SUMMARY

Bonding to tooth tissue can be achieved through an “etch&rinse,” “self-etch” or “glass-ionomer” approach. In this paper, the basic bonding mechanism to enamel and dentin of these three approaches is demonstrated by means of ultramorphological and chemical characterization of tooth-biomaterial interface interactions. Furthermore, bond-strength testing and measurement of marginal-sealing effectiveness (the two most commonly employed methodologies to determine “bonding effectiveness” in the laboratory) are evaluated upon their value and relevance in predicting clinical performance. A new dynamic methodology to test biomaterial-tooth bonds in a fatigue mode is introduced with a recently developed micro-rotary fatigue-testing device. Eventually, today’s adhesives will be critically weighted upon their performance in diverse laboratory studies and clinical trials. Special attention has been given to the benefits/drawbacks of an etch&rinse versus a self-etch approach and the long-term performance of these adhesives. Correlating data gathered in the laboratory with clinical results clearly showed that laboratory research CAN predict clinical effectiveness. Although there is a tendency to simplify bonding procedures, the data presented confirm that conventional three-step etch&rinse adhesives still perform most
favorably and are most reliable in the long-term. Nevertheless, a self-etch approach may have the best future perspective. Clinically, when adhesives no longer require an “etch&rinse” step, the application time, and probably more importantly, the technique-sensitivity are substantially reduced. Especially “mild,” two-step self-etch adhesives that bond through a combined micromechanical and chemical interaction with tooth tissue closely approach conventional three-step systems in bonding performance.

INTRODUCTION

Adhesive dentistry evolves rapidly. Two main incentives drive this evolution. Adhesive techniques combined with using tooth-colored restorative materials are frequently requested by patients. They want us to restore their teeth not only anatomically and functionally, but also esthetically and, thus, nearly invisibly. From our perspective, today’s operative dentistry should primarily involve “minimally invasive” (Degrange & Roulet, 1997) or “minimum intervention” (Tyas, Anusavice & Frencken, 2000) care. This means that only the lost or diseased tooth tissue is replaced by the restorative material that is directly bonded to the remaining sound tissue. Also, the more recent approach of promoting “maintenance and repair” (Bouschlicher, Reinhardt & Vargas, 1997; Denhey, Bouschlicher & Vargas, 1998; Roetters, 2000; Wilson, Setcos & Brunton, 2001), rather than replacing entire restorations (exhibiting marginal discolorations and/or defects) has further boosted the use of adhesive techniques in diverse applications of everyday clinical practice.

Major drawbacks of this approach are that adhesive techniques still accompany a higher placement complexity and technique sensitivity (risk of making manipulation errors). Also, though the retention of adhesive restorations for a reasonable time is no longer a clinical problem, maintaining the margins of adhesive restorations sealed against leakage phenomena remains the major factor that shortens clinical longevity.

The fundamental principle of adhesion to tooth substrate is based upon an exchange process by which inorganic tooth material is exchanged for synthetic resin (Van Meerbeek & others, 2001a). This process involves two phases. One phase consists of removing calcium phosphates by which microporosities are exposed at both the enamel and dentin tooth surface. The other so-called hybridization phase involves infiltration and subsequent in situ polymerization of resin within the created surface microporosities. This results in micro-mechanical interlocking that is primarily based on mechanisms of diffusion. While micro-mechanical interlocking is believed to be a prerequisite to achieving good bonding within clinical circumstances, the potential benefit of additional chemical interaction between functional monomers and tooth substrate components has recently gained new attention. This paper sketches the current status of adhesives in terms of bonding effectiveness measured in the laboratory and in clinical practice. Special attention is given to the potential roles of both micro-mechanical and chemical bonding mechanisms through correlating morphologic and chemical interfacial characteristics of tooth-biomaterial interactions using diverse kinds of adhesives.

MECHANISMS OF ADHESION

TO ENAMEL AND DENTIN

Using contemporary adhesives, the substance exchange between biomaterial and tooth tissue is carried out in one, two or three clinical application steps, respectively. Besides the number of application steps, adhesives can further be classified based on the underlying adhesion strategy in “etch&rinse,” “self-etch” and “(resin-modified) glass-ionomer adhesives” (Van Meerbeek & others, 2001a) (Figure 1). The degree of substance exchange substantially differs among these adhesives. In general, the exchange intensity induced by etch&rinse adhesives exceeds that of self-etch adhesives, though among the latter, systems that rather intensively interact with tooth tissue also exist, even when applied in only a single step.

Etch&Rinse Approach

This adhesion strategy involves at least two steps and, in its most conventional form, three steps with successive application of the conditioner or acid etchant, followed by the primer or adhesion promoting agent, and eventually, application of the actual bonding agent or adhesive resin (Figure 1). The simplified two-step version combines the second and third step but still follows a separate “etch&rinse” phase.

This etch&rinse technique is still the most effective approach to achieving efficient and stable bonding to enamel and basically only requires two steps. Selective dissolution of hydroxyapatite crystals through etching (commonly with a 30-40% phosphoric-acid gel) is followed by in situ polymerization of resin that is readily absorbed by capillary attraction within the created etch pits, thereby, enveloping individually exposed hydroxyapatite crystals (Figure 2). Two types of resin tags interlock within the etch-pits. “Macro”-tags fill the space surrounding the enamel prisms (Figure 2a), while numerous “micro”-tags result from resin infiltration/polymerization within the tiny etch-pits at the cores of the etched enamel prisms (Figure 2b). The latter are especially thought to contribute the most with regard to retention to enamel.

At dentin, this phosphoric-acid treatment exposes a microporous network of collagen that is nearly totally deprived of hydroxyapatite (Figures 3 and 4). High-resolution transmission electron microscopy (TEM) and
chemical surface analysis by energy dispersive X-ray spectroscopy (EDXS) and X-ray photoelectron spectroscopy (XPS) have confirmed that nearly all calcium phosphates were removed or at least became under detection limit (Figure 5) (Van Meerbeek & others, 1996; Yoshida & Van Meerbeek, 2002). As a result, the primary bonding mechanism of etch&rinse adhesives to dentin is primarily diffusion-based and depends on hybridization or infiltration of resin within the exposed collagen fibril scaffold, which should be as complete as possible (Figure 6). True chemical bonding is rather unlikely, because the functional groups of monomers may have only weak affinity to the "hydroxyapatite-depleted" collagen. Such challenging monomer-collagen interaction might be the principle reason for what has been documented as manifesting in the form of "nanoleakage" phenomena (Sano & others, 1994b, 1995).

Most critical in the etch&rinse approach is the priming step. When an acetone-based adhesive is used, the highly technique-sensitive "wet-bonding" technique is mandatory (Tay & others, 1996). Otherwise, gentle post-conditioning air-drying of acid-etch dentin (and enamel) following a "dry-bonding" technique still guarantees effective bonding when a water/ethanol-based adhesive is used (Van Meerbeek & others, 1996, 1998c).
Glass-Ionomer Approach

Glass-ionomers remain as the only materials that are self-adhesive to tooth tissue, in principle, without any surface pre-treatment (Figure 7). Although this is certainly true, pre-treatment with a weak polyalkenoic-acid conditioner significantly improves bonding efficiency (Inoue & others, 2001a). Hence, this glass-ionomer approach can be achieved following a one- or two-step application procedure (Figure 1). The additional conditioning step becomes more important, especially when coarse cutting diamonds are used and, consequently, thicker and more compact smear layers are produced. In general, such a polyalkenoic-acid conditioner is applied for 10-to-20 seconds and gently rinsed off, followed by gently air-drying without dehydrating the surface (Figures 7 and 8a). The increase in bonding efficiency must be partially attributed to (1) a “cleaning” effect, by which loose cutting debris is removed, (2) a partial “demineralization” effect, by which the surface area is increased and microporosities for micromechanical interlocking or hybridization are exposed, but also in part to (3) chemical interaction of polyalkenoic acid with residual hydroxyapatite (see below). A network of “hydroxyapatite-coated” collagen fibrils interspersed by pores is typically exposed to a depth no deeper than 1 µm. TEM and XPS have demonstrated that (depending on the product) this polyalkenoic acid conditioner cannot be completely rinsed off (Van Meerbeek & others, 1998b, 2001b). An up to 0.5 µm thick layer, often referred to as “gel phase,” remains attached to the tooth surface despite the conditioner being rinsed off (Figure 8b).

The actual auto-adhesion of glass ionomers to tooth tissue has recently been determined to be twofold. Micromechanical interlocking is achieved by shallow hybridization of the micro-porous, hydroxyapatite-coated collagen fibril network (Figure 8) (Van Meerbeek & others, 1998b, 2001b; Tay & others, 2001; Yip & others, 2001). In this respect, glass ionomers can be considered as adhering to tooth tissue through a “mild” self-etch approach (see below). The basic difference with the resin-based self-etch approach is that glass ionomers are self-etching through the use of a relatively high molecular weight (8,000-15,000) polycarboxyl-based polymer. Resin-based self-etch adhesives make use of acidic low-molecular weight monomers.

As the second component of the self-adhesion mechanism, true primary chemical bonding occurs through
forming ionic bonds between the carboxyl groups of the polyalkenoic acid and calcium of hydroxyapatite that remains around the exposed surface collagen (Figures 9-11). This was proven for polyalkenoic acids applied to hydroxyapatite (Yoshida & others, 2000). Interaction of the polyalkenoic acid with hydroxyapatite resulted in a significant shift of the peak representing the carboxyl groups (-COO-) to a lower binding energy, suggesting the formation of an ionic bond to hydroxyapatite as schematically explained in Figure 10.

This interaction was relatively strong, as this peak shift was recorded after ultrasonically rinsing off the polyalkenoic acid solution. This shifted peak at the XPS spectrum in Figure 9 represents the binding ener-
gy of the C atom (C 1s) of the carboxyl group, being 288.6 eV for the unreacted polyalkenoic acid itself. This binding energy results from two oxygen atoms that pull on the carbon atom. As explained in Figure 10, when one oxygen atom of the carboxyl functional group of the polyalkenoic acid reacts chemically with calcium of hydroxyapatite, it consumes energy to form an ionic bond. Consequently, its pull to the carbon atom of the carboxyl group is less intense, thus, reducing its binding energy to 288.2 eV. However, the carboxyl peak in Figure 9 did not shift entirely to 288.2 eV, indicating that not all carboxyl groups interacted with hydroxyapatite. In fact, deconvolution disclosed that the shifted peak consists of two sub-peaks (Figure 11), representing carboxyl groups that interacted with hydroxyapatite (sub-peak at 288.2 eV) and those that did not (sub-peak at 288.6 eV). It was also demonstrated that the actual molecular formula of the polyalkenoic acid significantly influences the chemical bonding potential (Yoshida & others, 2000; Fukuda & others, 2003). XPS clearly showed that a polyalkenoic acid based upon 10:1 acrylic/maleic acid units has about two-thirds of its carboxyl groups bonded to hydroxyapatite versus only half of the carboxyl groups of pure polyacrylic acid (Yoshida & others, 2000; Fukuda & others, 2003). Based on these XPS data.
Yoshida & others, 2000, 2001; Yoshioka & others, 2002), the authors proposed an “Adhesion-Decalcification model” (AD-model) that explains why certain acids adhere to tooth tissue more than they decalcify it (Figure 12). This largely depends on the solubility of the formed calcium salt at the hydroxyapatite surface in its own acidic solution. The more soluble the calcium salts of the acids (or the adhesive monomer/polymer), the less it will adhere to the mineral substrate. As the calcium salts of polyalkenoic acids could hardly be dissolved, they have an adequate chemical bonding potential to hydroxyapatite-based tissues.

Figure 15. Fe-SEM photomicrographs of dentin either treated (left/a) with the strong self-etching primer Non-Rinse Conditioner (Dentsply) or (right/b, image taken by J Perdigão) with the mild self-etching primer of Clearfil Liner Bond 2 (Kuraray). Non-Rinse Conditioner clearly opened the dentin tubules and exposed a micro-porous collagen fibril network similar to the effect of an etch&rinse approach using phosphoric acid. However, Clearfil Liner Bond 2 primer interacted clearly less intense with some exposure of collagen, while most tubules remained occluded.

Figure 16. TEM photomicrographs of an unstained non-demineralized (left/a) and stained demineralized (right/b) section through the resin-dentin interface produced by the strong one-step self-etch adhesive Adper Prompt (3M ESPE). Note that dentin was rather deeply demineralized up to about 3 µm. All hydroxyapatite around collagen was dissolved and the demineralization front stopped abruptly. A rather thick hybrid layer of about 3 µm was formed and resembles a hybrid layer as it would typically be produced following a etch&rinse approach. The typical phosphate-based composition of the adhesive resulted in a strong pick-up of heavy metal stain, by which the infiltration of the electron-dense resin within the hydroxyapatite-depleted collagen can be clearly detected. Some phase separation between electron lucent hydrophilic and electron dense hydrophobic adhesive components can be observed within the adhesive resin layer on top of the hybrid layer.

Figure 17. (a) TEM photomicrograph (left) of an unstained, non-demineralized section through the resin-dentin interface produced by Clearfil SE (Kuraray). Note that dentin was only partially demineralized for about 1 µm deep, leaving hydroxyapatite crystals within the hybrid layer. (b) TEM photomicrograph of a stained, demineralized section through the resin-dentin interface produced by Clearfil SE (Kuraray). Note the formation of a 1-µm thick hybrid layer with a typical shag-carpet appearance at the transition to the adhesive resin and individual cross-banded collagen fibrils separated by electron lucent interfibrillar spaces. The chemical formula of the functional monomer 10-MDP is presented in the insert.

Figure 18. TEM photomicrographs of the resin-dentin interface produced by the “intermediary strong” self-etch adhesive AdheSE (Vivadent). A relatively thick hybrid layer of about 2 µm can be observed on the photomicrograph representing a stained, demineralized section. The insert shows an unstained, non-demineralized TEM section, on which can be seen that the top 1.5-2 µm of the hybrid layer does not contain any residual hydroxyapatite crystals. The 0.5-1 µm layer at the hybrid layer base still contains residual hydroxyapatite and forms a rather gradual transition to the underlying affected dentin.
Typical of some glass ionomers is the morphologic manifestation of a “gel-phase” at the interface, as was shown correlative by transmission electron microscopy (Figure 8) and atomic force microscopy (Van Meerbeek & others, 1998b, 2001b; Yoshida & others, 1999). Correlating TEM and XPS data elucidated that this gel phase represents the formation of a calcium polycarboxylate salt resulting from either the polyalkenoic acid conditioner or the glass ionomer material itself (Van Meerbeek & others, 2001b). This phase has been shown to be stable and strong, intermediary between the shallow 0.5-1 µm hybrid layer and the glass-ionomer matrix. In microtensile bond strength testing, the interface typically fractured well above the gel phase within the matrix of the glass-ionomer material itself (Van Meerbeek & others, 2001b). This actual function and contribution of this phase to the bond integrity needs to be further elucidated.

Self-Etch Approach

Probably, in regard to user-friendliness and technique-sensitivity, clinically, the most promising approach is self-etch. It no longer needs an “etch&rinse” phase, which not only lessens clinical application time, but also significantly reduces technique-sensitivity or the risk of making errors during application and manipulation. Another important advantage of the self-etch approach is that infiltration of resin occurs simultaneously with the self-etching process, by which the risk of discrepancy between both processes is low or non-existent. However, little is known about the long-term effects of incorporating dissolved hydroxyapatite crystals and residual smear layer remnants within the bond. How much of the primer/adhesive solvent is kept within the interfacial structure should also be investigated. Such solvent surplus will directly weaken the bond integrity, provide channels for nanoleakage or may affect polymerization of the infiltrated monomers. The resultant interfacial structure also becomes more hydrophilic and, thus, more prone to hydrolytic degradation (Tay & others, 2002a; Tay, Pashley & Yoshiyama, 2002b).

A self-etch approach involves either a two- or one-step application procedure (Figure 1). The self-etch effect should be ascribed to monomers to which one or more carboxylic or phosphate acid groups are grafted (Van Meerbeek & others, 2001a). Depending on etching aggressiveness, they can be subdivided into “strong” and “mild” self-etch adhesives (Figure 13).

“Strong” self-etch adhesives usually have a pH of 1 or below (Table 1). This high acidity results in rather deep demineralization effects. At enamel, the resulting acid-etch pattern resembles a phosphoric-acid treatment following an etch&rinse approach (Figure 14a) (Inoue & others, 2000; Pashley & Tay, 2001). At dentin, collagen is exposed and nearly all hydroxyapatite is dissolved (Figures 15a and 16). Consequently, the underlying bonding mechanism of “strong” self-etch adhesives is primarily diffusion-based, similar to the etch&rinse approach. Such low-pH self-etch adhesives have often been documented with rather low bond strength values, especially at dentin, and quite a high...
The acidity (pH) of diverse adhesive solutions

<table>
<thead>
<tr>
<th>Adhesive</th>
<th>Classification</th>
<th>pH primer*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adper Prompt L-Pop (3M ESPE)</td>
<td>One-step self-etch</td>
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</tr>
<tr>
<td>Prompt L-Pop 2 (3M ESPE)</td>
<td>One-step self-etch</td>
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</tr>
<tr>
<td>Xeno III (Dentsply)</td>
<td>One-step self-etch</td>
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<tr>
<td>i-Bond (Kulzer)</td>
<td>One-step self-etch</td>
<td>1.6</td>
</tr>
<tr>
<td>Non-Rinse Conditioner (Dentsply)</td>
<td>Two-step self-etch</td>
<td>1.0</td>
</tr>
<tr>
<td>AdheSE primer (Vivadent)</td>
<td>Two-step self-etch</td>
<td>1.4</td>
</tr>
<tr>
<td>OptiBond Solo Plus SE primer (Kerr)</td>
<td>Two-step self-etch</td>
<td>1.5</td>
</tr>
<tr>
<td>Clearfil SE Bond primer (Kuraray)</td>
<td>Two-step self-etch</td>
<td>1.9</td>
</tr>
<tr>
<td>Clearfil SE Bond Plus primer (Kuraray)</td>
<td>Two-step self-etch</td>
<td>2.0</td>
</tr>
<tr>
<td>Unifil Bond primer (GC)</td>
<td>Two-step self-etch</td>
<td>2.2</td>
</tr>
<tr>
<td>Panavia ED primer mixed (Kuraray)</td>
<td>Two-step self-etch</td>
<td>2.6</td>
</tr>
<tr>
<td>OptiBond Solo Plus primer/adhesive (Kerr)</td>
<td>Two-step etch&amp;rinse</td>
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</tr>
<tr>
<td>Prime&amp;Bond NT primer/adhesive (Dentsply)</td>
<td>Two-step etch&amp;rinse</td>
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</tr>
<tr>
<td>Scotchbond 1 primer/adhesive (3M)</td>
<td>Two-step etch&amp;rinse</td>
<td>4.7</td>
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<tr>
<td>OptiBond FL primer (Kerr)</td>
<td>Three-step etch-rinse</td>
<td>1.8</td>
</tr>
</tbody>
</table>

* Measured in-house using a digital pH meter (Inolab pH Level 2, WTW, Weilheim, Germany).

number of pre-testing failures when tested following a microtensile bond strength approach (Inoue & others, 2001b, 2003; De Munck & others, 2003a). Besides the high initial acidity that appears to dramatically weaken the bonding performance, another concern is the effect of residual solvent (water) that remains within the adhesive interface, which can hardly be completely removed. Further study needs to investigate the long-term stability of this strong self-etch approach.

“Mild” self-etch systems, in general, have a pH of around 2 (Table 1) and demineralize dentin only to a depth of 1 µm (Figures 15b and 17). This superficial demineralization occurs only partially, keeping residual hydroxyapatite still attached to collagen. Nevertheless, sufficient surface-porosity is created to obtain micromechanical interlocking through hybridization. The thickness of the hybrid layer is, however, much smaller than that produced by the strong self-etch or etch&rinse approach but has been proven to be minor in importance with regard to actual bonding effectiveness (Inoue & others, 2000, 2001b; De Munck & others, 2003a). The preservation of hydroxyapatite within the submicron hybrid layer may serve as a receptor for additional chemical bonding (Van Meerbeek & others, 2000; Yoshida & others, 2003). Carboxylic acid-based monomers like 4-MET (4-methacryloyloxyethyl trimellitic acid) and phosphate-based monomers, such as phenyl-P (2-methacryloyloxyethyl phenyl hydrogen phosphate), and 10-MDP (10-methacryloxydecyl dihydrogen phosphate) have a chemical bonding potential to calcium of residual hydroxyapatite (Yoshida & others, 2003). One may hypothesize that a weak self-etching effect is mandatory in order to (1) deal with the smear layer resulting from cavity preparation, (2) achieve micromechanical interlocking within etch pits at enamel and (3) achieve shallow micromechanical interlocking through hybridization at dentin. Micromechanical retention is thought to be necessary to resist acute de-bonding forces (such as those to which composite-tooth bonds are typically subjected during bond-strength testing). In addition, the exposed hydroxyapatite enamel surface and the hydroxyapatite crystals that remain around collagen (in the case of a “mild” self-etching or a glass-ionomer approach) are expected to be particularly advantageous. They enable more intimate chemical interaction with the functional monomers on a molecular level and may help prevent/retard marginal leakage. The challenge now is to have the functional monomers interact with hydroxyapatite so that the resulting calcium-carboxylate or calcium-phosphate bonds are stable within a hydrophilic environment long-term. Keeping hydroxyapatite around collagen may also better protect the collagen against hydrolysis and, thus, early degradation of the bond (Sano & others, 1999; Hashimoto & others, 2000, 2002). The weakest property of mild self-etch adhesives is their bonding potential to enamel. Therefore, developing monomers with stronger chemical bonding potential to hydroxyapatite may also help to further improve their bonding performance to enamel.

Some new adhesives, AdheSE (Vivadent) and OptiBond Solo Plus Self-etch (Kerr, Orange, CA, USA), were recently marketed and cannot be classified as “mild” or “strong” two-step self-etching adhesives. The pH of their self-etching primers is about 1.5 (Table 1) and, based on their interaction with dentin, the authors refer to them as "intermediary strong" two-step self-etch adhesives. Most typical is the two-fold build-up of the dentinal hybrid layer with a completely demineralized top layer and a partially demineralized base (Figures 18 and 19). Following an “etch&rinse” or “strong” self-etch approach, the transition of the exposed collagen fibril network to the underlying unaffected dentin is quite abrupt (see Figures 4, 5 and 16, respectively). Following an “intermediary strong” self-etch approach, the deepest region of the hybrid layer up to a maximum of 1 µm still con-
tains hydroxyapatite, by which the transition of the hybrid layer to the underlying unaffected dentin is more gradual (Figures 18 and 19). These adhesives are more acidic than the “mild” self-etch adhesives, by which better micromechanical interlocking is achieved at enamel and dentin. The residual hydroxyapatite at the hybrid layer base may still allow for chemical intermolecular interaction, as was shown before for the “mild” self-etch adhesives. Based on the acidity (Table 1), the one-step self-etch adhesives i-Bond (Kulzer) and Xeno III (Dentsply, Milford, DE, USA) must also categorized as “intermediary strong” self-etch adhesives. Their resultant interfacial interaction is consequently expected to be similar to that produced by the intermediary “strong” two-step self-etch adhesives documented above.

Unicem (3M ESPE, St Paul, MN, USA) was recently launched as a possible first step towards self-adhesive resin-based restorative materials. This luting material is designed to be applied without any pre-treatment. TEM of the resultant interface showed a very superficial interaction with dentin (Figure 20). When applied to bur-cut dentin, a layer about 0.5-1 µm deep appeared less mineralized and most likely represented infiltration of Unicem components with a partially dissolved bur smear layer. This layer did not appear when Unicem was applied to fractured dentin that was free of cutting smear. Then, the interaction of Unicem with dentin could barely be morphologically detected. The actual bonding mechanism of this self-adhesive cement should be investigated in depth.

MEASURING BONDING EFFECTIVENESS:
LABORATORY VERSUS CLINICAL TESTING

Laboratory Testing of Adhesives: Can They Predict Clinical Effectiveness?

Clinical trials are the ultimate test for dental restorations, but they cannot differentiate the true reason for failure due to the simultaneous impact of diverse stresses on restorations within the aggressive oral cavity. Lab testing can evaluate the effect of a single variable, while keeping all other variables constant. Based on this type of research, clear recommendations can be formulated toward clinicians with regard to the appropriate use and selection of dental materials. In general, laboratory testing is easy, fast and relatively cheap to screen new materials/techniques. They are useful in determining the “effectiveness” of adhesive materials within the specific test set-up. Ideally, the final objective should always be predicting clinical behavior long-term, though direct translation to the clinical situation is often difficult or even impossible.

Bond Strength Testing

In the mouth, the interface between restoration and tooth is exposed to diverse forces that act simultaneously. Already during setting of composite, resin shrinkage puts stress on the bond, pulling it away from the cavity wall (Versluis, Tantbirojn & Douglas, 1998). During function, mechanical stress by chewing forces, thermal and chemical stress with changes in temperature and pH will have an effect on the bond integrity as part of bio-tribocorrosive effects. The rationale behind bond strength testing is that the higher the actual bonding capacity of an adhesive, the better it will withstand such stresses and the longer the restoration will survive in vivo. Bond strength testing is relatively easy and fast and, in fact, besides a material tester does not require special equipment. It, therefore, remains the most popular methodology for measuring bonding effectiveness in the laboratory. Van Noort & others (1989), however, emphasized that bond strength cannot be regarded as a material property. The data obtained from bond strength tests largely depend on the actual test set-ups that may differ between laboratories for parameters such as specimen geometry, size of surface area, the type of composite and more. It is, therefore, not surprising that bond strength data substantially vary among laboratories throughout the world. The many variables involved make standardization of test methodologies for bond-strength measurements hardly achievable.

Most commonly, bond strength is measured by subjecting composites bonded-to-enamel/dentin to tensile or shear stress. However, at bond strength values higher than 20 MPa in a shear test, cohesive failures of the substrate will more likely occur (Schreiner & others, 1998). Therefore, a new test needed to be developed that differentiates between adhesives that produce higher bond strengths. A microtensile bond strength (µTBS) methodology was introduced by Sano & others in 1994(a). These authors showed that microtensile bond strength was inversely related to the bonded surface area (Sano & others, 1994a; Shono & others, 1999; Phrukkanon, Burow & Tyas, 1998a,b; Pashley & others, 1999) and that although much higher bond strengths were measured, most failures still occurred at the interface between tooth substrate and adhesive. Other advantages of µTBS-testing are that regional bond strengths and bonding effectiveness to clinically relevant tooth substrates such as carious (Nakajima & others, 1995; Yoshiyama & others, 2000) and sclerotic dentin (Tay & others, 2000; Kwong & others, 2002) can be measured (Pashley & others, 1999). The major disadvantage of µTBS-testing is the rather labor-intensive, technically demanding and relatively fragile sample preparation technique. Special care should be taken to avoid/reduce the production of microfractures at the interface during specimen preparation. They may weaken the bond and, thus, reduce the actual bond strength (Ferrari & Cardoso, 2002). Otherwise, one could argue that clinical restoration margins are...
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Figure 21. Chart presenting the micro-tensile bond strength (µTBS) to enamel of diverse commercial adhesives. The data were gathered from diverse laboratory studies carried out at BIOMAT Leuven strictly following the same experimental protocol. The color code refers to the different kinds of adhesives following the classification presented in Figure 1. For the two-step self-etch adhesives, the light-green colored bars represent the data produced by “mild” self-etch adhesives, the intermediary-green colored bars those produced by “intermediary strong” self-etch adhesives, and the dark-green bars represent the µTBS-data produced by the “strong” self-etch adhesives. For the one-step self-etch adhesives, the light-yellow colored bars represent the data produced by “mild” self-etch adhesives, the intermediary-yellow colored bars those produced by “intermediary strong” self-etch adhesives, and the dark-yellow bars represent the µTBS-data produced by the “strong” self-etch adhesives. All data are pooled per group of adhesives underneath the chart.

Figure 22. Statistical analysis of the pooled enamel µTBS-data demonstrate that three-step etch&rinse adhesives bond equally well to enamel as two-step etch&rinse adhesives. Etch&rinse adhesives bond slightly, but statistically significantly better to enamel than two-step self-etch adhesives, that on their turn bond significantly much better than one-step self-etch adhesives. The actual p-values are mentioned in the table insert. All red-colored figures indicate statistical significant difference; black-colored figures indicate absence of statistical difference.

Figure 23. Chart presenting the micro-tensile bond strength (µTBS) to dentin of diverse commercial adhesives. The data were gathered from diverse laboratory studies carried out at BIOMAT Leuven strictly following the same experimental protocol. The color code refers to the different kinds of adhesives following the classification presented in Figure 1. For the two-step self-etch adhesives, the light-green colored bars represent the data produced by “mild” self-etch adhesives, the intermediary-green colored bars those produced by “intermediary strong” self-etch adhesives, and the dark-green bars represent the µTBS-data produced by the “strong” self-etch adhesives. For the one-step self-etch adhesives, the light-yellow colored bars represent the data produced by “mild” self-etch adhesives, the intermediary-yellow colored bars those produced by “intermediary strong” self-etch adhesives, and the dark-yellow bars represent the µTBS-data produced by the “strong” self-etch adhesives. All data are pooled per group of adhesives underneath the chart.

Figure 24. Statistical analysis of the pooled dentin µTBS-data demonstrated that three-step etch&rinse adhesives bonded significantly better to dentin than two-step etch&rinse adhesives and two-step self-etch adhesives. Both two-step etch&rinse and self-etch bonded significantly much better than one-step self-etch adhesives. The actual p-values are mentioned in the table insert. All red-colored figures indicate statistical significant difference; black-colored figures indicate absence of statistical difference.

subjected to similar stresses during finishing of composite restorations with diamonds. They also induce microfractures at the restoration-tooth transition. In this way, µTBS-sample preparation may actually better simulate clinical circumstances. Eventually, if all specimens are prepared in the same manner, no additional variable is introduced. In order to standardize sample preparation, at BIOMAT Leuven, the Iowa MicroSpecimen Former (Armstrong, Keller & Boyer, 2001; De Munck & others, 2003a,c) is used, which
enables the production of stick-type specimens that are cylindrically constricted at the interface to ensure that the maximum tensile stress is concentrated at the actual interface. Hence, this µTBS-testing protocol must become the new standard for measure bonding effectiveness in the laboratory.

A modification of this test is the “micro-shear” test, which makes it more difficult to standardize the location of the force (Shimada & others, 2000). Nevertheless, the results obtained did not differ substantially from those gathered following a µTBS-protocol (Phrukkanon & others, 1998a,b).

Another less common technique is the push-out test (Frankenberger, Krämer & Petschelt, 1999, 2000b; Frankenberger & others, 2000a). A small resin composite cylinder in the middle of a dentin disc is pushed out, resulting in a shear stress at the interface. The main advantage of this technique is that failure is forced to occur along the adhesive interface (Drummond & others, 1996). However, this test is more time-consuming and cannot be applied for evaluating enamel bond strength. Also, push-out data are very comparable to traditional shear-bond strength testing (Drummond & others, 1996).

µTBS to Enamel—At Leuven, the µTBS of a large group of commercial and experimental adhesives to bur-cut enamel and dentin has been determined (Inoue & others, 2001a,b, 2003; De Munck & others, 2003a,c), always following the same experimental protocol, using one particular composite material (Z100, 3M ESPE). When bonding to enamel, an etch&rinse approach still results in the highest bonding effective-

In-house clinical results in Class-V lesions

Figure 25. Schematic explaining the principle of micro-rotary fatigue testing (µRF) and illustrating a specimen prepared for it. The sample is clamped at one end in the specimen grip and at the other end loaded with an adjustable weight. When the specimen is rotated, each spot “x” will be stressed cyclically in tensile and compression. The stress applied diminishes towards the center of the specimen.

Figure 26. Fe-SEM photomicrograph illustrating a resin-enamel µRF-specimen prepared using Clearfil SE (Kuraray) that failed after 25680 cycles at 19 MPa. The failure pattern was clearly mixed with “A” representing adhesive failure and “C” cohesive failure in the adhesive. The arrow probably represents the area where the crack was initiated. Typical beach marks are indicated by “B,” where “R” represents the rest fracture.

Figure 27. Chart presenting the retention percentage of Class-V restorations in function of time when diverse adhesive/composite combinations were used to restore non-carious cervical lesions as part of in-house clinical trials, which were started before 1990.
ness irrespective of a two- or three-step procedure and the product tested (Figure 21). When pooling the μTBSs of all etch&rinse adhesives tested (or of which the μTBS was repeatedly measured as part of different studies), a μTBS of 39 and 40 MPa was achieved, respectively, for the three-step and two-step etch&rinse adhesives. As has been known from Buonocore (1955), bonding to enamel only requires an acid-etch step followed by the application of a fluid resin without the need for an intermediary primer step. The latter, on the other hand, does not negatively influence bonding effectiveness and is even mandatory when a “wet-bonding” procedure is carried out.

A self-etch procedure, in general, has resulted in a lower bonding effectiveness, though some adhesives approached the bonding effectiveness of etch&rinse adhesives (Figure 21). A pooled μTBS of about 30 MPa was obtained for two-step self-etch adhesives. The μTBS of the “strong” two-step self-etch adhesive NRC/Prime&Bond NT (Dentsply) was not significantly lower than that of the etch&rinse adhesives. Although Clearfil SE (Kuraray) and the experimental successor Clearfil SE Plus (Kuraray; containing the anti-microbial monomer methacryloyloxydodecylpyridinium bromide or MDPB; Imazato & others, 1997) belong to the group of “mild” two-step self-etch adhesives, their μTBS is not that much lower than etch&rinse adhesives. This may indicate that although they only interact superficially with enamel and, thus, their potential for micromechanical interlocking is much less than a phosphoric-acid treatment, the additional chemical bonding capacity to hydroxyapatite may have contributed to the actual favorable bonding effectiveness. Likewise, the bonding effectiveness of the “intermediary strong” two-step self-etch adhesive Optibond Solo Plus Self-etch (Kerr, Orange, CA, USA) approached that of etch&rinse adhesives.

One-step self-etch adhesives produced significantly lower μTBSs than etch&rinse and two-step self-etch adhesives (Figure 21). The pooled μTBS was about 16 MPa for the one-step self-etch adhesives. This most likely can be attributed to their higher acidity and, consequently, higher potential to achieve micromechanical interlocking at enamel.

The glass-ionomer adhesive Fuji Bond LC (GC) performed equally well as the two-step self-etch adhesives (Figure 21). However, during bond-strength testing, the glass-ionomer adhesive tended to fail more frequently in the glass-ionomer material itself than at the actual interface, where their actual bonding effectiveness to enamel was never measured and should at least be higher than the cohesive strength of the glass-ionomer adhesive (Inoue & others, 2000, 2001a).

Statistical analysis of the pooled enamel μTBS data (Figure 22) showed that etch&rinse adhesives, irrespective of a two- or three-step application procedure, bonded slightly but significantly stronger to enamel than two-step self-etch adhesives and significantly more strong than one-step self-etch adhesives.

μTBS to Dentin—At dentin, three-step etch&rinse adhesives still surpassed all other adhesives that use simplified application procedures (Figure 23). No significant difference could be recorded between the bonding effectiveness to dentin of two-step etch&rinse and two-step self-etch adhesives. Again, the “mild” two-step self-etch adhesive Clearfil SE (Kuraray) and the “intermediary strong” two-step self-etch adhesive Optibond Solo Plus Self-etch (Kerr) most closely approached the bonding effectiveness of the conventional three-step adhesives. The lowest μTBS was again...
recorded for the one-step self-etch adhesives that performed similarly to the glass-ionomer adhesive Fuji Bond LC (GC).

Statistical analysis of the pooled dentin µTBS-data (Figure 24) showed that three-step etch&rinse adhesives bonded significantly more strongly to dentin than two-step etch&rinse and two-step self-etch adhesives. Both latter systems did not perform significantly different from each other. Again, the significantly least favorable µTBS-results were recorded for one-step self-etch adhesives.

**µTBS to Hydroxyapatite**—As mentioned above, the actual bonding effectiveness of glass ionomers and “mild” self-etch adhesives may result from combined micromechanical and chemical interaction with the tooth substrate. It is, however, currently not known how much chemical interaction contributes to the actual bonding effectiveness. Therefore, the authors determined the µTBS of a various group of adhesive materials to synthetic hydroxyapatite that, besides not having organic collagen, was highly polished and, thus, devoid of mechanical interlocking sites (Van Meerbeek & others, 2003). Among the adhesives tested, all specimens prepared with three-step etch&rinse adhesive Optibond FL (Kerr) failed prior to µTBS-testing (pre-testing failures), proving that any micromechanical retention was excluded. The two-step self-etch adhesive Clearfil SE (Kuraray) presented with a rather low µTBS, along with a high number of pre-testing failures, indicating that there is some chemical interaction that, however slightly (for about 7% as compared to its µTBS to dentin), contributed to the actual bond strength achieved at dentin. Clearly, much less pre-testing failure and a significantly higher µTBS were recorded for the resin-modified glass-ionomer adhesive Fuji Bond LC (GC) and the conventional glass-ionomer restorative material Fuji IX (GC). For Fuji Bond LC, the chemical interaction accounted for about 40% of the actual bond strength achieved at dentin. Chemical bonding of glass ionomer to hydroxyapatite depended greatly on the use of a separate polyalkenoic acid conditioner (Cavity Conditioner (GC); see also Figure 8). Without this pre-treatment, all specimens failed prior to testing. Equally effective as glass-ionomer materials with regard to chemical bonding potential, the resin-based luting material Panavia F (Kuraray) presented with a µTBS that accounted for about 67% of its actual bond strength to dentin. No pre-testing failures were recorded for Panavia F, indicating its relatively strong chemical bonding potential. Although Panavia F was applied following a self-etch approach using a 10-MDP-based primer solution, as in Clearfil SE, its chemical bonding effectiveness is much higher than the two-step self-etch adhesive Clearfil SE. Further in-depth analysis of the actual differences in composition and application procedures should help explain this difference in chemical bonding potential.

Finally, the self-adhesive luting material Unicem (3M ESPE) presented with a relatively negligible chemical bonding potential in the same range recorded for the two-step self-etch adhesive (Clearfil SE). Note that the ratio of chemical bonding to eventual “total” bonding effectiveness must be regarded as arbitrary, since differences in substrate properties such as roughness, stiffness and so on between the hydroxyapatite and dentin specimens were ignored.

**Effect of Aging**—Most current adhesives perform well in bond-strength tests, at least when tested shortly after application and under controlled in vitro conditions (Inoue & others, 2001b, 2003; De Munck & others, 2003a,c). However, the oral cavity with temperature changes, chewing loads and chemical attacks by acids and enzymes forms a severe challenge for tooth-composite bonds to survive for a long time. Clinically, marginal deterioration of composite restorations remains problematic and forms the major reason that dramatically shortens the lifetime of adhesive restorations (Van Meerbeek & others, 1998a). A factor known to degrade tooth-composite bonds is exposure to water (Gwinnett & Yu, 1995; Sano & others, 1999, Armstrong & others, 2001). Among different forms of marginal leakage, nanoleakage, or the ingress of oral fluids through nanometer-sized channels along collagen fibrils within the hybrid layer, is considered detrimental to the bond integrity (Sano & others, 1995; Hashimoto & others, 2000, 2002).

In a recent paper (De Munck & others, 2003c), the authors studied the long-term degradation of resin-dentin bonds using a µTBS-testing methodology through exposure to water for four years, either directly or indirectly, when the resin-dentin interface was surrounded by resin bonded to enamel. The microtensile bond strength (µTBS) to dentin of two three-step etch&rinse adhesives (Optibond Dual-Cure, Kerr; Scotchbond Multi-Purpose, 3M ESPE) was compared to two two-step etch&rinse adhesives (Optibond Solo, Kerr; Scotchbond 1, 3M ESPE) after four years of storage in water. Direct exposure to water resulted in a significant decrease in the µTBS of the two-step but not of the three-step etch&rinse adhesives. Indirect exposure to water did not significantly reduce the µTBS of any adhesive, indicating that resin bonded to enamel protected the resin-dentin bond against degradation. This means that, in the clinical situation, one can rely on durable dentin bonding using three- or two-step etch&rinse adhesives if all cavity margins are located in enamel. For cavities with margins ending in dentin, three-step total-etch adhesives are preferred.

**Marginal Sealing Effectiveness**

Clinically, early loss of restoration is no longer a clinical problem when reliable (mostly conventional three-step etch&rinse) adhesives are used, even long-term (Van Meerbeek & others, 1994, 1998a; van Dijken,
A number of studies have tested the performance of adhesives by applying a non-parametric scale evaluation method. It can be made semi-quantitative by using SEM (Sano & others, 1994b, 1995). Many methodologies have been introduced to assess microleakage and can be further subdivided in qualitative, semi-quantitative or true quantitative measurements of sealing effectiveness.

**Qualitative Measurement of Sealing Effectiveness**—The use of organic dyes as tracers is one of the oldest, most common methods of detecting leakage in vitro. A number of dyes varying in particle size and affinity to substrates have been used and are known to significantly influence microleakage results (Alani & Toh, 1997). In general, this method involves immersion of a restored tooth into a dye solution after having coated the unrestored tooth parts covered with a waterproof varnish until close to the restoration margin. After a certain time interval, the specimens are washed and sectioned into two or more slices to visually determine the extent of dye penetration along the restoration margin (Alani & Toh, 1997). The main problem is that this methodology basically is a qualitative evaluation method. It can be made semi-quantitative by applying a non-parametric scale (Castelnuovo, Tjan & Liu, 1996).

**Semi-Quantitative Measurement of Sealing Effectiveness or Marginal Analysis**—A number of in vitro studies have tested the performance of adhesives by semi-quantitatively evaluating by using SEM the marginal gap formation around restorations placed in extracted teeth (Roulet & others, 1989; Krejci, Kuster & Lutz, 1993; Roulet, 1994; Gladys & others, 1995; Blunck, Neumann & Roulet, 2000; Blunck & Roulet, 1999, 2002). This method assumes that if the forces generated during shrinkage or thermo-mechanical strains exceed the bond strength to enamel/dentin, an observable gap will form at the margin of the restoration. Although the literature also lacks clear evidence of any correlation of gap formation in vitro with the interfacial failures observed in vivo, it is reasonable to assume that this semi-quantitative marginal gap analysis is clinically relevant (Roulet, 1994), certainly, when measurements are repeated after thermocycling (Krejci & others, 1993, Schuckar & Geurtsen, 1997).

Blunck and Roulet (2002) have semi-quantitatively analyzed the marginal adaptation of cervical restorations for a diverse group of adhesives, consistently following the same experimental protocol. Basically, their results correlated well with the µTBS-data recorded by the authors of this study at BIOMAT Leuven. After one-year water storage and two thermocycling sessions, still on average, 93% of the restoration margin length was gap-free for the three-step etch&rinse adhesive Optibond FL (Kerr) and 91% for the “mild” two-step self-etch adhesive Clearfil SE (Kuraray) (Blunck & Roulet, 2002). Two-step etch&rinse adhesives such as Excite (Vivadent), Optibond Solo Plus (Kerr) and Scotchbond 1 (3M ESPE) revealed significantly lower percentages of gap-free margin lengths of 80%, 82% and 63%, respectively. Less than half (48%) of the margin length was gap-free for the “strong” one-step self-etch adhesive Prompt-L Pop (3M ESPE).

**Quantitative Measurement of Sealing Effectiveness or Flow Measurement**—A quantitative method to assess microleakage is to measure the flow along the interface (Pagliarini & others, 1996) or from the pulp to a sealed dentin surface (Derkson, Pashley & Derkson, 1986; Del-Nero, Escribano & de la Macorra, 2000; Bouillaguet & others, 2000). The marginal sealing effectiveness is quantified using a “Flodec device” (De Marco Engineering, Geneva, Switzerland). The adhesively-restored tooth is brought under pressure with water from inside the dental pulp. The permeability of the tooth-restoration interface is then quantitatively determined through accurate measurement of the displacement of an air bubble within a water-filled micro-pipet (Ø=0.7 mm) using a computer-driven optical system (Flodec device). The main advantages of this method are that it is fully quantitative and that the specimens can be longitudinally followed since it is a non-destructive method. However, one major problem using this technique is that leakage may also occur through the dental substrate itself and, thus, falsely increase the leakage values.

**Nanoleakage**—Sano and others (1994b, 1995) revealed that leakage can occur between the hybrid layer and intact dentin, even in the absence of a marginal gap. This leakage was assessed using Ag-ions that are extremely small (0.059 nm). It is hypothesized that it represents permeation through demineralized sub-micron spaces that have not been filled with adhesive resin (Sano & others, 1995). These voids are so small that bacteria may not be able to pass through, but these spaces may be more susceptible to degrada-
tion by water and bacterial side products such as acids and enzymes (Paul & others, 1999). This phenomenon can be quantitatively assessed by measuring the dye penetration depth using, for instance, confocal laser scanning microscopy (Dörfer & others, 2000; Pioch & others, 2001, 2002) or TCM (Tay & others, 2002b).

Dynamic Fatigue Testing

Bonding effectiveness to tooth tissue is typically measured statically, for example, by shear bond or microtensile bond strength (µTBS) testing (see above). In the clinical situation, however, tooth-composite bonds are seldom imposed to such acute tensile/shear stresses. During its lifetime, a restoration is subjected to cyclic loading, each load is insufficient to provoke failure, but in the long-term, can possibly lead to marginal deterioration and loss of the restoration. Therefore, fatigue testing of dental adhesives is expected to better predict their in vivo performance.

There is, however, no standard fatigue test for dental adhesives. Possible methods are a cyclic shear test (Ruse, Shew & Feduik, 1995; Drummond & others, 1996; Devji & others, 1998), a cyclic tensile test (Aquilino, Diaz-Arnold & Piotrowski, 1991; Givan & others, 1995), a cyclic fracture toughness test (Destoop, 2002) or a cyclic push-out test (Frankenberger & others, 1999). Another possibility is loading only not the interface but the whole tooth until the tooth-restoration complex fails (Fissore, Nicholls & Yuodelis, 1991).

At BIOMAT Leuven, the authors have developed a micro-rotary fatigue device that dynamically tests tooth-composite interfaces (De Munck & others, 2002). A macro-version was used prior to determine the fatigue resistance of soldered joints (Wiskott, Nicholls & Belser, 1994). In our test set-up, standard microtensile bond strength (µTBS) bar-type samples prepared with a rounded, constricted interface (Figure 25 and 26) were clamped in a pin-chuck and connected to a stepping motor with the free end loaded with a certain weight. By rotating the specimen, each spot at the outer surface of the interface underwent successively compressive and tensile loading following a sinusoidal function (Wiskott & others, 1994). Depending on the survival/failure of each sample after 10^5 cycles, the load imposed to the next sample was increased/decreased with ± 5%. The results of the fatigue test were analyzed using a logistic regression to determine the load at which 50% of the samples failed and was called the median micro-Rotary Fatigue Resistance (µRFR). In a pilot study, the µRFR of the three-step etch&rinse adhesive Optibond FL (Kerr) and the two-step self-etch adhesive Clearfil SE (Kuraray) to enamel and dentin was determined. The ranking of median µRFRs was in accordance with the ranking of the respective µTBSs obtained for the two adhesives bonded to enamel and dentin. They were about three-fourths of the respective µTBSs, except for Optibond FL bonded to dentin, which appeared to lose more of its static bond strength when tested dynamically. From this preliminary study, it could be concluded that fatiguing of tooth-composite interfaces is feasible, with consistent results provided. Because of the cyclic loading and high number of cycles (10^5), the resulting data might also be more clinically relevant, especially for assessing long-term bonding effectiveness, which is still a major shortcoming of contemporary adhesives.

Clinical Testing of Adhesives

New adhesives are continually being introduced to the dental profession, unfortunately, often without sufficient clinical validation (Van Meerbeek & others, 1998a, 2001a). In the mouth, multiple and mutually-interactive clinical variables related to the quality of tooth substrate and its immediate oral environment co-determine the eventual effectiveness of adhesives (Van Meerbeek & others, 1994). Adhesives have mainly been clinically tested in non-prepared cervical abrasions and erosions. Such “model” lesions are ideal test cavities, because they are located mainly in dentin and are widely available. They present no macro-mechanical undercuts, and they are usually found in anterior teeth or premolars with good access and in patients who have better than average oral hygiene. However, such clinical trials are limited in number and require several years with regular recalls in order to achieve sufficient clinical validation. Nevertheless, the more expensive and long-lasting clinical trials remain necessary to validate laboratory observations. Laboratory testing on near ideal substrates and under optimal in vitro conditions is valuable as a pre-clinical screening test of adhesive materials, at best, only a good prediction of clinical performance. Most Class-V clinical trials run for three years, although longer follow-up times may be desirable. However, after three years, most adhesives are outdated and are replaced by a successor that claims to be better.

At Leuven, the clinical effectiveness of adhesives has been routinely investigated in controlled follow-up studies using the same experimental protocol for almost 20 years. The clinical effectiveness of modern adhesives has significantly improved, allowing adhesive restorations to be placed with a high predictable level of clinical success. Most modern adhesive systems are superior to their predecessors, especially in terms of retention, making it no longer the main cause of premature clinical failure. This must, in part, be attributed to the introduction in the early 1990s of the “total-etch” (now referred to as “etch&rinse”) technique, by which phosphoric acid is also applied to dentin. Earlier adhesives often showed many failures within the first six months when applied strictly to dentin without any selective phosphoric acid-etching of adjacent enamel (Figure 27). When following the same protocol in more
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recent clinical trials (etch&rinse systems applied selectively to dentin), almost any early debonding failures were recorded (Figure 28). This must, to a great extent, be attributed to the enamel immediately adjacent to dentin always being (unintentionally) etched and the guarantee of a durable bond to the enamel margin. Adequate bonding to enamel, alone, may also keep such restorations in place. Nevertheless, bonding to dentin has improved substantially. However, in order to be considered clinically effective, adhesive systems should not only keep the restoration in place for a significant period of time, but also, and what clinically may even be more important, completely seal the restoration margins against the ingress of oral fluids and microorganisms. However, none of today's systems yet appears able to guarantee leakage-free margins for a significant amount of time, especially at the dentin site (Van Meerbeek & others, 1998a; De Munck & others, 2003b).

At Leuven, the excellent clinical performance of the three-step etch&rinse adhesive Optibond FL (Kerr), with a 100% retention rate at five years, is noteworthy (De Munck & others, 2003b). Likewise, 96% of the restorations were still in place at five years when the three-step etch&rinse adhesive Permaquick (Ultradent, South Jordan, UT, USA) was used (De Munck & others, 2003b). Besides the favorable clinical performance of etch&rinse adhesives, glass ionomers commonly present with high retention results, even up to three years of clinical service (Figure 28). In a recent double-blind, split-mouth, randomized controlled clinical trial, the clinical effectiveness of the “mild” two-step self-etch adhesive Clearfil SE (Kuraray) was evaluated following two experimental protocols (Peumans & others, 2003). Clearfil SE was applied either following the manufacturer’s instructions or including prior selective acid etching of the enamel cavity margins with 40% phosphoric acid. At two years, no restoration losses were recorded for either experimental group (Figure 28). Besides a higher tendency toward small (but of clinically negligible relevance) marginal defects at the enamel side (when enamel was not etched beforehand with phosphoric acid), the “mild” self-etch approach of Clearfil SE still appears to be a clinically reliable, predictable and simplified adhesive technique.

In general, two-step etch&rinse adhesives perform clinically less favorably than conventional three-step adhesives (Sunnegardh & van Dijken, 2000; van Dijken, 2000a). For instance, still favorable seven-year retention rates of 84% and 79%, respectively, were recorded for the three-step etch&rinse adhesives Clearfil Liner Bond (Kuraray) and Optibond Dual-Cure (Kerr) (van Dijken, 2001). Two-step etch&rinse adhesives generally perform clinically less favorably in Class-V lesions. The results reported for this group vary more among the different research centers, which is probably indicative of their higher technique sensitivity. For instance, only 45% of the acetone-based adhesive One-Step (BISICO, Schaumberg, IL, USA) were retained at five years (van Dijken, 2001), and only 52% of Gluma 2000 (Kulzer) at five years (van Dijken, 2000a). Also, 25% of the Scotchbond 1 restorations were already lost at the three-year recall in a study by van Dijken (2001), while only 3% were lost at three years in a study by Ripps, Burgess and Rappold (2000). A loss rate of only 7% was recorded for Optibond Solo at three years (Swift & others, 2001). At three years, excellent and reasonably good clinical effectiveness was reported for Prime&Bond 2.1 (Dentsply), with a retention rate of 100% at three years by Martin, Jednakiewicz and Fletcher (2002), and 89% at three years by Swift and others (2001).

Regarding the clinical effectiveness of two-step self-etch adhesives, less data is available in the literature. Latta and others (2000) reported a still favorable 92% retention rate at three years for Clearfil Liner Bond 2 and van Dijken (2002) reported a 91% retention rate at two years.

Finally, regarding one-step self-etch adhesives, strongly varying results were recorded for PSA (applied along with Dyract, Dentsply). Only a 5% loss rate at five years was reported by Folwaczny and others (2001), whereas, even 41% of the restorations placed using PSA (Dentsply) de-bonded within a four-year observation period, as reported by Unlu, Belli and Özer (2001). A rather favorable retention rate of 84% at five years was reported by van Dijken (2000a). Several studies reported on the clinical performance of Prompt L-Pop (3M ESPE). Rather favorable short-term retention rates of 100% at six months and 96% at one year, respectively, were recorded by Munoz and others (2001) and by Boghosian (2002). However, relatively high loss rates of 21% at two years and 35% at one year were reported, respectively, by van Dijken (2002) and Brackett, Covey and St Germain (2002).

CONCLUSIONS

A great diversity in laboratory testing of adhesives exists. Modern determination of bonding effectiveness in the laboratory should involve (1) microtensile bond strength testing, (2) sealing effectiveness testing using semi-quantitative marginal analysis or fully quantitative margin permeability measurement and possibly (3) dynamic fatigue testing. There is a lack of standardization of testing methodologies. Nevertheless, good correlation exists between laboratory and clinical effectiveness, by which it can be concluded that laboratory testing CAN predict clinical effectiveness.

Diverse types of adhesives exist which can be classified following their bonding mechanism and clinical
application approach into etch&rinse, glass-ionomer and self-etch adhesives. Although there is a tendency toward adhesives with simplified application procedures, simplification does not guarantee equal or improved bonding effectiveness. Three-step etch&rinse adhesives still perform best in laboratory and clinical research. Because of an additional chemical bonding potential to hydroxyapatite, the mild self-etch approach may be most promising in terms of durable bonding to dental hard tissue using a simple, low, technique-sensitive application technique.

Acknowledgements
The Buonocore Memorial lecture is supported by a grant from Caulk Dental Manufacturing Co to the Academy of Operative Dentistry.

(Presented 27 February 2003)

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