.

195

196 **5. CONCLUSION**

197 Aging did not reduce the bond strength of the conventional GIC to dentin whether the surface
198 was pre-treated with a polyalkenoic acid conditioner or not. Conditioning of dentin appears to
199 increase the durability of the GIC to dentin.

200

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202 commercial, or not-for-profit sectors.

203

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Legends to figures

Figure 1: Micro-tensile bond strength of GIC bonded to polyalkenoic acid conditioned (Cavity Conditioner) and non-conditioned dentin for 1 week and 1 year. Mean μ TBS are presented in MPa. $n=10$. The same letters indicate no statistically significant difference ($p>0.05$).

Figure 2: Representative TEM photomicrographs of unstained, non-demineralized sections of the GIC/dentin interface with polyalkenoic acid conditioning using Cavity Conditioner stored for 1 week and 1 year. (a,b) A partially demineralized dentin layer (De) of about 0.5-1 μ m was seen at the dentin-conditioned interface (a,b). Hydroxyapatite (HAp) remained within this partially demineralized layer. On top of the partially demineralized layer, a matrix-rich layer (ML) of a width of a few hundred nanometers at 1 week and about 100 nanometers at 1 year was seen (a,b). On top of the matrix-rich layer, an intermediate layer (IL) of a few hundred nanometers was deposited, and this zone was typically demarcated from the GIC matrix by small electron-lucent globules (a,b). [GI = Glass ionomer cement; IL = Intermediate Layer; ML = Matrix-rich Layer; De = Demineralized Layer; Ud = Unaffected dentin.]

Figure 3: Representative TEM photomicrographs of unstained, non-demineralized sections of the GIC/dentin interface without polyalkenoic acid conditioning stored for 1 week and 1 year (a,b). The GIC material was closely attached to the dentin surface without a smear layer and no other layer could be detected (a,b). The bond appeared intact. There were no clear signs of bond degradation after 1 year of water storage. [GI = Glass ionomer cement; Ud = Unaffected dentin.]

Figure 4: The image of GIC/dentin interface with polyalkenoic acid conditioning after 1 week storage as captured by FE-TEM. Plot 1 indicated the GIC area, Plot 2 indicated the IL area and Plot 3 indicated the ML area.

Figure 5: Chemical compositions were analyzed by STEM mode. Plots 1 and 2 showed the components of GIC such as Si, Sr, Al. Plots 1, 2 and 3 showed almost the same tendency while Plot 3 showed more Ca content.

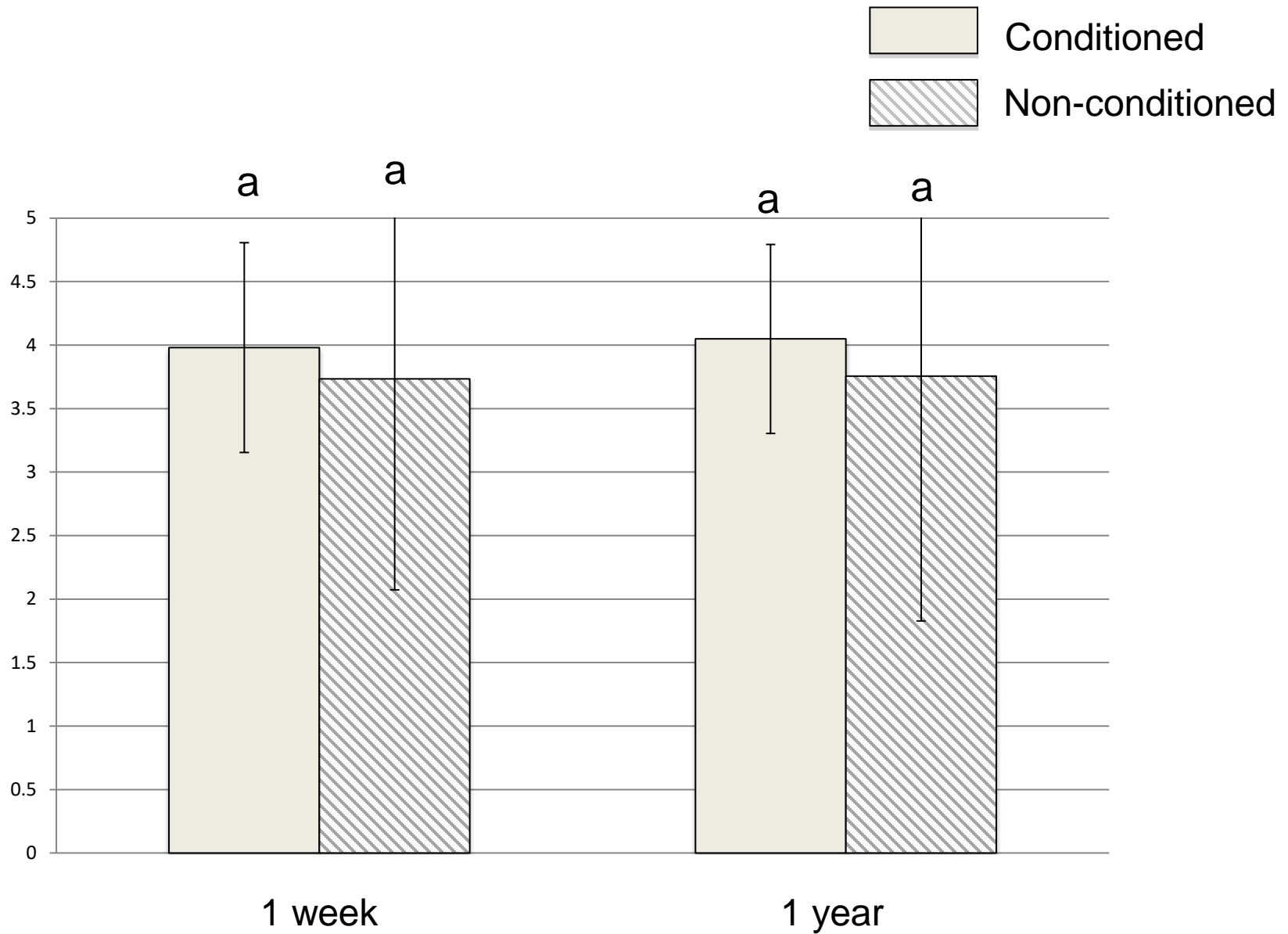
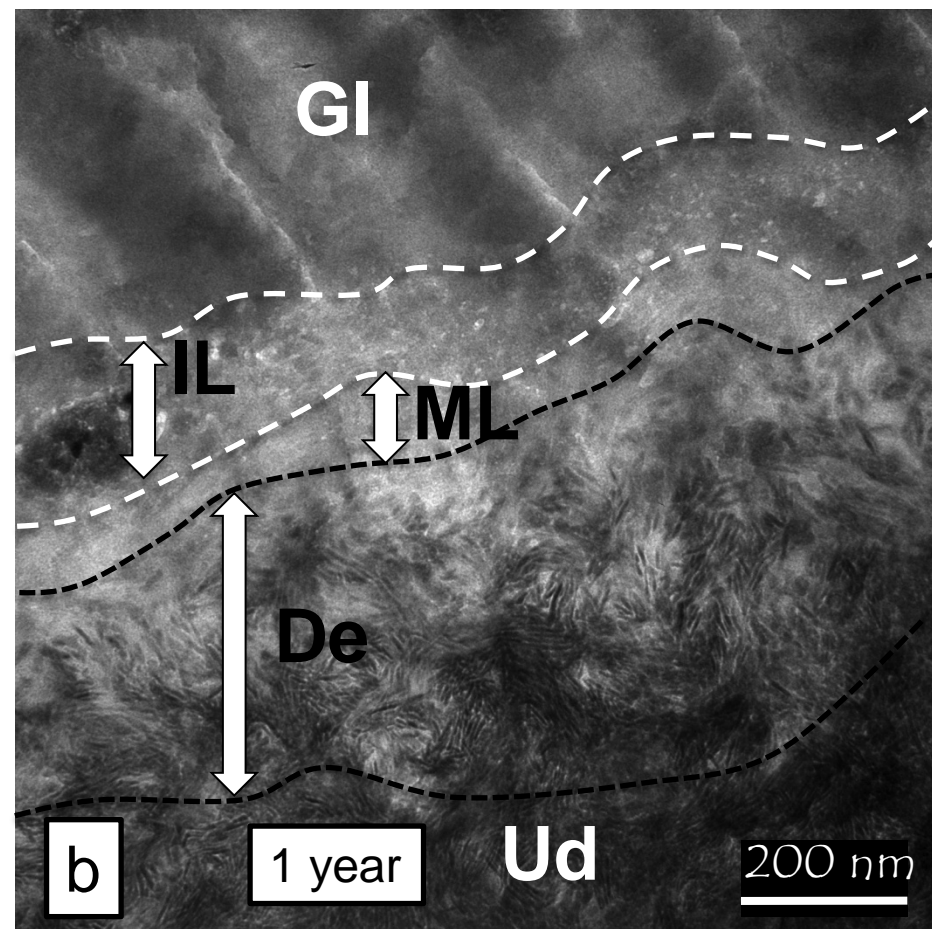
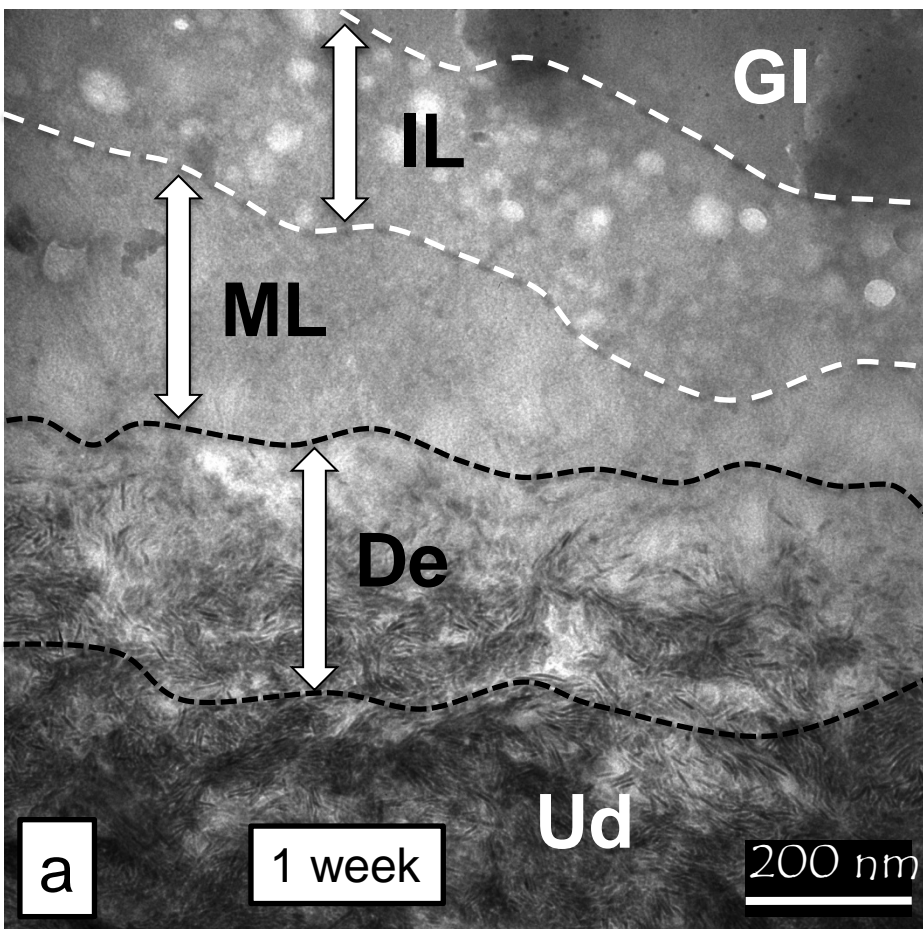
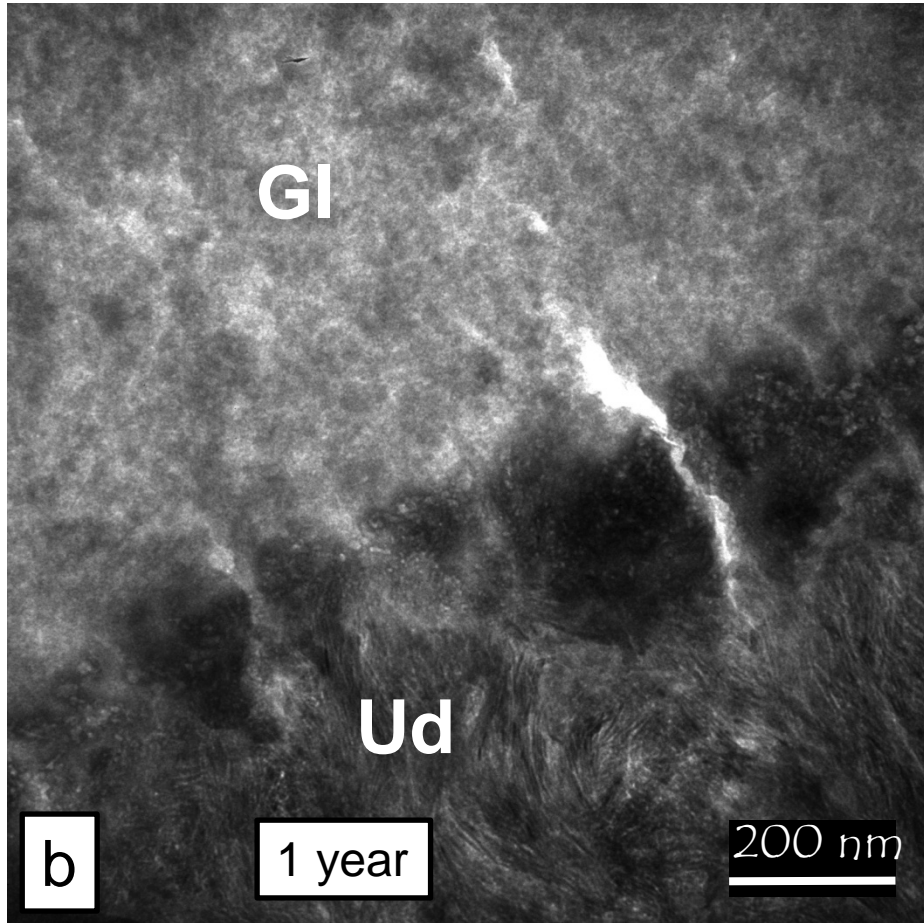
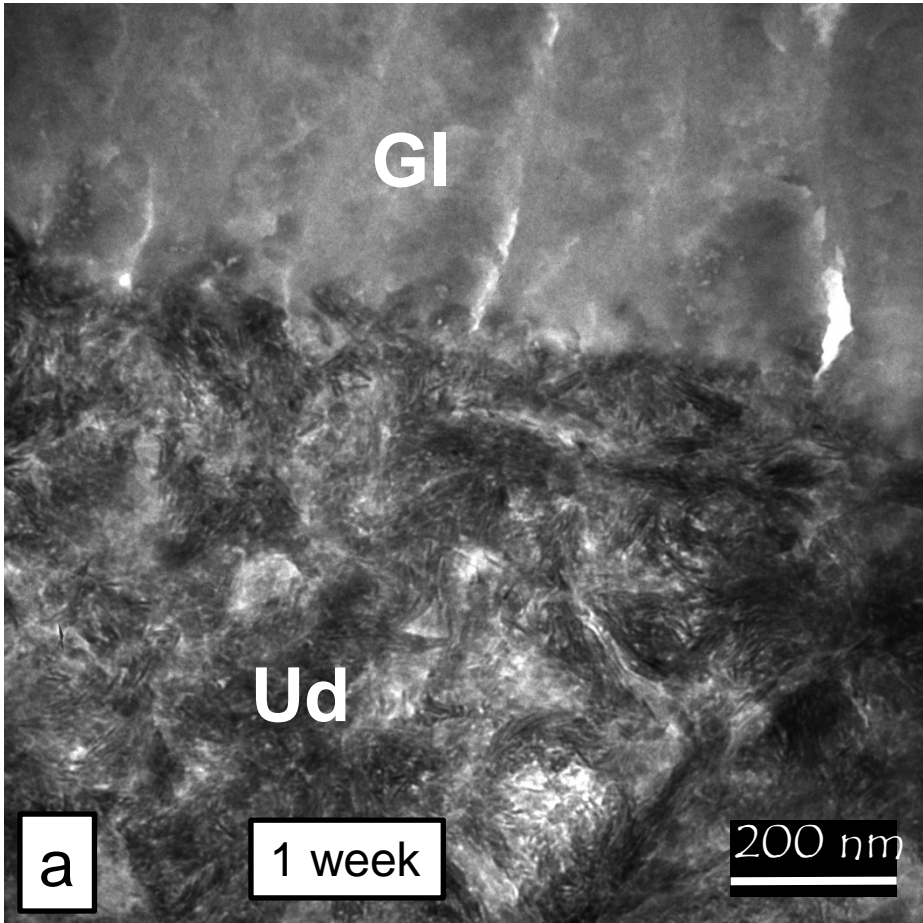


Fig 1



GI: Glass ionomer cement
IL: Intermediate Layer
ML: Matrix-rich Layer
De: Demineralized Layer
Ud: Unaffected Dentin

Fig 2



GI: Glass ionomer cement
Ud: Unaffected Dentin

Fig 3

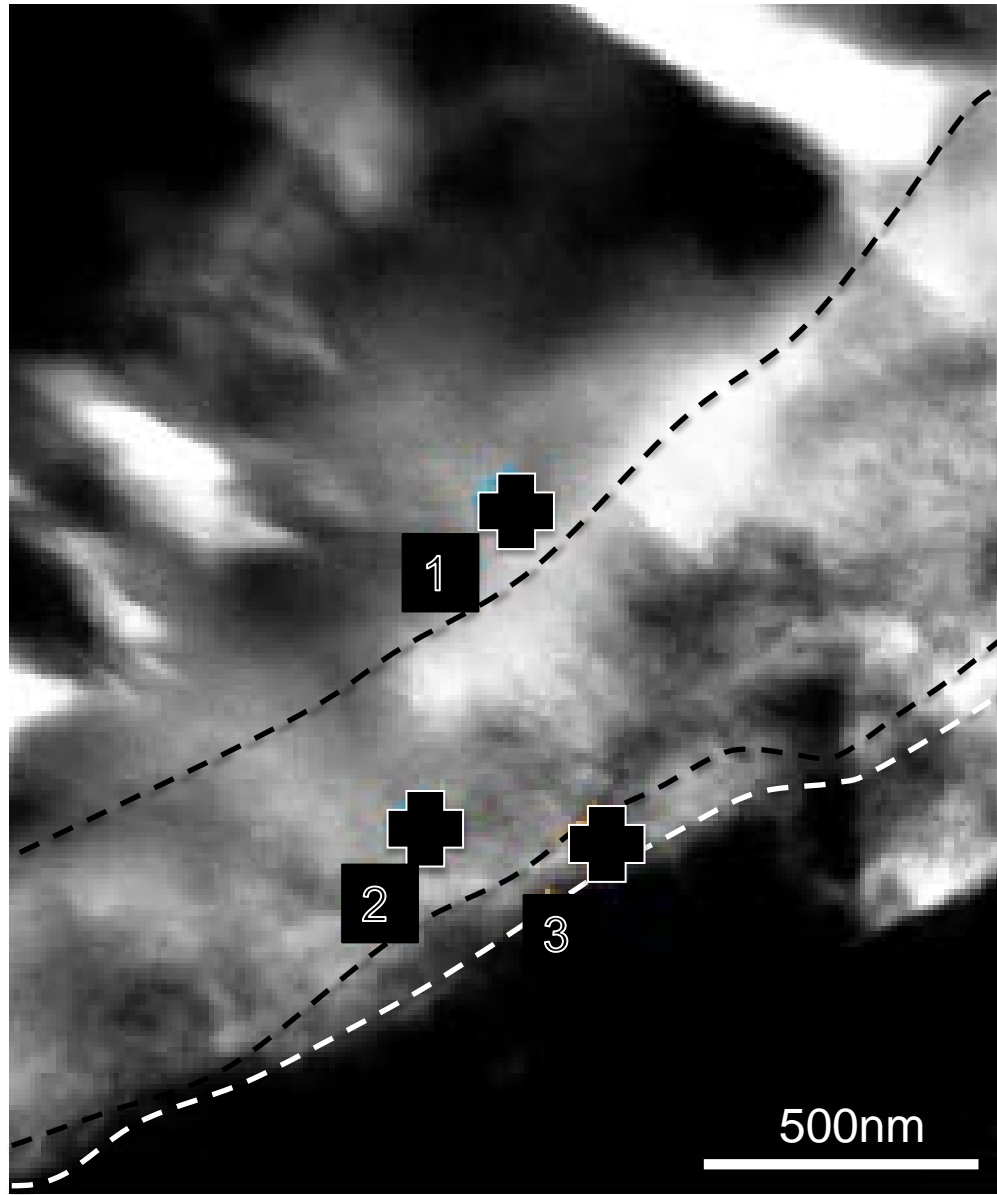


Fig 4

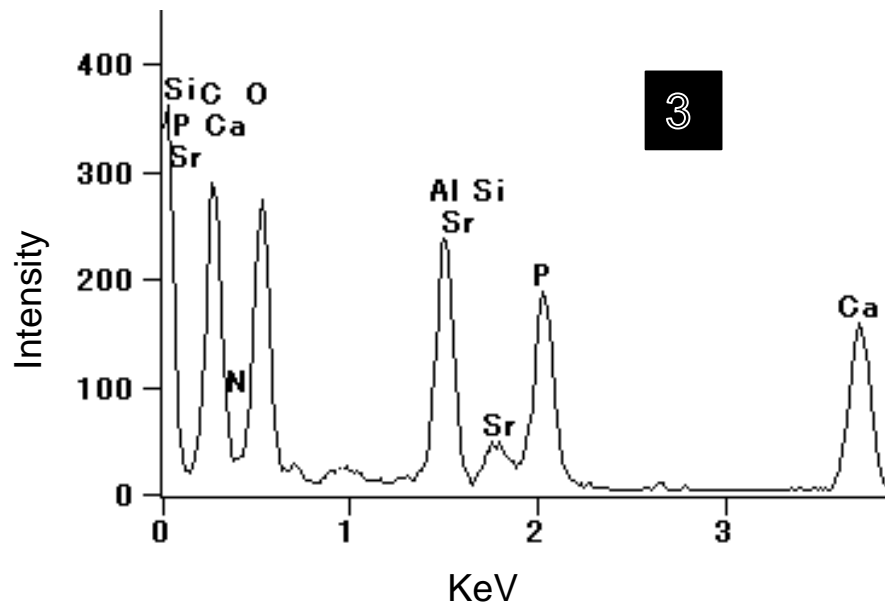
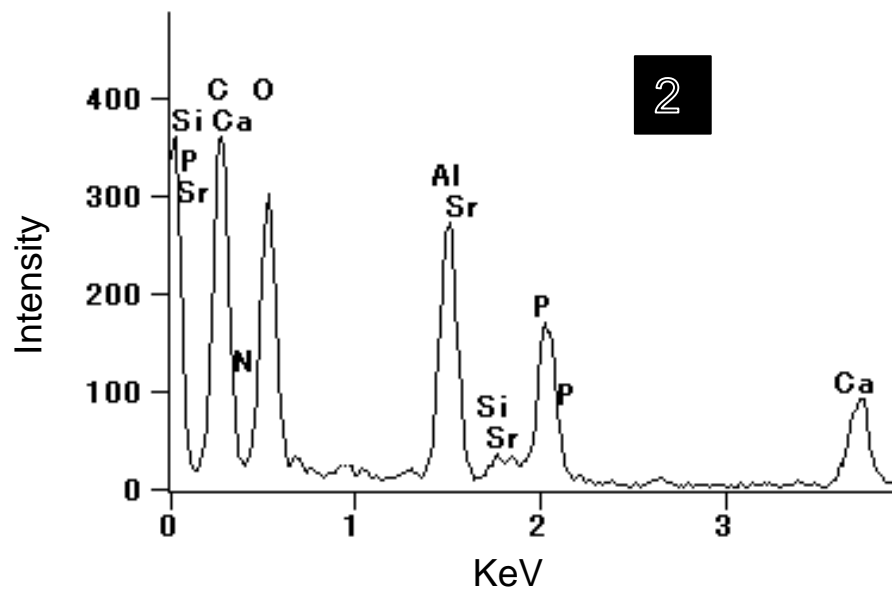
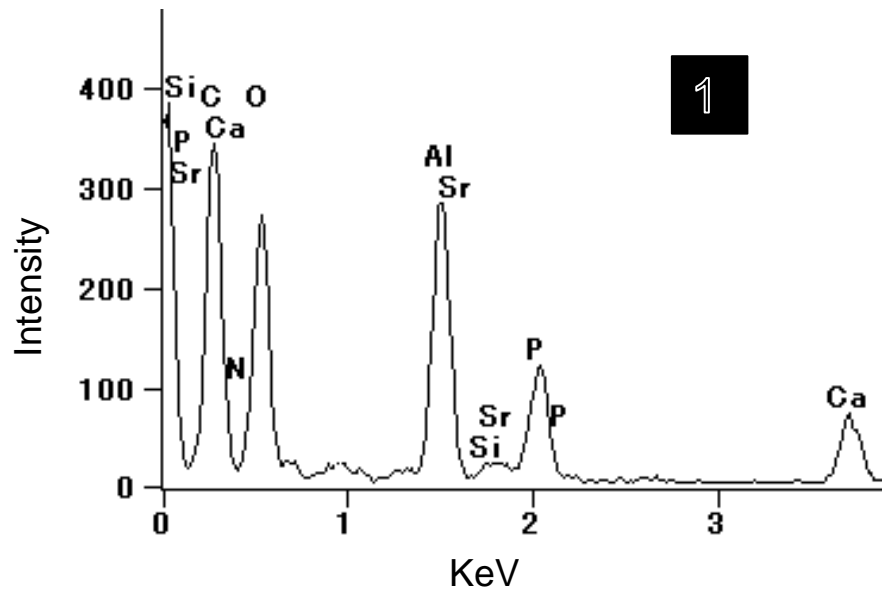


Fig 5