

## INTRODUCTION

The luting procedure is a key to the long-term success of fixed restorations. The strength and durability of the bond between the prosthesis, the luting agent and the enamel/dentin interface play an important role in the outcome of fixed prosthetics.<sup>1</sup>

With an increasing demand for esthetic restorations and the increased use of thinner ceramic/porcelain restorations such as veneers, the luting procedure becomes more critical. High failure rates have been associated with such restorations, especially when a large amount of dentin is involved.<sup>1</sup> That is why the importance of the predictability of adhesive luting agents has increased.

There is a wide choice of luting agents and products, each with advantages and disadvantages. The selection procedure is based on each product's properties and the published studies, but most of all on operator preference.

As newer materials are introduced into the market to overcome the disadvantages of previous materials, it is more difficult to keep track of each material's properties. According to the ISO 4049 (2000)/96 specifications and the ADA professional product review for resin-based cements/water-based cements, water sorption and solubility are tests required to validate any resin-based cement.

The prognosis of prosthetic restorations is largely impacted by the maintenance of the luting agent and the adhesive bond.<sup>2</sup> Clinically, the luting interface has to withstand masticatory and parafunctional stresses in a wet, warm environment. When exposed to water or saliva, most restorative materials undergo hydrolytic degradation.<sup>3</sup>

Some studies identify dissolution rather than physical disintegration as the mechanism for cement erosion.<sup>4</sup>

Water can penetrate the polymer and result in the breaking of secondary bonds (van der Waals forces) between the polymer chains. As a result, the mechanical properties of the resin decrease. Water will have a plasticizing effect on the polymer, reducing both the bond strength and the mechanical properties.<sup>5-7</sup> Some have argued that the amount of water sorption can be beneficial to overcome the shrinking stresses.<sup>8</sup>

This study evaluated the water solubility and water sorption characteristics of nine different polymeric luting agents over a 180-day, water-storage period.

#### NULL HYPOTHESES

The null hypotheses to be tested were:

1. There is no significant difference in water sorption between the different materials.
2. There is no significant difference in solubility between the different materials.

The alternative hypotheses were:

1. The cements based on more hydrophilic monomers (self-etching resin cements and resin-modified glass-ionomer cements) will show more water sorption when compared with the other cements.
2. The self-etch resin cements will show significantly greater solubility.

LITERATURE REVIEW

## LUTING AGENTS

Luting agents, viscous materials placed between tooth structure and a prosthesis, harden through chemical reaction to firmly attach the prosthesis to the tooth structure.<sup>9</sup>

An ideal luting agent should be: biocompatible with the tissue that it contacts; able to adhere to tooth substance and restoration; able to prevent leakage by producing a good marginal seal; cariostatic; insoluble in the oral cavity; able to resist water-sorption; low in film thickness; able to resist the forces transmitted to the lute through the restoration; able to achieve maximum physical properties as quickly as possible; able to allow easy removal of excess; available in a range of different shades, and radiopaque. There is no such ideal material.

Luting agents can be divided into three categories according to their adhesive potential: low, such as zinc phosphate; medium, represented by the polycarboxylate cement, and high, represented by the glass-ionomers, resin-modified glass-ionomers and resin-based luting agents.<sup>10</sup>

Table I summarizes some properties of available luting materials.<sup>11</sup>

## THE GLASS-IONOMER LUTING AGENTS

Conventional glass-ionomer cements (GIC) were introduced to the dental profession by Wilson and Kent in 1972.<sup>12</sup> GICs are formed by the acid-base reaction of an aqueous polymeric acid (polyalkenoic acid) such as polyacrylic acid and a glass component that is usually a fluoroaluminosilicate. GIC was derived from silicate cements

and polycarboxylate cements. The reaction leads to the slow formation of an amorphous silicate structure by hydration of silicon.<sup>13</sup>

Clinical success of glass-ionomer cement as a luting agent has been well documented.<sup>14</sup> It gained popularity due to its fluoride release and bonding to tooth structure. Muzynski et al. examined the solubility of glass-ionomer cements by measuring the fluoride release from simulated dental restorations, and their results agree with other studies.<sup>15</sup> However, fluoride release is not an indication of solubility because the fluoride salts are embedded in the matrix without being incorporated into the reaction. Glass-ionomer luting agents have been shown *in vitro* to reduce demineralization around crowns despite reduced solubility.<sup>16</sup> *In vivo*, glass-ionomer cement has been found to increase the fluoride level in the saliva in the short-term,<sup>17</sup> to increase adjacent enamel fluoride levels<sup>18</sup> and to modify the caries-causing organisms.<sup>19</sup>

Tested for compressive strength, glass-ionomer cement showed high values. The compressive strength of glass-ionomer cement continues to increase over several weeks to reach about 200 MPa.<sup>20,21</sup> This continued increase is thought to be due to reconstruction of the silicate network.<sup>22</sup>

Glass-ionomer cements are sensitive to water erosion. Early water contamination of glass-ionomer leaches  $\text{Ca}^{2+}$  and  $\text{Al}^{3+}$  ions resulting in excessive opacity, rapid disintegration and clinical wear. Clinical success with glass-ionomer cements depends on early protection from both hydration and dehydration. It is weakened by early exposure to moisture, while desiccation produces shrinkage cracks in the recently set cement.<sup>23</sup>

Some studies have concluded that glass-ionomer cements are more resistant to degradation than zinc phosphate cements, although Knibbs and Walls reported that

marginal defects around crowns appeared sooner with glass-ionomer than with zinc phosphate, possibly because of the greater susceptibility of glass-ionomer to contamination by moisture.<sup>24</sup> Contaminated glass-ionomer is more susceptible to erosion, and glass-ionomer aged in water is mechanically weaker.<sup>25</sup>

A clinical study with patients wearing luting specimens in the lingual flanges of inferior complete dentures showed that polycarboxylate and zinc phosphate cements dissolve more than glass-ionomer cement. Under scanning electron microscopy, glass-ionomer showed pits and extensive cracks on their surfaces.<sup>26</sup> Keyf et al. compared the water sorption and solubility of glass-ionomer luting agents with four provisional, three permanent luting cements (zinc polycarboxylate, zinc phosphate and glass-ionomer) and five restorative cements. The glass-ionomer luting agent showed the lowest solubility with significant differences between water sorption and water solubility. Some glass-ionomer cement showed negative values for solubility implying the uptake of water into the cement structure.<sup>27</sup>

#### THE RESIN-MODIFIED GLASS-IONOMER LUTING AGENTS

In 1988, resin-modified glass-ionomer cements (RM-GIC) were introduced to overcome some of the glass-ionomer cements' problems such as sensitivity to humidity and early, weak mechanical strength.<sup>28</sup> RM-GICs were formed by the replacement of the polyacid with a modified polyacid grafted with unsaturated groups, and the incorporation of polymerizable hydrophilic resins.<sup>28</sup> The hydrophilic resin, such as HEMA, is added as a co-solvent. It also polymerizes or copolymerizes with the modified polyacid.<sup>29</sup> RM-GIC shows some advantages over the conventional GIC. They particularly allow a longer working time as they are photo-chemically initiated, reducing the early sensitivity to

moisture and dehydration associated with the early stage of the acid-base setting reaction in the conventional GIC.<sup>30</sup> They show rapid hardening of their surface. The inclusion of resin in the glass-ionomers leads to an increase in flexural and tensile strength of the cement. It is not clear if the inclusion of resin into the cements increases the surface microhardness and resistance to compression.<sup>31, 32</sup> Ellakuria et al. compared the microhardness of resin-modified versus conventional glass-ionomer cements after one-year water storage. The resin-modified glass-ionomer cements showed a significantly lower hardness than the conventional GIC.<sup>33</sup> They explained their findings as potentially due to the interposition of the HEMA matrix preventing the complete formation of the poly-salt matrix,<sup>34</sup> inhibiting the acid-base reaction.<sup>35</sup> On the other hand, the decrease in microhardness may be due to the hypothetical separation of the phases described in the microstructure of these materials.<sup>30, 36</sup> A third explanation for the reduction in microhardness could be the high proportion of functional hydrophilic groups contained within the matrix absorbing a large quantity of water and thus producing a plasticizing effect.<sup>37, 38</sup> Bourke et al. stated that RM-GIC reaches its maximum hardness at one day after which no significant increase is detected.<sup>39</sup>

A luting agent should have sufficient mechanical properties to resist functional forces over the lifetime of the restoration. Glass-ionomer was found to be significantly harder than the resin modified glass-ionomer.<sup>32, 34, 38, 40</sup> These results are possibly related to the presence of a solid silicate phase around the non-reacting glasses responsible for the hardening.<sup>13, 22</sup>

Saskalauskaite et al. tested the flexural strength and elastic modulus of RM-GIC and resin luting cements. The RM-GIC was characterized by lower flexural strength and



elastic modulus.<sup>41</sup> The resin-modified glass-ionomers are expected to absorb water easily due to the presence of the hydrophilic species. It has been argued that water sorption is beneficial in terms of stress reduction during setting.<sup>8</sup> Water sorption also reduces the cement compressive strength.<sup>36</sup> Cattani-Lorinte et al. tested the effect of water on the physical properties of the resin-modified glass-ionomer cements, and stated that the RM-GICs are very sensitive to water sorption. Samples left in contact with water showed lower flexural strength, lower elastic modulus and a softer surface than dry samples.<sup>42</sup>

#### THE RESIN-BASED LUTING AGENTS

Resin-based luting material has gained in popularity due to the benefit of the acid-etch technique for attaching resin to tooth substrate and the possibility of attaching the resin to the prosthetic material if properly prepared. In addition, low solubility has been reported with the resin luting agents.

The resin-based luting materials are suitable for use with all indirect restorative materials. They have shown the best mechanical properties of all luting cements. One drawback is that they require more complicated clinical procedures, such as different bonding systems for the dentin, enamel, and for the restoration.<sup>43</sup>

Adhesive resin cements have shown reduced microleakage *in vitro*,<sup>44-46</sup> and *in vivo*.<sup>47</sup> Filled resin cement showed higher values in flexural strength,<sup>21, 48</sup> modulus of elasticity,<sup>49</sup> fracture toughness, and hardness testing when compared with traditional luting agents, GIC, RM-GIC and unfilled resin.<sup>50, 51</sup>

## SELF-ADHESIVE LUTING AGENTS

The concept of self-etching adhesives is based on the use of polymerizable acidic monomers, which will etch both enamel and dentin simultaneously. Commercially available self-etching adhesive products contain monomers that can be divided into three main groups according to their function: 1) Self-etching adhesive monomers 2) Cross-linking monomers, and 3) Additional monofunctional co-monomers (Figure 1).<sup>52</sup>

Phosphorous-containing monomers are capable of etching both enamel and dentin. They were first used in dental adhesives of the second generation; the first commercially introduced compound being the glycerol dimethacrylate ester of phosphoric acid (GDMP).<sup>53</sup> These compounds are well known in the literature for their hydrolytic instability.<sup>54</sup> Applying monomers containing a more hydrolytically stable bond between the polymerizable group and the strong acidic phosphate group solves hydrolytic instability of methacrylate phosphates. Anbar et al. carried out the first evaluation of polymerizable phosphates for dental adhesives.<sup>55, 56</sup> They showed that vinylphosphonic acid (VPA) and 4-vinylbenzylphosphonic acid (VBPA) (Figure 2) or corresponding copolymers can improve the adhesion of restorative composites on etched enamel. Other acrylic ether phosphoric acids were also introduced (AEPA) with improved hydrolytic stability and reactivity in the free-radical polymerization. All self-adhesive luting agents tested in this study contained phosphorous monomers (Figure 3).<sup>57</sup>

Fillers in the luting agents are divided into two groups. One group is sialinated to bond with the polymerizable monomer that improves the mechanical properties of the luting agent. The other group is alkaline in nature, acting as a buffering agent to raise the

pH after conditioning and facilitating proper penetration reducing the hydrophilicity and thus the viscosity, increasing the wettability.<sup>58</sup>

The self-etch luting agent should have a degree of hydrophilicity to improve its wettability and penetration into conditioned enamel/dentin; but, it becomes more hydrophobic during the setting reaction.

De Munck et al. examined the bonding of auto-adhesive luting agents to enamel and dentin using high-resolution electron microscopy. De Munck stated that the micro-tensile bond strength of RelyX Unicem (3M ESPE, Seefeld, Germany) to enamel was significantly lower than that of the control group Panavia-F (Kuraray Medical, Tokyo, Japan) while there was no significant difference in the bond strength to dentin.<sup>59</sup> This finding agrees with other studies comparing Rely X Unicem to Panavia-F.<sup>60, 61</sup> Panavia showed a high bond strength to both enamel and dentin.

Abo-Hamar et al. compared the bond strength of Panavia-F, Rely X Unicem, and Ketac-Cem (3M ESPE, Seefeld, Germany) after being subjected to thermocycling. The bond strength of Rely X Unicem to enamel was significantly decreased, but was still significantly higher than Ketac-Cem.<sup>62</sup> Wiedig et al. tested the bond strength of two new self-adhesive luting cements to human dentin, (Rely X Unicem and Maxcem, Kerr, Orange, CA) and an experimental self-adhesive paste/paste material. Maxcem showed a significantly lower bond strength compared with the other two. On the other hand, no significant difference between Rely X Unicem and the experimental self-adhesive paste/paste material was found.<sup>63</sup>

Fabianelli et al. investigated the adaptation of self-adhesive resin cement used for luting gold and ceramic inlays. Rely X Unicem, Fuji-Cem (GC America, Illinois) and

Variolink II (Ivoclar Vivadent, Schaan, Liechtenstein) were compared. There was no significant difference on marginal adaptation of the three-tested cements.<sup>64</sup> These results agree with those of Behr et al.<sup>65</sup> and Ibarra et al.<sup>66</sup>

In a study comparing the traditionally used luting agents, represented by Calibra (Dentsply DeTry, Konstanz, Germany) and Panavia F2, to the new self-etching approach luting agents represented by Rely X Unicem and Maxcem, it was stated that all systems involving the etch-and-rinse approach resulted in a significantly higher percentage of gap-free margins in enamel than the other luting systems. The systems significantly differed in gap formation in dentin.<sup>67</sup> These results agree with other studies that all imply that the simplification may ease handling for the general practitioner but may not improve adhesive effectiveness.<sup>68-70</sup> All-in-one adhesives exhibit a certain permeability that has been demonstrated both *in vitro* and *in vivo*.<sup>71-76</sup> Tay et al. explained these findings as a result of excessive primer/adhesive solvent at the interfacial layer of self-etching systems that can possibly provide channels for nanoleakage and may lead to hydrolytic degradation of the bond.<sup>75</sup>

## SORPTION AND SOLUBILITY

Multiple studies have reported that long-time storage in water affects the mechanical properties of the cements.<sup>21, 31, 77, 78</sup> Cattani-Lorente et al. concluded that deterioration of the physical properties of the cements after long-term storage in an aqueous environment could be related to the water sorption of these materials.<sup>79</sup> Part of the water absorbed could act as a plasticizer and result in a weakening of the cement.<sup>80</sup>

One study by Piwowarczyk et al. investigated the effect of water storage on flexural strength and compressive strength of 12 luting cements. The 12 cements included

two glass ionomer cements (Fuji I and Ketac-Cem), three resin-modified glass-ionomers (Fuji Plus, Fuji Cem and Rely X Luting), four resin cements (Rely X ARC, Panavia F, Variolink II, and Compolar) and one self-etch resin cement (Rely X Unicem). They found that Panavia and Rely X Unicem light-cured had a significant decrease in flexural strength after 150 days of storage in water. Fuji I, Ketac-Cem and Rely X Luting showed a significant increase in their flexural strength. Rely X Unicem showed a significant decrease in its compressive strength after 150 days.<sup>81</sup> Another study by Ortengren et al. showed a significant decrease in modulus of elasticity and strength of resin cement specimens after 60 days of water storage compared with the dry storage.<sup>82</sup>

Pedreira et al. investigated the effect of water storage on the microhardness of resin cements; Rely X Unicem and Panavia F were two of the cements tested. After seven days' water storage, Rely X Unicem showed high initial hardness. Three-month water-storage had no influence on the hardness of most of the cements, with the exception of Rely X Unicem, which showed a significant increase in the hardness values.<sup>83</sup>

Gerdolle et al. evaluated the water sorption characteristics and the solubility behavior of four luting cements; two of the tested luting agents were a composite resin (Panavia F) and a resin-modified glass-ionomer (Fuji Plus). Fuji Plus exhibited overall higher values of water sorption and solubility, while Panavia showed low values. Gerdolle concluded that the behavior of resin-based materials in water varies according to the composition characteristics, in particular, the high portion of hydrophilic chemical species as well as the filler characteristics.<sup>84</sup> Mese et al. agreed with Gerdolle's conclusion that solubility and sorption values were found to depend on the type and

content of fillers, filler concentration, mean particle size, the coupling agents, the nature of the filler particles, and the type of solvent.<sup>85</sup>

Water has an effect on the bonding strength of the cement to both the tooth structure and the restoration. Oyagüe et al. stated that, after a six-month water-storage period, the bond strength of dual-cured resin cements was significantly decreased (Clearfil Esthetic Cements and Calibra) but that Rely X Unicem showed no significant change in bond strength after water-storage.<sup>86</sup> In a different study Abdulla et al. investigated the effect of direct and indirect water storage (where the luting agent was bonded to dentin, then covered in a way that the water exposure was limited through a dentin bridge) on the microtensile dentin bond strength of total etch and two self-etch adhesives; after one year of indirect water storage, bond strength decreased but showed no significant difference from that after 24 hrs of water storage. After one year of direct water storage, a significant decrease in microtensile bond strength was detected.<sup>87</sup> These results do not agree with those of Oyagüe et al., but we can argue that Abdulla was investigating dentin bonding adhesives, which are expected to behave better in a wet environment.

There is more than one technique to assess the water uptake of dental materials. One technique used to assess the actual water content of a sample directly is weight change. The technique described in ISO Specification No. 4049 for assessing water sorption and solubility was used in this study.

According to the ISO specification, the water sorption and solubility behavior of a material is assessed in a one-week period. This study covered a longer period because it

has been shown in different studies that polymeric materials uptake water continuously over a long period of time before reaching equilibrium.<sup>27, 33, 42, 81, 88</sup>

MATERIALS AND METHODS



Ten luting agents were investigated in this study (Table II). Panavia F (Kuraray Medical Tokyo, Japan) and Fuji I (GC America, Illinois) were used as control groups. Some complications occurred preparing the Fuji I samples, which led to the use of Panavia F only as a control.

### SPECIMEN PREPARATION

Discs were prepared using molds made of natural acetal (Delrin) with internal dimensions of  $15 \text{ mm} \pm 0.1 \text{ mm}$  in diameter and  $1.0 \pm 0.1 \text{ mm}$  depth. These discs were placed on top of a glass slide covered with a Mylar sheet. Then, the discs were slightly overfilled with the materials to be tested. The materials were prepared according to the manufacturer's instructions (Table III). After the molds were filled, they were covered with a second Mylar sheet, and a glass slide was used to remove excess material. Each specimen was examined with the naked eye against a light to check for internal porosities or defects. The light-cured samples were examined before and after light curing. A light-curing unit (L.E.Demetron I, Kerr Corporation, Orange, CA) with an output of  $790 \text{ mW/cm}^2$  was used to photo-activate the dual cured cements. Curing was done using a 13 mm tip with each specimen cured in overlapping sections each for 40 seconds on both upper and lower surfaces until the whole specimen was irradiated (total of eight curing times). Then, specimens were transferred to an oven maintained at  $37 \text{ }^\circ\text{C} \pm 1 \text{ }^\circ\text{C}$ . The self-cured specimens were placed in the oven for 60 min while the light and the dual-cured specimens were placed for 15 min. Discs then were randomly assigned to each of

the storage periods in water ( $n = 52$ , 13 for each storage time): seven, 30, 90 and 180 days.

#### SORPTION AND SOLUBILITY ANALYSIS

A microbalance (Mettler Toledo, AG285, Switzerland) with a precision of 0.01/mg was used for weighing the specimens. The water sorption/solubility test was performed according to the ISO 4049 (2000) specification for resin-based restorative materials. Each specimen was finished by holding the periphery against 1000-grit abrasive papers on a non-rotating grinding table; specimens were rotated so that the periphery was abraded, assuring the removal of flash. A visual inspection of the periphery ensured their smoothness. After that, the specimens were transferred to a desiccator maintained at  $37\text{ }^{\circ}\text{C} \pm 1^{\circ}\text{C}$ . After 22 hours, each specimen was removed and stored in a second desiccator maintained at  $23\text{ }^{\circ}\text{C} \pm 1^{\circ}\text{C}$  for 2 hrs and then weighed. This cycle was repeated until the weight loss of each specimen was not more than 0.1 mg in any 24-hour period; this constant weight was  $W_0$  (the initial weight). After the final drying, the mean diameter was determined by calculating the mean of two measurements at right angles to each other across the specimen surface. Then, the mean thickness by calculating the mean of two measurements'  $180^{\circ}$  angle from each other was measured using a digital caliper with a precision of 0.01 mm (Max-cal, Cole Parmer Instrument Co., Chicago). The area and then the volume,  $V$ , in cubic millimeters were calculated. Before immersing the specimens in water, the specimens' densities were determined from weight and volume measurements using the equation:  $\rho = W_0/V$  ( $\text{g}/\text{mm}^3$ ). Any samples with a density value less than 10 percent of the average were discarded due to the possible presence of internal voids. The specimens then were immersed in water for the

selected storage period: seven, 30, 90 and 180 days. Each specimen was immersed in 10 ml of water in an individual glass container. The water was changed every week. The specimens immersed for the one-week period were not subjected to a water change.

Water pH was measured each time ( $\text{pH} = 5.5 \pm 0.45$ ).

After each period, each specimen was removed, washed with water, blotted till no water was visible at the surface, and then air dried for 15 seconds. The weight for each specimen was measured to 0.1-mg accuracy within one minute of removal from the water storage and the data were recorded. The weight measured after removal from the water storage was  $W_1$ . After measuring the specimens for weight gain, they were placed in a desiccator with fresh dried silica gel at an elevated temperature ( $90^\circ\text{C}$ ) and then weighed at an equal interval till until a constant weight was reached; this weight was the final weight  $W_2$ . Weight gains were measured by subtracting the original sample weight from the post-storage weight by the equation:  $W_1 - W_2$ . Water sorption percent  $W_{\text{SP}}(\%)$  and water solubility percent  $W_{\text{SL}}(\%)$  were calculated by the following equations:

$$W_{\text{SP}}(\%) = (W_1 - W_2) \times 100 / W_0$$

$$W_{\text{SL}}(\%) = (W_0 - W_2) \times 100 / W_0.$$

## RESULTS

## STATISTICAL METHODS

A full factorial two-way analysis of variance (ANOVA) model was used to model the effect of luting agent and time period on water sorption and solubility. Pair-wise comparisons were adjusted using Tukey's multiple comparison procedure. A significance level of 0.05 was used for all statistical tests.

Five observations were excluded from the analysis. These observations had values that were negative or that were extremely large or small and hence were considered to be outliers (Table IV).

## WATER SORPTION

There was a significant difference in water sorption among the nine luting materials ( $p < 0.001$ ), among the four time periods ( $p < 0.001$ ). There was a significant interaction between luting materials and time ( $p < 0.001$ ) (Table IX).

The resin-modified glass-ionomers showed the highest percentage of water sorption of all the luting materials tested. Rely X Luting Plus had the highest water sorption of all, followed by Fuji Cem, then Fuji Plus, with significant difference between each one of them (Table XI).

The resin-based luting agents showed the lowest percentage of water sorption. Rely X ARC had the lowest water sorption followed by Panavia F. Rely X ARC was significantly higher than Panavia F (Table XI).

The self-adhesive luting materials showed a varied range of water sorption results. Rely X Unicem had the lowest water sorption percentage among the self-adhesives and BisCem had the highest. There was significant difference among all the self-adhesives (Table XI).

Comparing all the luting materials water sorption results among the different time periods (7, 30, 90 and 180 days), a significant interaction between the luting materials and time was detected (Table IX). There was no significant difference between water sorption results observed over the periods of seven days and 30 days. Then, the water sorption results significantly increased with increasing storage time (90, 180 days) with no significant difference between the two storage periods (90, 180 days) (Table XIII).

There was a significant interaction between the luting materials and time. Rely X ARC, Panavia F, BisCem, Breeze and Maxcem Elite shows no significant change in their sorption with time changes resulting in a plot close to a straight line. In the other hand Rely X Unicem, Rely X Luting Plus, Fuji Cem and Fuji Plus shows significant increase in their sorption comparing the seven-day storage time and the 90-day storage time, but all reached a plateau during the time period from 90 days to 180 days (Table XVI).

## WATER SOLUBILITY

There was a significant difference in water solubility among the nine luting materials ( $p < 0.001$ ) and among the four time periods ( $p < 0.001$ ). There was a significant interaction between luting materials and time ( $p < 0.001$ ) (Table XXI).

Rely X Unicem followed by the resin-based luting materials observed the lowest water solubility result. Rely X ARC was significantly lower than Panavia F (Table XXIII).

The resin-modified glass-ionomer luting materials showed the highest water solubility results. FujiCem was the highest followed by Rely X Unicem then Fuji Plus, with significant differences among the three luting materials (Table XXIII).

The self-adhesive materials, except for Rely X Unicem, showed lower results compared with the resin-based luting agents, but still higher than the resin-based luting materials. All the self-adhesive materials were significantly different compared with each other, where Maxcem Elite showed the highest water solubility percentage among the self-adhesives followed by BisCem and Breeze (Table XXIII).

A significant interaction was observed between luting materials and time ( $p < 0.001$ ) (Table XXI). Water solubility increased with the increase in storage period. Water solubility results for each storage period were significantly different when compared with each other (Table XXV).

Figure 9 shows the mean water solubility by luting material and time period. Breeze, Panavia F and Rely X Unicem showed no significant changes in water solubility results with change in time, which resulted in a plot close to straight line. Maxcem Elite showed no significant changes in water solubility until the 90-day storage time, after which the solubility significantly increased during the time period from 90 days to 180 days. Fuji Cem, Rely X Luting, Fuji Plus, and BisCem showed significant increases in solubility at 90 days, reaching a peak. Then, the water solubility reached a plateau.

TABLES AND FIGURES



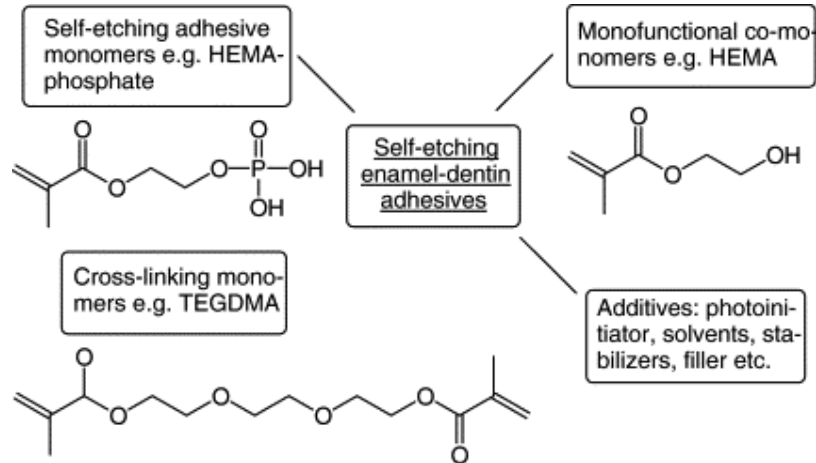


FIGURE 1. Components of currently available self-etching enamel-dentin adhesives.

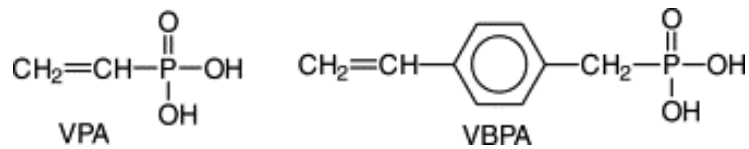


FIGURE 2. Structure of the monomeric phosphonic acids VPA and VPBA.

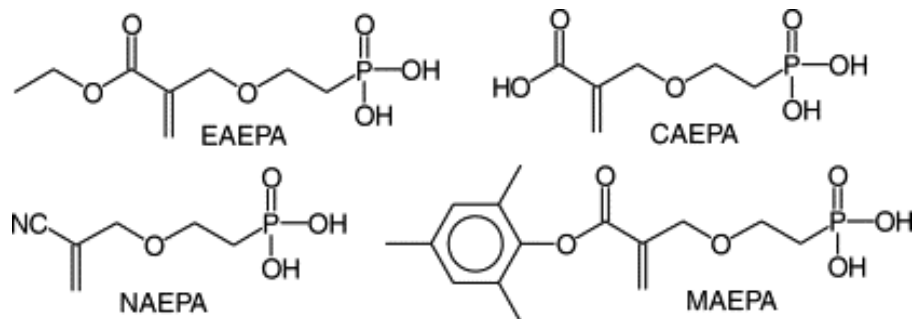


FIGURE 3. Structure of various acrylic ether phosphonic acids AEPA.

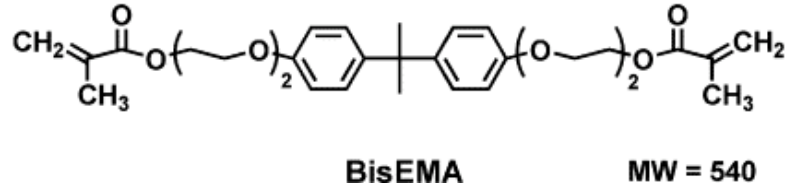
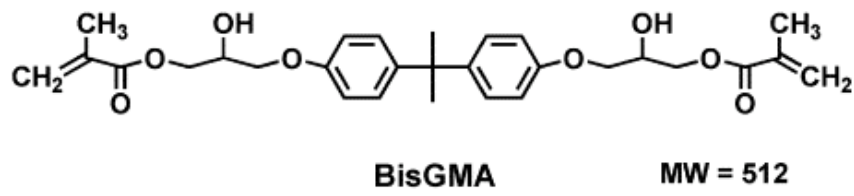
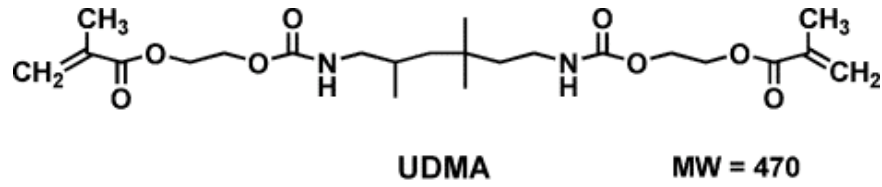


FIGURE 4. Chemical structure of commonly used dentin resin dimethacrylate monomers.

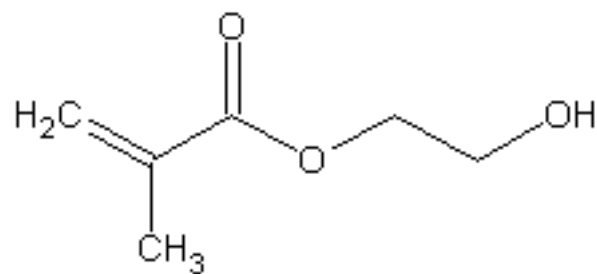


FIGURE 5. Chemical structure of HEMA 2-Hydroxyethyl methacrylate.

TABLE I

## Properties of different luting agents

PROPERTIES	IDEAL MATERIAL	Zinc phosphate	Glass Ionomer	Resin modified Ionomer	Composite Without the use of adhesives	Composite with adhesives
Film thickness ( $\mu\text{m}$ )	Low	>25	<25	>25	>25	<25
Working time (min)	Long	1.5-5	2-3.5	2-4	3-10	0.5-5
Setting time (min)	Short	5-14	6-9	2	3-7	1-15
Compressive strength (MPa)	High	62-101	122-162	40-141	194-200	179-255
Elastic Modulus (GPa)	Dentin =13.7 Enamel = 83 - 130	13.2	11.2	nt	17	4.5-9.8
Solubility	Very low	High	Low	Very Low	Very Low	Very low
Microleakage	Very low	High	Low to very high	Very low	High to very high	Very low to low
Removal of excess	Easy	Easy	Medium	Medium	Medium	Difficult
Retention	High	Moderate	Moderate to high	nt	Moderate	High

TABLE II

## List of the materials tested

LUTING MATERIALS	NATURE	COMPONENTS	MANUFACTURER
Panavia F (PF)	Resin Cement	Base: 10-MDP, 5-NMSA, silica, dimethacrylates, initiator. Catalyst: barium glass, sodium fluoride, dimethacrylates, BPO	Kuraray Medical Tokyo, Japan
Fuji I (FI)	Glass Ionomer		GC America, Illinois, USA
Rely X ARC (RA)	Resin Cement	Past A: BisGMA, TEGDMA, zirconia/silica filler 67.5% wt, dimethacrylate monomer. Past B: contain peroxide.	3M ESPE Seefeld, Germany
Rely X Unicem (RU)	Self-adhesive resin cement	Self-etch cement powder: glass powder, silica, calcium hydroxide, pigment, substitute pyrimidine and peroxy compound. (Filler load 72% wt, particles size < 9.5 $\mu\text{m}$ ) Liquid initiator: methacrylated phosphoric, dimethacrylate, acetate, stabilizer and initiator.	3M ESPE Seefeld, Germany
Rely X luting plus (RL)	Resin Modified Glass Ionomer Cement	Past A: fluoroaluminosilicate glass, proprietary reducing agent, HEMA, water, opacity. Past B: methacrylate polycarboxylic acid, BisGMA, HEMA, water, potassium persulfate, zirconia silica fillers.	3M ESPE Seefeld, Germany
Breez (BZ)	Self-adhesive resin cement	BisGMA, UDMA, TEGDMA, HEMA, 4-MET, barium glass, silica, BiOCl, Ca, Al, F	Pentron, Wallingford, CT, USA
Maxcem Elite (MX)	Self-adhesive resin cement	Glyceroldimethacrylate dihydrogen phosphate (GPDM), mono-, di-, tri-functional methacrylate monomers, self-cured redox initiator, photoinitiator (camphorquinone), stabilizer. (67% wt fillers, filler size 3.6 $\mu\text{m}$ ): barium glass, fluoroaluminosilicate and silica.	Kerr, Orange, CA, USA
BisCem	Self-adhesive resin cement	Bis (Hydroxyethylmethacrylate) 10-30%, Phosphate (base) 40-70%, Tetraethylene glycol, dimethacrylate, dental glass.	Bisco, Illinois, USA
FujiCem (FC)	Resin Modified Glass Ionomer Cement	(% Chemical components by WT and exposure limits) Distilled water 30-40%, Polyacrylic acid 30-40%, Benzenesulfonic acid sodium salt 2-3%, Silica powder 2%.	GC America, Illinois, USA
Fuji Plus (FP)	Resin Modified Glass Ionomer Cement	Powder: silica glass, Liquid: polyacrylic acid, 2-hydroxyethyl methacrylate, di-2-methacryloxyethyl-2, 2,4, trimethyl hexamethylene dicarbamate, tartaric acid.	GC America, Illinois, USA

TABLE III  
Manufacturers' instructions

MATERIALS	MANUFACTURERS' INSTRUCTIONS
Panavia F (PF) (Kuraray Medical, Tokyo, Japan)	<ol style="list-style-type: none"> <li>1. Dispense Equal amount of paste A and paste B.</li> <li>2. Mix sufficient paste A and paste B on the mixing plate for 20 seconds.</li> <li>3. Light cure for 20 sec.</li> </ol>
Fuji I (FI) (GC America, Illinois, USA)	<ol style="list-style-type: none"> <li>1. Powder and liquid dispensing: powder to liquid ratio is 1.8/1.0g. 1 level scoop of powder to 2 drops of liquid.</li> <li>2. . Mixing: Dispense powder and liquid onto the pad. Using the plastic spatula, add all the powder to the liquid. Mix rapidly for 20 seconds.</li> <li>3. Setting time is 4 minutes 30 seconds after start of mixing. Remove excess cement at the first formation of gel stage.</li> </ol>
Fuji-Plus (FP) (GC America, Illinois, USA)	<ol style="list-style-type: none"> <li>1. Powder and liquid dispensing: powder to liquid ratio is 2.0g / 1.0g. (1 level large scoop of powder to 3 drops of liquid.</li> <li>2. Mixing: Dispense powder and liquid onto the pad. Using the plastic spatula, add all the powder to the liquid. Mix rapidly for 20 seconds.</li> <li>3. Maintain isolation until set is verified (Approx. 4 minutes).</li> </ol>
FujiCem (FC) (GC America, Illinois, USA)	<p>- Cement supplied as a paste pack.</p> <ol style="list-style-type: none"> <li>1. Insert Paste Pak into dispenser and twist into position.</li> <li>2. Dispense desired amount of material.</li> <li>3. Mix with spatula for 10 seconds.</li> <li>4. Cement set in 3 min, after remove the excess.</li> </ol>
Rely X ARC (RA) (3M ESPE Seefeld, Germany)	<ol style="list-style-type: none"> <li>1. Apply and evenly distribute a thin layer of cement to the bonding surface of the indirect restoration.</li> <li>2. Setting time 3–5 min.</li> <li>3. Light cure for 40 seconds or allowed to self-cure for 10 minutes from start of mix.</li> </ol>
Rely X Unicem (RU) (3M ESPE Seefeld, Germany)	<ol style="list-style-type: none"> <li>1. Mix the 3M™ESPE™RelyX™Unicem Self-Adhesive Universal Resin Cement capsule in a high-frequency mixing unit (e.g. Capmix™) for 15 sec or in the Rotomix™capsule-mixing unit for 10 sec (see also the section on “Times”).</li> <li>2. Application: Insert the capsule in the Aplicap Applier after mixing and open the nozzle as far as possible. Protect the working area from water and saliva during application. Working time from the start of mixing 2 min.</li> <li>3. Light curing: 20 sec for each surface.</li> <li>4. Self-curing: set time after start of mixing 5 min.</li> </ol>
Rely X Luting Plus (RL) (3M ESPE Seefeld, Germany)	<ol style="list-style-type: none"> <li>1. Mixing: Using a cement spatula, mix the powder into the liquid. To minimize water evaporation and maximize working time, continue spatulation of the powder and liquid to a small area of the mixing pad. All of the powder should be incorporated into the liquid within 30 seconds.</li> <li>2. Working Time of the standard powder/liquid ratio is at least 2.5 minutes from the start of mix at a room temperature of 73°F (23°C).</li> </ol>
Breez (BZ) (Pentron, Wallingford, CT, USA)	<ol style="list-style-type: none"> <li>1. Dispense: Dispense Breeze™Cement directly into restoration.</li> <li>2. Place: Seat restoration.</li> <li>3. Cure: Light cure or self-cure.</li> </ol>
Maxcem Elite (MX) (Kerr, Orange, CA, USA)	<ol style="list-style-type: none"> <li>1. Dispensing the material</li> <li>2. Allow Maxcem to sit undisturbed for 1 1/2 minutes before light curing.</li> <li>3. Light-cure all surfaces including margins for 20 seconds*.</li> </ol>
BisCem (BC) (BISCO, Illinois, USA)	<ol style="list-style-type: none"> <li>1. Cement is supplied in a single syringe.</li> <li>2. Fill restoration with BisCem.</li> <li>3. Seat the restoration.</li> <li>4. Light cure for 3-5 seconds, to aid in cement removal.</li> <li>5. Excess cement is then easily removed.</li> <li>6. Light cure for 20-30 secon3.</li> </ol>

TABLE IV

Outliers

id	Agent	Period	W0	W1	W2	Wsp	Wsl
BC51	BC	7	377.20	398.60	377.70	5.54	-0.13
FC22	FC	90	394.60	325.00	274.70	12.75	30.39
FC25	FC	90	312.90	344.30	391.90	-15.21	-25.25
RL2	RL	180	290.00	332.00	374.10	-14.52	-29.00
RL30	RL	30	373.80	315.70	266.40	13.19	28.73

These samples gave extreme observation,  
far into the negative side or far high.

TABLE V

Water sorption by time, agent

Wsp - Water sorption (%)							
Time period (days)	Luting agent	N	Mean	Std Dev	Minimum	Maximum	Median
7	BC	12	6.51	0.28	6.21	7.08	6.4
	BZ	13	4.66	0.18	4.4	5.08	4.67
	FC	13	15.67	0.55	14.93	16.74	15.69
	FP	12	12.78	1.24	10.55	14.86	12.85
	MX	13	3.68	0.21	3.43	4.04	3.61
	PF	12	1.6	0.21	1.14	1.92	1.62
	RA	13	1.87	0.19	1.62	2.2	1.85
	RL	13	16.92	0.46	16.19	17.58	16.96
	RU	11	1.86	0.11	1.59	2.04	1.86
30	BC	13	6.55	0.21	6.26	6.93	6.54
	BZ	13	5.31	0.26	4.76	5.68	5.38
	FC	13	16.21	0.47	15.25	17.01	16.22
	FP	13	12.79	1.02	10.81	14.09	12.94
	MX	13	3.88	0.19	3.62	4.22	3.86
	PF	13	2.21	0.17	1.96	2.49	2.2
	RA	13	1.76	0.08	1.69	1.93	1.74
	RL	12	17.44	0.6	16.71	18.41	17.35
	RU	13	2.53	0.21	2.1	2.86	2.57
90	BC	13	6.69	0.27	5.97	7.03	6.68
	BZ	12	5.47	0.17	5.2	5.71	5.46
	FC	11	17.08	0.58	16.14	18.08	16.91
	FP	12	14.12	1.99	10.76	16.63	13.61
	MX	8	4.14	0.16	3.99	4.41	4.05
	PF	13	2.49	0.21	2.1	2.83	2.42
	RA	13	1.79	0.06	1.65	1.87	1.81
	RL	13	18.32	0.34	17.77	18.95	18.27
	RU	12	3.27	0.35	2.69	3.69	3.26
180	BC	13	6.59	0.17	6.44	6.96	6.52
	BZ	13	5.55	0.17	5.2	5.85	5.59
	FC	12	16.97	0.67	15.59	17.92	17.03
	FP	13	14.14	1.53	12.38	16.9	14.01
	MX	12	4.01	0.25	3.57	4.48	4
	PF	13	2.7	0.18	2.45	3.13	2.69
	RA	13	1.8	0.08	1.71	1.97	1.78
	RL	12	17.97	0.8	16.01	19.08	18.05
	RU	12	3.95	0.42	3.64	5.23	3.86

TABLE VI

Water sorption by agent, time

Wsp - Water sorption (%)							
Luting agent	Time period (days)	N	Mean	Std Dev	Minimum	Maximum	Median
BC	7	12	6.51	0.28	6.21	7.08	6.4
	30	13	6.55	0.21	6.26	6.93	6.54
	90	13	6.69	0.27	5.97	7.03	6.68
	180	13	6.59	0.17	6.44	6.96	6.52
BZ	7	13	4.66	0.18	4.4	5.08	4.67
	30	13	5.31	0.26	4.76	5.68	5.38
	90	12	5.47	0.17	5.2	5.71	5.46
	180	13	5.55	0.17	5.2	5.85	5.59
FC	7	13	15.67	0.55	14.93	16.74	15.69
	30	13	16.21	0.47	15.25	17.01	16.22
	90	11	17.08	0.58	16.14	18.08	16.91
	180	12	16.97	0.67	15.59	17.92	17.03
FP	7	12	12.78	1.24	10.55	14.86	12.85
	30	13	12.79	1.02	10.81	14.09	12.94
	90	12	14.12	1.99	10.76	16.63	13.61
	180	13	14.14	1.53	12.38	16.9	14.01
MX	7	13	3.68	0.21	3.43	4.04	3.61
	30	13	3.88	0.19	3.62	4.22	3.86
	90	8	4.14	0.16	3.99	4.41	4.05
	180	12	4.01	0.25	3.57	4.48	4
PF	7	12	1.6	0.21	1.14	1.92	1.62
	30	13	2.21	0.17	1.96	2.49	2.2
	90	13	2.49	0.21	2.1	2.83	2.42
	180	13	2.7	0.18	2.45	3.13	2.69
RA	7	13	1.87	0.19	1.62	2.2	1.85
	30	13	1.76	0.08	1.69	1.93	1.74
	90	13	1.79	0.06	1.65	1.87	1.81
	180	13	1.8	0.08	1.71	1.97	1.78
RL	7	13	16.92	0.46	16.19	17.58	16.96
	30	12	17.44	0.6	16.71	18.41	17.35
	90	13	18.32	0.34	17.77	18.95	18.27
	180	12	17.97	0.8	16.01	19.08	18.05
RU	7	11	1.86	0.11	1.59	2.04	1.86
	30	13	2.53	0.21	2.1	2.86	2.57
	90	12	3.27	0.35	2.69	3.69	3.26
	180	12	3.95	0.42	3.64	5.23	3.86



TABLE VII

Class level information

Class	Levels	Values
trt	9	BC BZ FC FP MX PF RA RL RU
period	4	7 30 90 180

TABLE VIII

Number of observations

Number of observations read	448
Number of observations used	448

TABLE IX

ANOVA table for water sorption

Source	DF	Type III SS	Mean Square	F Value	Pr > F
trt	8	15786.94212	1973.36777	5674.98	<.0001
period	3	62.87163	20.95721	60.27	<.0001
trt*period	24	36.43582	1.51816	4.37	<.0001

TABLE X

Model summary – water sorption

R-Square	Coeff Var	Root MSE	wsp Mean
0.991077	7.561339	0.589688	7.798718

TABLE XI

Means with the same letter are not significantly different

Tukey Grouping	Mean	N	trt
A	4.83	49	FC
B	3.25	50	RL
C	1.99	50	FP
D	1.11	46	MX
E	0.94	51	BC
E	0.93	51	BZ
F	0.67	51	PF
G	0.46	52	RA
H	0.13	48	RU

TABLE XII

Details for luting agent comparisons – water sorption

Alpha	0.05
Error Degrees of Freedom	412
Error Mean Square	0.347731
Critical Value of Studentized Range	4.40996
Minimum Significant Difference	0.3688
Harmonic Mean of Cell Sizes	49.71416

TABLE XIII

Means with the same letter are not significantly different

Tukey Grouping	Mean	N	period
A	8.19	113	90
A	8.10	107	180
B	7.55	116	30
B	7.39	112	7

TABLE XIV

Details for time period comparison – water sorption

Alpha	0.05
Error Degrees of Freedom	412
Error Mean Square	0.060419
Critical Value of Studentized Range	3.64808
Minimum Significant Difference	0.0848
Harmonic Mean of Cell Sizes	111.905

TABLE XV

## Water sorption comparison within agents

Pair-wise comparisons within luting agent; Tukey adjusted p-values							
trt	period	Mean	period	7	30	90	180
BC	7	6.51	7		1.0000	1.0000	1.0000
BC	30	6.55	30	1.0000		1.0000	1.0000
BC	90	6.69	90	1.0000	1.0000		1.0000
BC	180	6.59	180	1.0000	1.0000	1.0000	
BZ	7	4.66	7		0.6276	0.1846	0.0533
BZ	30	5.31	30	0.6276		1.0000	1.0000
BZ	90	5.47	90	0.1846	1.0000		1.0000
BZ	180	5.55	180	0.0533	1.0000	1.0000	
FC	7	15.67	7		0.9205	<.0001	<.0001
FC	30	16.21	30	0.9205		0.1197	0.2991
FC	90	17.08	90	<.0001	0.1197		1.0000
FC	180	16.97	180	<.0001	0.2991	1.0000	
FP	7	12.78	7		1.0000	<.0001	<.0001
FP	30	12.79	30	1.0000		<.0001	<.0001
FP	90	14.12	90	<.0001	<.0001		1.0000
FP	180	14.14	180	<.0001	<.0001	1.0000	
MX	7	3.68	7		1.0000	0.9990	1.0000
MX	30	3.88	30	1.0000		1.0000	1.0000
MX	90	4.14	90	0.9990	1.0000		1.0000
MX	180	4.01	180	1.0000	1.0000	1.0000	
PF	7	1.60	7		0.8105	0.0767	0.0025
PF	30	2.21	30	0.8105		1.0000	0.9786
PF	90	2.49	90	0.0767	1.0000		1.0000
PF	180	2.70	180	0.0025	0.9786	1.0000	
RA	7	1.87	7		1.0000	1.0000	1.0000
RA	30	1.76	30	1.0000		1.0000	1.0000
RA	90	1.79	90	1.0000	1.0000		1.0000
RA	180	1.80	180	1.0000	1.0000	1.0000	
RL	7	16.92	7		0.9603	<.0001	0.0058
RL	30	17.44	30	0.9603		0.0811	0.9683
RL	90	18.32	90	<.0001	0.0811		1.0000
RL	180	17.97	180	0.0058	0.9683	1.0000	
RU	7	1.86	7		0.6757	<.0001	<.0001
RU	30	2.53	30	0.6757		0.3584	<.0001
RU	90	3.27	90	<.0001	0.3584		0.6029
RU	180	3.95	180	<.0001	<.0001	0.6029	
RL	30	17.44	30	0.9603		0.0811	0.9683

TABLE XVI

Water sorption comparison within period

Pair-wise comparisons within period; Tukey adjusted p-values												
period	trt	Mean	i/j	BC	BZ	FC	FP	MX	PF	RA	RL	RU
7	BC	6.51	BC		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
7	BZ	4.66	BZ	<.0001		<.0001	<.0001	0.0136	<.0001	<.0001	<.0001	<.0001
7	FC	15.67	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
7	FP	12.78	FP	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001
7	MX	3.68	MX	<.0001	0.0136	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001
7	PF	1.60	PF	<.0001	<.0001	<.0001	<.0001	<.0001		1.0000	<.0001	1.0000
7	RA	1.87	RA	<.0001	<.0001	<.0001	<.0001	<.0001	1.0000		<.0001	1.0000
7	RL	16.92	RL	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001		<.0001
7	RU	1.86	RU	<.0001	<.0001	<.0001	<.0001	<.0001	1.0000	1.0000	<.0001	
30	BC	6.55	BC		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
30	BZ	5.31	BZ	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
30	FC	16.21	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	0.0002	<.0001
30	FP	12.79	FP	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001
30	MX	3.88	MX	<.0001	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001
30	PF	2.21	PF	<.0001	<.0001	<.0001	<.0001	<.0001		0.9948	<.0001	1.0000
30	RA	1.76	RA	<.0001	<.0001	<.0001	<.0001	<.0001	0.9948		<.0001	0.2593
30	RL	17.44	RL	<.0001	<.0001	0.0002	<.0001	<.0001	<.0001	<.0001		<.0001
30	RU	2.53	RU	<.0001	<.0001	<.0001	<.0001	<.0001	1.0000	0.2593	<.0001	
90	BC	6.69	BC		0.0002	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
90	BZ	5.47	BZ	0.0002		<.0001	<.0001	0.0007	<.0001	<.0001	<.0001	<.0001
90	FC	17.08	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	0.0002	<.0001
90	FP	14.12	FP	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001
90	MX	4.14	MX	<.0001	0.0007	<.0001	<.0001		<.0001	<.0001	<.0001	0.2814
90	PF	2.49	PF	<.0001	<.0001	<.0001	<.0001	<.0001		0.4539	<.0001	0.2485
90	RA	1.79	RA	<.0001	<.0001	<.0001	<.0001	<.0001	0.4539		<.0001	<.0001
90	RL	18.32	RL	<.0001	<.0001	0.0002	<.0001	<.0001	<.0001	<.0001		<.0001
90	RU	3.27	RU	<.0001	<.0001	<.0001	<.0001	0.2814	0.2485	<.0001	<.0001	
180	BC	6.59	BC		0.0041	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
180	BZ	5.55	BZ	0.0041		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
180	FC	16.97	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	0.0189	<.0001
180	FP	14.14	FP	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001
180	MX	4.01	MX	<.0001	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	1.0000
180	PF	2.70	PF	<.0001	<.0001	<.0001	<.0001	<.0001		0.0438	<.0001	0.0001
180	RA	1.80	RA	<.0001	<.0001	<.0001	<.0001	<.0001	0.0438		<.0001	<.0001
180	RL	17.97	RL	<.0001	<.0001	0.0189	<.0001	<.0001	<.0001	<.0001		<.0001
180	RU	3.95	RU	<.0001	<.0001	<.0001	<.0001	1.0000	0.0001	<.0001	<.0001	

TABLE XVII

Water solubility by time, agent

Wsl - Water solubility (%)							
Time period (days)	Luting agent	N	Mean	Std Dev	Minimum	Maximum	Median
7	BC	12	0.62	0.23	0.48	1.32	0.57
	BZ	13	0.75	0.05	0.66	0.82	0.74
	FC	13	2.29	0.26	1.61	2.62	2.32
	FP	12	0.97	0.12	0.83	1.23	0.94
	MX	13	0.75	0.1	0.56	0.87	0.78
	PF	12	0.42	0.13	0.26	0.65	0.41
	RA	13	0.74	0.22	0.43	1.17	0.72
	RL	13	1.88	0.3	1.57	2.46	1.85
	RU	11	0.05	0.05	0	0.18	0.05
30	BC	13	0.73	0.07	0.66	0.91	0.71
	BZ	13	0.83	0.08	0.69	0.98	0.82
	FC	13	3.24	0.79	2.3	5.52	3.14
	FP	13	1.07	0.13	0.89	1.26	1.05
	MX	13	1.04	0.13	0.82	1.22	1.08
	PF	13	0.56	0.16	0.35	0.87	0.54
	RA	13	0.55	0.13	0.37	0.86	0.55
	RL	12	2.28	0.21	2.02	2.65	2.22
	RU	13	0.07	0.03	0.02	0.13	0.07
90	BC	13	1.27	0.17	1.04	1.77	1.22
	BZ	12	0.99	0.09	0.86	1.15	1
	FC	11	6.72	0.46	6.13	7.55	6.63
	FP	12	2.78	0.37	2.23	3.27	2.76
	MX	8	1.16	0.11	0.97	1.3	1.18
	PF	13	0.82	0.14	0.67	1.08	0.78
	RA	13	0.31	0.04	0.22	0.36	0.31
	RL	13	4.42	0.21	4.13	4.83	4.48
	RU	12	0.16	0.05	0.07	0.24	0.16
180	BC	13	1.12	0.12	0.92	1.26	1.15
	BZ	13	1.15	0.13	0.91	1.35	1.18
	FC	12	7.59	0.56	6.65	8.66	7.65
	FP	13	3.11	0.38	2.49	3.72	3.13
	MX	12	1.55	0.12	1.41	1.71	1.52
	PF	13	0.85	0.18	0.64	1.3	0.8
	RA	13	0.23	0.02	0.2	0.29	0.23
	RL	12	4.45	0.34	3.91	5.21	4.42
	RU	12	0.22	0.05	0.16	0.3	0.22

TABLE XVIII

Water solubility by agent, time

Wsl - Water solubility (%)							
Luting agent	Time period (days)	N	Mean	Std Dev	Minimum	Maximum	Median
BC	7	12	0.62	0.23	0.48	1.32	0.57
	30	13	0.73	0.07	0.66	0.91	0.71
	90	13	1.27	0.17	1.04	1.77	1.22
	180	13	1.12	0.12	0.92	1.26	1.15
BZ	7	13	0.75	0.05	0.66	0.82	0.74
	30	13	0.83	0.08	0.69	0.98	0.82
	90	12	0.99	0.09	0.86	1.15	1
	180	13	1.15	0.13	0.91	1.35	1.18
FC	7	13	2.29	0.26	1.61	2.62	2.32
	30	13	3.24	0.79	2.3	5.52	3.14
	90	11	6.72	0.46	6.13	7.55	6.63
	180	12	7.59	0.56	6.65	8.66	7.65
FP	7	12	0.97	0.12	0.83	1.23	0.94
	30	13	1.07	0.13	0.89	1.26	1.05
	90	12	2.78	0.37	2.23	3.27	2.76
	180	13	3.11	0.38	2.49	3.72	3.13
MX	7	13	0.75	0.1	0.56	0.87	0.78
	30	13	1.04	0.13	0.82	1.22	1.08
	90	8	1.16	0.11	0.97	1.3	1.18
	180	12	1.55	0.12	1.41	1.71	1.52
PF	7	12	0.42	0.13	0.26	0.65	0.41
	30	13	0.56	0.16	0.35	0.87	0.54
	90	13	0.82	0.14	0.67	1.08	0.78
	180	13	0.85	0.18	0.64	1.3	0.8
RA	7	13	0.74	0.22	0.43	1.17	0.72
	30	13	0.55	0.13	0.37	0.86	0.55
	90	13	0.31	0.04	0.22	0.36	0.31
	180	13	0.23	0.02	0.2	0.29	0.23
RL	7	13	1.88	0.3	1.57	2.46	1.85
	30	12	2.28	0.21	2.02	2.65	2.22
	90	13	4.42	0.21	4.13	4.83	4.48
	180	12	4.45	0.34	3.91	5.21	4.42
RU	7	11	0.05	0.05	0	0.18	0.05
	30	13	0.07	0.03	0.02	0.13	0.07
	90	12	0.16	0.05	0.07	0.24	0.16
	180	12	0.22	0.05	0.16	0.3	0.22

TABLE XIX

Class level information

Class	Levels	Values
trt	9	BC BZ FC FP MX PF RA RL RU
period	4	7 30 90 180

TABLE XX

Number of observations

Number of Observations Read	448
Number of Observations Used	448

TABLE XXI

ANOVA table for water solubility

Source	DF	Type III SS	Mean Square	F Value	Pr > F
trt	8	966.4730054	120.8091257	1999.53	<.0001
period	3	142.9166673	47.6388891	788.48	<.0001
trt*period	24	236.2180614	9.8424192	162.9	<.0001

TABLE XXII

Model summary – water solubility

R-Square	Coeff Var	Root MSE	wsl Mean
0.981322	15.52391	0.245802	1.583379

TABLE XXIII

Means with the same letter are not significantly different

Tukey Grouping	Mean	N	trt
A	4.83	49	FC
B	3.25	50	RL
C	1.99	50	FP
D	1.11	46	MX
E	0.94	51	BC
E	0.93	51	BZ
F	0.67	51	PF
G	0.46	52	RA
H	0.13	48	RU

TABLE XXIV

Details for luting agent comparisons – water solubility

Alpha	0.05
Error Degrees of Freedom	412
Error Mean Square	0.060419
Critical Value of Studentized Range	4.40996
Minimum Significant Difference	0.1537
Harmonic Mean of Cell Sizes	49.71416

TABLE XXV

Means with the same letter are not significantly different

Tukey Grouping	Mean	N	period	Tukey Grouping
A	2.21	113	180	A
B	2.05	107	90	B
C	1.14	116	30	C
D	0.96	112	7	D

TABLE XXVI

Details for time period comparison – water solubility

Alpha	0.05
Error Degrees of Freedom	412
Error Mean Square	0.060419
Critical Value of Studentized Range	3.64808
Minimum Significant Difference	0.0848
Harmonic Mean of Cell Sizes	111.905



TABLE XXVII

Water solubility comparison within agents

Pair-wise comparisons within luting agent; Tukey adjusted p-values							
trt	period	Mean	i/j	7	30	90	180
BC	7	0.62	7		1.0000	<.0001	0.0005
BC	30	0.73	30	1.0000		<.0001	0.0356
BC	90	1.27	90	<.0001	<.0001		0.9999
BC	180	1.12	180	0.0005	0.0356	0.9999	
BZ	7	0.75	7		1.0000	0.8547	0.0194
BZ	30	0.83	30	1.0000		0.9996	0.2593
BZ	90	0.99	90	0.8547	0.9996		0.9999
BZ	180	1.15	180	0.0194	0.2593	0.9999	
FC	7	2.29	7		<.0001	<.0001	<.0001
FC	30	3.24	30	<.0001		<.0001	<.0001
FC	90	6.72	90	<.0001	<.0001		<.0001
FC	180	7.59	180	<.0001	<.0001	<.0001	
FP	7	0.97	7		1.0000	<.0001	<.0001
FP	30	1.07	30	1.0000		<.0001	<.0001
FP	90	2.78	90	<.0001	<.0001		0.2338
FP	180	3.11	180	<.0001	<.0001	0.2338	
MX	7	0.75	7		0.4821	0.0844	<.0001
MX	30	1.04	30	0.4821		1.0000	0.0002
MX	90	1.16	90	0.0844	1.0000		0.1569
MX	180	1.55	180	<.0001	0.0002	0.1569	
PF	7	0.42	7		1.0000	0.0227	0.0076
PF	30	0.56	30	1.0000		0.7204	0.4845
PF	90	0.82	90	0.0227	0.7204		1.0000
PF	180	0.85	180	0.0076	0.4845	1.0000	
RA	7	0.74	7		0.9907	0.0051	0.0001
RA	30	0.55	30	0.9907		0.8676	0.2773
RA	90	0.31	90	0.0051	0.8676		1.0000
RA	180	0.23	180	0.0001	0.2773	1.0000	
RL	7	1.88	7		0.0230	<.0001	<.0001
RL	30	2.28	30	0.0230		<.0001	<.0001
RL	90	4.42	90	<.0001	<.0001		1.0000
RL	180	4.45	180	<.0001	<.0001	1.0000	
RU	7	0.05	7		1.0000	1.0000	0.9998
RU	30	0.07	30	1.0000		1.0000	0.9999
RU	90	0.16	90	1.0000	1.0000		1.0000
RU	180	0.22	180	0.9998	0.9999	1.0000	

TABLE XXVIII

## Water solubility comparison within storage periods

Pair-wise comparisons within period; Tukey adjusted p-values												
period	trt	Mean	i/j	BC	BZ	FC	FP	MX	PF	RA	RL	RU
7	BC	0.62	BC		1.0000	<.0001	0.1642	1.0000	0.9883	1.0000	<.0001	<.0001
7	BZ	0.75	BZ	1.0000		<.0001	0.9440	1.0000	0.2342	1.0000	<.0001	<.0001
7	FC	2.29	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	0.0116	<.0001
7	FP	0.97	FP	0.1642	0.9440	<.0001		0.9511	<.0001	0.9197	<.0001	<.0001
7	MX	0.75	MX	1.0000	1.0000	<.0001	0.9511		0.2201	1.0000	<.0001	<.0001
7	PF	0.42	PF	0.9883	0.2342	<.0001	<.0001	0.2201		0.2777	<.0001	0.1237
7	RA	0.74	RA	1.0000	1.0000	<.0001	0.9197	1.0000	0.2777		<.0001	<.0001
7	RL	1.88	RL	<.0001	<.0001	0.0116	<.0001	<.0001	<.0001	<.0001		<.0001
7	RU	0.05	RU	<.0001	<.0001	<.0001	<.0001	<.0001	0.1237	<.0001	<.0001	
30	BC	0.73	BC		1.0000	<.0001	0.1646	0.3417	0.9989	0.9953	<.0001	<.0001
30	BZ	0.83	BZ	1.0000		<.0001	0.8624	0.9695	0.6663	0.5414	<.0001	<.0001
30	FC	3.24	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
30	FP	1.07	FP	0.1646	0.8624	<.0001		1.0000	0.0001	<.0001	<.0001	<.0001
30	MX	1.04	MX	0.3417	0.9695	<.0001	1.0000		0.0007	0.0003	<.0001	<.0001
30	PF	0.56	PF	0.9989	0.6663	<.0001	0.0001	0.0007		1.0000	<.0001	0.0003
30	RA	0.55	RA	0.9953	0.5414	<.0001	<.0001	0.0003	1.0000		<.0001	0.0006
30	RL	2.28	RL	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001		<.0001
30	RU	0.07	RU	<.0001	<.0001	<.0001	<.0001	<.0001	0.0003	0.0006	<.0001	
90	BC	1.27	BC		0.6318	<.0001	<.0001	1.0000	0.0025	<.0001	<.0001	<.0001
90	BZ	0.99	BZ	0.6318		<.0001	<.0001	1.0000	0.9992	<.0001	<.0001	<.0001
90	FC	6.72	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
90	FP	2.78	FP	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001
90	MX	1.16	MX	1.0000	1.0000	<.0001	<.0001		0.4279	<.0001	<.0001	<.0001
90	PF	0.82	PF	0.0025	0.9992	<.0001	<.0001	0.4279		0.0001	<.0001	<.0001
90	RA	0.31	RA	<.0001	<.0001	<.0001	<.0001	<.0001	0.0001		<.0001	1.0000
90	RL	4.42	RL	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001		<.0001
90	RU	0.16	RU	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	1.0000	<.0001	
180	BC	1.12	BC		1.0000	<.0001	<.0001	0.0060	0.6771	<.0001	<.0001	<.0001
180	BZ	1.15	BZ	1.0000		<.0001	<.0001	0.0208	0.4133	<.0001	<.0001	<.0001
180	FC	7.59	FC	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001	<.0001
180	FP	3.11	FP	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001	<.0001
180	MX	1.55	MX	0.0060	0.0208	<.0001	<.0001		<.0001	<.0001	<.0001	<.0001
180	PF	0.85	PF	0.6771	0.4133	<.0001	<.0001	<.0001		<.0001	<.0001	<.0001
180	RA	0.23	RA	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001		<.0001	1.0000
180	RL	4.45	RL	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001		<.0001
180	RU	0.22	RU	<.0001	<.0001	<.0001	<.0001	<.0001	<.0001	1.0000	<.0001	

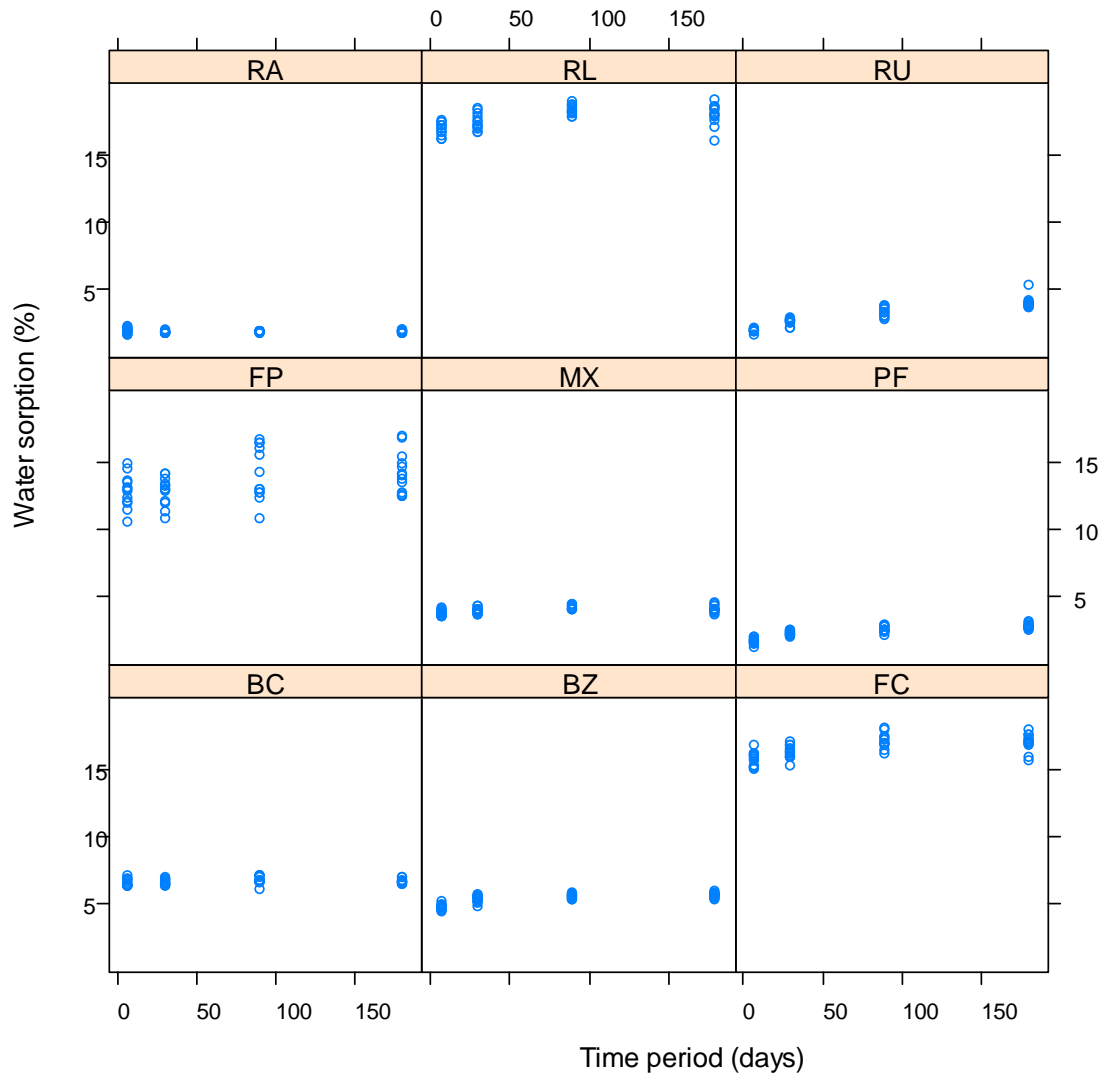


FIGURE 6. Water sorption.

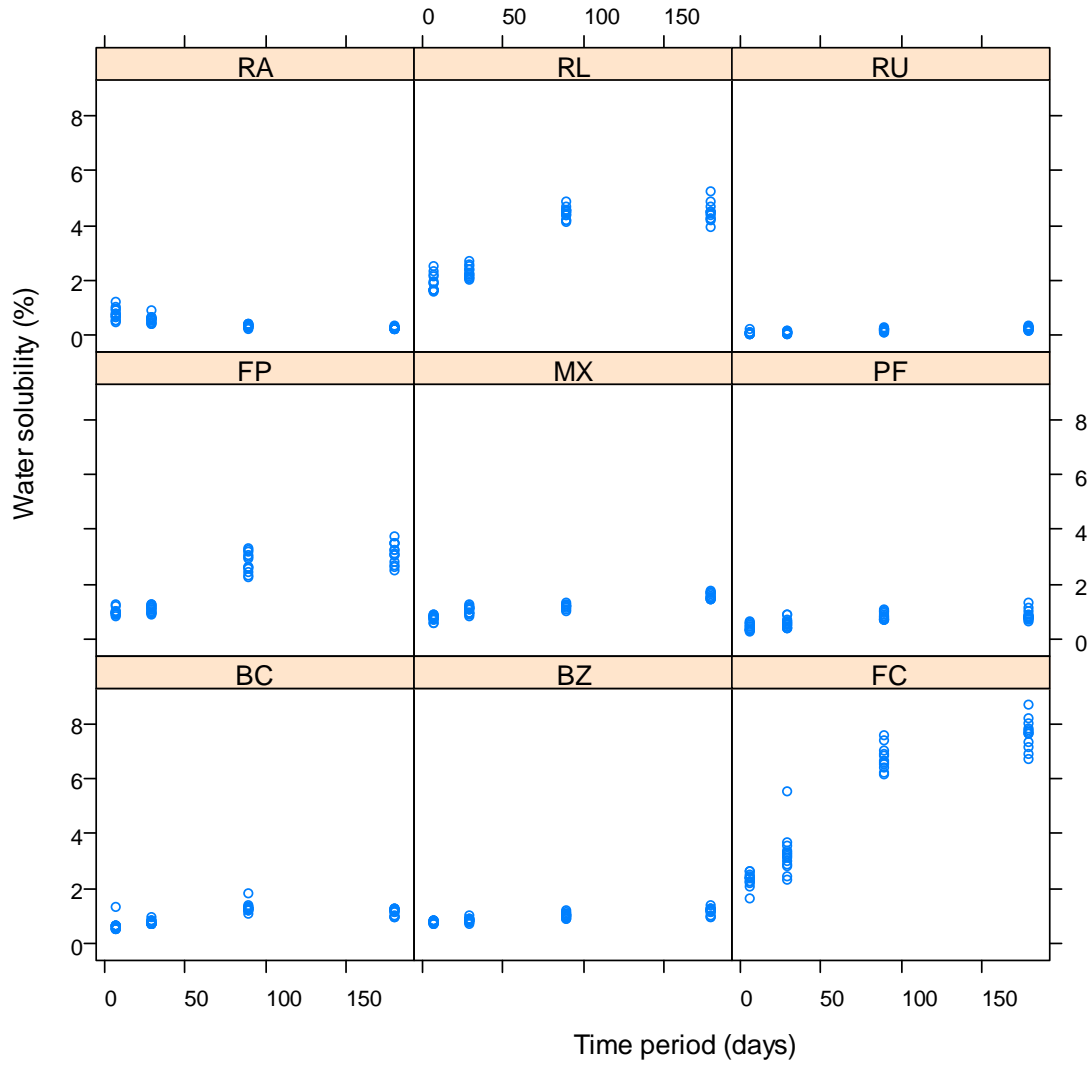


FIGURE 7. Water solubility.

FIGURE 8. Mean water sorption by luting agent and time period.

FIGURE 9. Mean water solubility by luting agent and time period.

## DISCUSSION

Water solubility and sorption behavior of luting materials is highly critical and cannot be neglected. Water can lead to a decrease in the mechanical properties of the luting agent as well as degradation of the bond between the luting agent and the restoration and the tooth.<sup>81-84, 86, 87</sup> This study was conducted following the ISO Specification No. 4049.

The glass-ionomer luting material represented by Fuji I could not be tested; the samples were fractured or chipped even before the first desiccating procedure. When the samples were placed in a humidifier for the first 24 hrs, samples survived the preparation but not the desiccation. This may be due to the small thickness of the discs prepared.

There were five sample observations excluded from the statistical analysis (Table IV). For the Fuji Cem and Rely X Luting excluded samples, the extreme results can be explained as a result of mixing errors. Both materials were hand-mixed during the preparation (Table III). It is well documented in the literature that dental luting materials properties can be affected by mixing errors.<sup>89, 90</sup>

After reviewing the results, both null hypotheses, which proposed that there is no difference in water sorption/solubility between the different luting materials, were rejected.

The resin-modified glass-ionomers showed the highest percentage of water sorption. As the resin-modified glass-ionomer undergoes its setting reaction, the net result is a multi-phase structure featuring hydrophilic (HEMA)/ionic components. The



boundaries between the different components are believed to be the sites where the water uptake and retention occurs during the water-storage periods. It has been argued that the water uptake may be beneficial to compensate for the setting shrinkage and reduce resulting stresses.<sup>8</sup> But the resin-modified glass-ionomers in this study showed water sorption percentages of 13.46 percent to 17.66 percent by weight, which is quite high. High water sorption results in hygroscopic expansion, which can explain the cracked and fractured resin-modified glass-ionomer samples observed during the water-storage period. This hygroscopic expansion exerts residual stresses on both the tooth and the restoration leading to post-operative sensitivity. These results agree with the majority of published literature.<sup>84, 85</sup>

FujiCem showed significantly higher sorption percentages compared with Fuji Plus, although they are from the same manufacturer (Table XI). This behavior can be attributed to the different chemical structure of both luting materials. FujiCem is made of the simplest form of the resin-modified glass-ionomer materials; some of the water content is replaced with polyacrylic acid (30 percent to 40 percent). The setting reaction is of two parts, a slow acid-base reaction similar to that of the conventional glass-ionomer cements and a photo-initiated and co-polymerization reaction of the methacrylate group of the HEMA.<sup>91</sup> Fuji Plus is of more complicated chemistry, in addition to the two reactions previously mentioned, a third polymerization initiation occurs through chemically initiated free radicals of the more complicated polymeric liquid.<sup>37, 92, 93</sup> Table II shows the chemical composition of both luting agents. This third reaction leads to a more complicated net structure with more linked polymeric chains and may explain the lower percent of water sorption observed by Fuji Plus. The water sorption results of Fuji

Plus is high, compared with the self-adhesive and the resin-based luting materials, and in the end, it still has the same multi-phase structure of a resin-modified glass-ionomer (Table XI).

For the self-adhesive luting materials, the results showed a wide range. The self-adhesive showed significantly higher sorption percentages compared with the conventional resin-based luting materials (Table XI). This was expected due to the chemistry of such materials; they are expected to be somewhat more hydrophilic, improving its wettability and penetration into the tooth structures. They become less hydrophilic and more hydrophobic during the polymerization reaction. But, compared with the conventional resin-based luting materials, they experience inferior hydrolytic stability.<sup>54</sup> The difference between the materials can be attributed to the difference in the type of resin matrix used or the filler content. An increase in the filler content may lead to a decrease in the sorption/solubility.

The filler content for all the self-adhesive luting materials were not available, so that it is not really clear if there was any impact of the filler content between the different materials. The information was available for Rely X Unicem (72% wt) and Maxcem Elite (67% wt); Maxcem Elite water sorption were significantly higher than Rely X Unicem (Table II).

Covering the resin matrix composition of the self-adhesive luting materials, UDMA (Figure 4) polymers show significantly more water uptake than polymers based on non-hydroxylated Bis-GMA analogues.<sup>94</sup> HEMA (Figure 5) polymers also have a more hydrophilic portion that leads to more water sorption compared with the BisGMA polymers. It was found that water could induce stresses between the different phases of

the final matrix leading to the formation of small cracks at these regions. These cracks could act as channels for more water diffusion and later formation of water pools.<sup>93</sup>

Figure 8 shows the mean water sorption by luting agent and time period. The resin and self-adhesives (except for Rely X Unicem) water sorption did not change with increase in storage time. Rely X Unicem sorption results kept increasing, reaching a significant peak at 90 days, after which no significant change in the sorption was detected. The resin-modified glass-ionomers all behaved the same, reaching a peak at 90 days, then a plateau after that. All the luting materials tested showed no changes in water sorption after the 90-day storage time, which means that most of the water sorption occurred during the first 90 days (Table XV).

BisCem started with a sorption significantly higher than the rest of the resin and the self-adhesive luting agents at 7- and 30-day storage time. After that, BisCem sorption was not different than Breeze for 90- and 180-day storage time. MaxCem and Breeze sorption were indifferent at 30 days, and then Breeze showed a significantly higher sorption than Maxcem. Rely X Unicem started with a sorption similar to that of Panavia and Rely X ARC for the 7- and 30-day storage time. At 90 days Rely X Unicem sorption was significantly higher compared with Rely X ARC but not different when compared with Panavia and Maxcem. At 180 days, Rely X U not different when compared with Panavia and Rely X ARC. Panavia and Rely X ARC sorption results were not significantly different. From these results, MaxCem elite is the most comparable self-adhesive to resin luting agents when considered for sorption. Rely X Unicem starts with sorption close to that of the resin luting agents; however, the plot showed rapid increase in the sorption especially at the 90-day storage time. Although Rely X Unicem expressed

no change in sorption percentages for the period from 90 to 180 day, it is still not clear if the sorption will increase after 180 day, considering the way the plot is going (Table XV - XVI).

The resin-modified glass-ionomers had different sorption in the beginning, starting from the 30-day storage period. Both Rely X Luting and Fuji Cem continue to have close water sorption, which are not significantly different. Fuji Plus shows the lower sorption compared with Rely X Luting and Fuji Cem (Table XVI).

There is no relation between water sorption behavior and water solubility. Both are two separate behaviors of a material; high sorption does not indicate high solubility and vice versa.

The resin-modified glass-ionomers showed the greatest water solubility, in particular FujiCem (4.83 percent) followed by Rely X Luting Plus (3.25 percent). The setting reaction of these materials make them less likely to have un-polymerized monomers leaching into solution as the polymerizable monomers are linked to the carboxylic acid groups in the matrix. But due to the hydrophilicity of such materials, they are more likely to be more soluble compared with less hydrophilic materials.<sup>95</sup> Fuji Plus was significantly less soluble than the two other resin-modified glass-ionomers. This may be explained by the setting reaction mentioned above, which will lead to a more highly cross-linked matrix. Fuji Cem was significantly more soluble at all the storage periods compared with Rely X Luting Plus.

Figure 9 shows the plotted water solubility means of the different luting materials against the change in time. The resin-modified glass-ionomers express the highest water solubility increase with time reaching its peak at 90 days. Rely X Luting and Fuji Plus

both reached a plateau for the period of 180 days. Fuji Cem solubility significantly increased with each time period and continued to increase until the 180-day storage period. Breeze, Panavia F and Rely X Unicem showed no significant change in solubility with the time change. Rely X Unicem had the lowest solubility results. Rely X ARC was not significantly different compared with Rely X Unicem for all the storage time periods. Panavia F was not significantly different compared with both Rely X Unicem and Rely X ARC for 7-, 30- and 90-day storage periods. At 180 days Panavia F solubility was significantly higher than Rely X Unicem and Rely X ARC (Table XXVII - XXVIII).

Maxcem Elite showed no change in water solubility during the 7-, 30- and 90-day time periods. During these time periods, Maxcem was not significantly different than Panavia F. Maxcem solubility significantly increased for the period of 180 days. The behavior of Maxcem Elite after 180 days of storage could not be predicted as to whether it would continue in increasing or reached a plateau. In the other hand, Rely X ARC showed no changes in its solubility during the first 90 days, then its solubility significantly decreased. Breeze, BisCem and Maxcem sorption were not significantly different compared with each other during the different storage periods; they were not significantly different compared with Panavia F during the 7-, 30- and 90-day storage periods. Breeze and BisCem solubility were not different from Panavia at 180 days, but Maxcem was significantly higher than Panavia at 180 days. Fuji Plus started having solubility close to the self-adhesives during the 7- and 30-day storage periods, then significantly increased (Table XXVIII).

Self-adhesive luting materials showed acceptable solubility comparable to the resin luting materials with the exception of Maxcem Elite, which exhibited a higher

solubility after 90 days. Rely X Unicem showed an unexpected solubility lower than both resin materials used in this study. At 180 days, both Rely X Unicem and Rely X ARC showed similar solubility. However, the behavior of Rely X ARC cannot be predicted after 180 days; it could decrease or increase beyond this point (Table XXVIII).

Based on solubility and sorption results, it is safe to say that the different self-adhesive luting materials do not behave the same. The behavior of one of the self-adhesive materials does not mean that any other self-adhesive material will behave similarly. Rely X Unicem was comparable to Panavia F and Rely X ARC in both solubility and sorption results; after reviewing the published literature, it can be interpreted as the most tested self-adhesive luting material.<sup>59-63, 65, 66</sup> Maxcem Elite sorption was comparable to the resin at first, but after 90 days it started to increase. Its solubility was comparable to Panavia F during the different storage times (Table XXVIII).

Although the resin-modified glass-ionomers showed the highest percentage of water sorption and solubility of all the materials tested in this study, they behave better compared with more conventional luting materials such as zinc phosphate and zinc polycarboxilate.<sup>26</sup>

As explained before, water sorption and solubility behavior of any given luting material are highly critical in predicting its durability. Based on the results of this study, it can be concluded that self-adhesive luting materials are comparable to the resin-based luting materials. Further testing is required to assess other behavior and properties of such luting agents.

It is difficult to compare the published data regarding water sorption/solubility because of so many variations between the studies. Different sample designs and dimensions, solutions other than water, such as artificial saliva and ethanol, and different methods of testing have been used.

For the resin-modified glass-ionomer luting materials, they showed lower results compared with self-adhesive and resin-based luting material; based on that, it is safe to say that the resin luting materials are the material of choice whenever it is possible.

In the end, this study covers only one aspect of many to be covered and further testing is still needed, focusing on clinical relevance. A beneficial test would be to test these materials when bonded to both tooth structure and restorative material and then placed in the oral cavity using an *in situ* model. A study done by Hersek et al. tested the solubility of the glass-ionomers through bonding the samples to the flanges of a denture and letting the patients wear it for a period of time.<sup>26</sup> Such a study could be duplicated with the luting agent bonded to tooth structure and restorative material and the whole sample attached to a denture flange to be worn by a patient. Such a methodology would allow investigating the solubility and sorption behavior in a situation closer to the actual clinical use of the material.

CONCLUSION



From the results of this study it can be concluded that:

1) Resin-based luting materials had the lowest sorption and solubility.

2) Self-adhesive luting materials behave differently, but generally they are somewhat comparable to resin luting materials with the exception of a few. They behave differently, showing a wide range, but they still behave better compared with the resin-modified glass-ionomers in terms of solubility and sorption behavior.

Resin-modified glass-ionomer luting material showed the greatest water sorption and solubility compared with both resin and self-adhesive materials.

Clinically, high sorption can induce residual stresses leading to post-operative pain and may further lead to failure of the restoration. High solubility leads to debonding, secondary caries, marginal discrepancies, and further failure of the restoration.

REFERENCES

1. Murahara S, Kajihara H, Hori S, Minesaki Y, Onizuka T, Tanaka T. [Bonding durability of commercially-available luting systems for ceramic restoration to dental zirconia]. *Nihon Hotetsu Shika Gakkai Zasshi* 2007;51(4):733-40.
2. Milleding P, Ortengren U, Karlsson S. Ceramic inlay systems: some clinical aspects. *J Oral Rehabil* 1995;22(8):571-80.
3. Soderholm KJ. Degradation of glass filler in experimental composites. *J Dent Res* 1981;60(11):1867-75.
4. Dupuis V, Laviolle O, Potin-Gautier M, Castetbon A, Moya F. Solubility and disintegration of zinc phosphate cement. *Biomaterials* 1992;13(7):467-70.
5. Rantala LI, Lastumaki TM, Peltomaki T, Vallittu PK. Fatigue resistance of removable orthodontic appliance reinforced with glass fibre weave. *J Oral Rehabil* 2003;30(5):501-6.
6. Braem MJ, Lambrechts P, Gladys S, Vanherle G. In vitro fatigue behavior of restorative composites and glass ionomers. *Dent Mater* 1995;11(2):137-41.
7. Indrani DJ, Cook WD, Televantos F, Tyas MJ, Harcourt JK. Fracture toughness of water-aged resin composite restorative materials. *Dent Mater* 1995;11(3):201-7.
8. Feilzer AJ, Kakaboura AI, de Gee AJ, Davidson CL. The influence of water sorption on the development of setting shrinkage stress in traditional and resin-modified glass ionomer cements. *Dent Mater* 1995;11(3):186-90.
9. Anusavice KJ. *Phillips' science of dental materials*. 11 ed. St. Louis: Elsevier Science; 2003.
10. de la Macorra JC, Pradies G. Conventional and adhesive luting cements. *Clin Oral Investig* 2002;6(4):198-204.
11. Rosenstiel SF, Land MF, Crispin BJ. Dental luting agents: a review of the current literature. *J Prosthet Dent* 1998;80(3):280-301.
12. Wilson AD, Kent BE. A new translucent cement for dentistry. The glass ionomer cement. *Br Dent J* 1972;132(4):133-5.

13. Wasson EA, Nicholson JW. New aspects of the setting of glass-ionomer cements. *J Dent Res* 1993;72(2):481-3.
14. Metz JE, Brackett WW. Performance of a glass ionomer luting cement over 8 years in a general practice. *J Prosthet Dent* 1994;71(1):13-5.
15. Muzynski BL, Greener E, Jameson L, Malone WF. Fluoride release from glass ionomers used as luting agents. *J Prosthet Dent* 1988;60(1):41-4.
16. Stannard JG, Sornkul E. Demineralization resistance and tensile bond strength of four luting agents after acid attack. *Int J Prosthodont* 1989;2(5):467-73.
17. Rezk-Lega F, Ogaard B, Rolla G. Availability of fluoride from glass-ionomer luting cements in human saliva. *Scand J Dent Res* 1991;99(1):60-3.
18. Scoville RK, Foreman F, Burgess JO. In vitro fluoride uptake by enamel adjacent to a glass ionomer luting cement. *ASDC J Dent Child* 1990;57(5):352-5.
19. Hallgren A, Oliveby A, Twetman S. Caries associated microflora in plaque from orthodontic appliances retained with glass ionomer cement. *Scand J Dent Res* 1992;100(3):140-3.
20. White SN, Yu Z. Compressive and diametral tensile strengths of current adhesive luting agents. *J Prosthet Dent* 1993;69(6):568-72.
21. Cattani-Lorente MA, Godin C, Meyer JM. Early strength of glass ionomer cements. *Dent Mater* 1993;9(1):57-62.
22. Matsuya S, Maeda T, Ohta M. IR and NMR. analyses of hardening and maturation of glass-ionomer cement. *J Dent Res* 1996;75(12):1920-7.
23. McLean J. Clinical application of glass-ionomer cements. *Oper Dent* 1992;5:184-90.
24. Knibbs PJ, Walls AW. A laboratory and clinical evaluation of three dental luting cements. *J Oral Rehabil* 1989;16(5):467-73.
25. Negm MM, Beech DR, Grant AA. An evaluation of mechanical and adhesive properties of polycarboxylate and glass ionomer cements. *J Oral Rehabil* 1982;9(2):161-7.
26. Hersek NE, Canay S. In vivo solubility of three types of luting cement. *Quintessence Int* 1996;27(3):211-6.
27. Keyf F, Tuna SH, Murat S, Safrany A. Water sorption and solubility of different luting and restorative dental cements. *Turk J Medical Sci* 2006;36(1):47-55.

28. Mathis RS, Ferracane JL. Properties of a glass-ionomer/resin-composite hybrid material. *Dent Mater* 1989;5(5):355-8.
29. Nicholson JW, Anstice HM. The physical chemistry of light-curable glass-ionomers. *J Mater Sci* 1994;5(3):119-22.
30. Wilson AD. Resin-modified glass-ionomer cements. *Int J Prosthodont* 1990;3(5):425-9.
31. Mitra SB, Kedrowski BL. Long-term mechanical properties of glass ionomers. *Dent Mater* 1994;10(2):78-82.
32. Attin T, Vataschki M, Hellwig E. Properties of resin-modified glass-ionomer restorative materials and two polyacid-modified resin composite materials. *Quintessence Int* 1996;27(3):203-9.
33. Ellakuria J, Triana R, Minguez N, et al. Effect of one-year water storage on the surface microhardness of resin-modified versus conventional glass-ionomer cements. *Dent Mater* 2003;19(4):286-90.
34. Peutzfeldt A, Garcia-Godoy F, Asmussen E. Surface hardness and wear of glass ionomers and compomers. *Am J Dent* 1997;10(1):15-7.
35. Nicholson JW, Anstice HM, McLean JW. A preliminary report on the effect of storage in water on the properties of commercial light-cured glass-ionomer cements. *Br Dent J* 1992;173(3):98-101.
36. Momoi Y, Hirosaki K, Kohno A, McCabe JF. In vitro toothbrush-dentifrice abrasion of resin-modified glass ionomers. *Dent Mater* 1997;13(2):82-8.
37. Swift EJ, Jr., Pawlus MA, Vargas MA, Fortin D. Depth of cure of resin-modified glass ionomers. *Dent Mater* 1995;11(3):196-200.
38. Yap AU. Post-irradiation hardness of resin-modified glass ionomer cements and a polyacid-modified composite resin. *J Mater Sci Mater Med* 1997;8(7):413-6.
39. Bourke AM, Walls AW, McCabe JF. Light-activated glass polyalkenoate (ionomer) cements: the setting reaction. *J Dent* 1992;20(2):115-20.
40. Gladys S, Van Meerbeek B, Braem M, Lambrechts P, Vanherle G. Comparative physico-mechanical characterization of new hybrid restorative materials with conventional glass-ionomer and resin composite restorative materials. *J Dent Res* 1997;76(4):883-94.

41. Saskalauskaite E, Tam LE, McComb D. Flexural strength, elastic modulus, and pH profile of self-etch resin luting cements. *J Prosthodont* 2008;17(4):262-8.
42. Cattani-Lorente MA, Dupuis V, Payan J, Moya F, Meyer JM. Effect of water on the physical properties of resin-modified glass ionomer cements. *Dent Mater* 1999;15(1):71-8.
43. Roberson TM, Heymann HO, Edward J Swift J. *Sturdevant's art and science of operative dentistry*. 5th ed. St Louis, Missouri: Mosby Inc.; 2006.
44. White SN, Furuichi R, Kyomen SM. Microleakage through dentin after crown cementation. *J Endod* 1995;21(1):9-12.
45. Ferrari M, Dalloca L, Kugel G, Bertelli E. An evaluation of the effect of the adhesive luting on microleakage of the IPS empress crowns. *Pract Periodontics Aesthet Dent* 1994;6(4):15-23; quiz 24.
46. Milleding P. Microleakage of indirect composite inlays. An in vitro comparison with the direct technique. *Acta Odontol Scand* 1992;50(5):295-301.
47. White SN, Yu Z, Tom JF, Sangsurasak S. In vivo microleakage of luting cements for cast crowns. *J Prosthet Dent* 1994;71(4):333-8.
48. Ban S, Hasegawa J, Anusavice KJ. Effect of loading conditions on bi-axial flexure strength of dental cements. *Dent Mater* 1992;8(2):100-4.
49. Scherrer SS, de Rijk WG, Belser UC, Meyer JM. Effect of cement film thickness on the fracture resistance of a machinable glass-ceramic. *Dent Mater* 1994;10(3):172-7.
50. Mueller HJ. Fracture toughness and fractography of dental cements, lining, build-up, and filling materials. *Scanning Microsc* 1990;4(2):297-307.
51. Wilson AD, Hill RG, Warrens CP, Lewis BG. The influence of polyacid molecular weight on some properties of glass-ionomer cements. *J Dent Res* 1989;68(2):89-94.
52. Moszner N, Salz U, Zimmermann J. Chemical aspects of self-etching enamel-dentin adhesives: a systematic review. *Dent Mater* 2005;21(10):895-910.
53. Brudevold F, Buonocore M, Wileman W. A report on a resin composition capable of bonding to human dentin surfaces. *J Dent Res* 1956;35(6):846-51.
54. Streitwieser A, Heathcock CH. *Introduction to organic chemistry*. New York: Macmillan Publishing Co.; 1976.

55. Anbar M, St John GA, Scott AC. Organic polymeric polyphosphonates as potential preventive agents of dental caries: in vitro experiments. *J Dent Res* 1974;53(4):867-78.
56. Moszner N, Salz U, Zimmermann J. Chemical aspects of self-etching enamel–dentin adhesives: a systematic review. *Dental Materials* 2005;21(10):895-910.
57. Salz U, Zimmermann J, Zeuner F, Moszner N. Hydrolytic stability of self-etching adhesive systems. *J Adhes Dent* 2005;7(2):107-16.
58. 3M ESPE AG · ESPE Platz SG. RelyXTMUnicem – Self-Adhesive Universal Resin Cement in the Clicker TMDispenser Technical Data Sheet.
59. De Munck J, Vargas M, Van Landuyt K, Hikita K, Lambrechts P, Van Meerbeek B. Bonding of an auto-adhesive luting material to enamel and dentin. *Dent Mater* 2004;20(10):963-71.
60. Escribano N, de la Macorra JC. Microtensile bond strength of self-adhesive luting cements to ceramic. *J Adhes Dent* 2006;8(5):337-41.
61. Hikita K, Van Meerbeek B, De Munck J, et al. Bonding effectiveness of adhesive luting agents to enamel and dentin. *Dent Mater* 2007;23(1):71-80.
62. Abo-Hamar SE, Hiller KA, Jung H, Federlin M, Friedl KH, Schmalz G. Bond strength of a new universal self-adhesive resin luting cement to dentin and enamel. *Clin Oral Investig* 2005;9(3):161-7.
63. Wiedig CA, Hecht R, Ludsteck M, Raia G, Rennschmid H. Microtensile testing of self-adhesive luting cements on human dentin. 3M ESPE AG, Seefeld, Germany 2006.
64. Fabianelli A, Goracci C, Bertelli E, Monticelli F, Grandini S, Ferrari M. In vitro evaluation of wall-to-wall adaptation of a self-adhesive resin cement used for luting gold and ceramic inlays. *J Adhes Dent* 2005;7(1):33-40.
65. Behr M, Rosentritt M, Regnet T, Lang R, Handel G. Marginal adaptation in dentin of a self-adhesive universal resin cement compared with well-trieed systems. *Dent Mater* 2004;20(2):191-7.
66. Ibarra G, Johnson GH, Geurtsen W, Vargas MA. Microleakage of porcelain veneer restorations bonded to enamel and dentin with a new self-adhesive resin-based dental cement. *Dent Mater* 2007;23(2):218-25.
67. Frankenberger R, Lohbauer U, Schaible RB, Nikolaenko SA, Naumann M. Luting of ceramic inlays in vitro: marginal quality of self-etch and etch-and-rinse adhesives versus self-etch cements. *Dent Mater* 2008;24(2):185-91.

68. Frankenberger R, Strobel WO, Kramer N, et al. Evaluation of the fatigue behavior of the resin-dentin bond with the use of different methods. *J Biomed Mater Res B Appl Biomater* 2003;67(2):712-21.
69. Frankenberger R, Tay FR. Self-etch vs etch-and-rinse adhesives: effect of thermo-mechanical fatigue loading on marginal quality of bonded resin composite restorations. *Dent Mater* 2005;21(5):397-412.
70. Frankenberger R, Strobel WO, Lohbauer U, Kramer N, Petschelt A. The effect of six years of water storage on resin composite bonding to human dentin. *J Biomed Mater Res B Appl Biomater* 2004;69(1):25-32.
71. Carvalho RM, Chersoni S, Frankenberger R, Pashley DH, Prati C, Tay FR. A challenge to the conventional wisdom that simultaneous etching and resin infiltration always occurs in self-etch adhesives. *Biomaterials* 2005;26(9):1035-42.
72. Itthagarun A, Tay FR, Pashley DH, Wefel JS, Garcia-Godoy F, Wei SH. Single-step, self-etch adhesives behave as permeable membranes after polymerization. (Pt 3). Evidence from fluid conductance and artificial caries inhibition. *Am J Dent* 2004;17(6):394-400.
73. Tay FR, Pashley DH, Garcia-Godoy F, Yiu CK. Single-step, self-etch adhesives behave as permeable membranes after polymerization.( Pt 2). Silver tracer penetration evidence. *Am J Dent* 2004;17(5):315-22.
74. Tay FR, Frankenberger R, Krejci I, et al. Single-bottle adhesives behave as permeable membranes after polymerization. (Pt 1). In vivo evidence. *J Dent* 2004;32(8):611-21.
75. Tay FR, Pashley DH, Suh BI, Carvalho RM, Itthagarun A. Single-step adhesives are permeable membranes. *J Dent* 2002;30(7-8):371-82.
76. Tay FR, Pashley DH, Suh BI, Hiraishi N, Yiu CK. Water treeing in simplified dentin adhesives--deja vu? *Oper Dent* 2005;30(5):561-79.
77. Mesu FP, Reedijk T. Degradation of luting cements measured in vitro and in vivo. *J Dent Res* 1983;62(12):1236-40.
78. Uno S, Finger WJ, Fritz U. Long-term mechanical characteristics of resin-modified glass ionomer restorative materials. *Dent Mater* 1996;12(1):64-9.
79. Malacarne J, Carvalho RM, de Goes MF, Svizero N, Pashley DH, Tay FR, et al. Water sorption/solubility of dental adhesive resins. *Dent Mater* 2006;22(10):973-80.



80. Pastila P, Lassila LV, Jokinen M, Vuorinen J, Vallittu PK, Mantyla T. Effect of short-term water storage on the elastic properties of some dental restorative materials--a resonant ultrasound spectroscopy study. *Dent Mater* 2007;23(7):878-84.
81. Piwowarczyk A, Lauer HC. Mechanical properties of luting cements after water storage. *Oper Dent* 2003;28(5):535-42.
82. Ortengren U, Elgh U, Spasenoska V, Milleding P, Haasum J, Karlsson S. Water sorption and flexural properties of a composite resin cement. *Int J Prosthodont* 2000;13(2):141-7.
83. Pedreira AP, Pegoraro LF, de Goes MF, Pegoraro TA, Carvalho RM. Microhardness of resin cements in the intraradicular environment: effects of water storage and softening treatment. *Dent Mater* 2009;25(7):868-76.
84. Gerdolle DA, Mortier E, Jacquot B, Panighi MM. Water sorption and water solubility of current luting cements: an in vitro study. *Quintessence Int* 2008;39(3):e107-14.
85. Mese A, Burrow MF, Tyas MJ. Sorption and solubility of luting cements in different solutions. *Dent Mater J* 2008;27(5):702-9.
86. Oyague RC, Monticelli F, Toledano M, Osorio E, Ferrari M, Osorio R. Effect of water aging on microtensile bond strength of dual-cured resin cements to pre-treated sintered zirconium-oxide ceramics. *Dent Mater* 2009;25(3):392-9.
87. Abdalla AI, El Eraki M, Feilzer AJ. The effect of direct and indirect water storage on the microtensile dentin bond strength of a total-etch and two self-etching adhesives. *Am J Dent* 2007;20(6):370-4.
88. Small IC, Watson TF, Chadwick AV, Sidhu SK. Water sorption in resin-modified glass-ionomer cements: an in vitro comparison with other materials. *Biomaterials* 1998;19(6):545-50.
89. Behr M, Rosentritt M, Loher H, et al. Changes of cement properties caused by mixing errors: the therapeutic range of different cement types. *Dent Mater* 2008;24(9):1187-93.
90. Fleming GJ, Marquis PM, Shortall AC. The influence of clinically induced variability on the distribution of compressive fracture strengths of a hand-mixed zinc phosphate dental cement. *Dent Mater* 1999;15(2):87-97.
91. Meerbeek BV, Landuyt KV, Inoue S, et al. Bonding to enamel and dentin. In: Summitt JB, Robbins JW, Hilton TJ, Schwartz RS. *Fundamentals of operative*

dentistry, a contemporary approach. Chicago: Quintessence Publishing Co., Inc; 2006.

92. Hunt PR. Glass ionomers: the next generation. A summary of the current situation. *J Esthet Dent* 1994;6(5):192-4.
93. Burgess J, Norling B, Summitt J. Resin ionomer restorative materials: the new generation. *J Esthet Dent* 1994;6(5):207-15.
94. Kerby RE, A.Knobloch L, Schricker S, Gregg B. Synthesis and evaluation of modified urethane dimethacrylate resins with reduced water sorption and solubility. *Dental Materials* 2009;25(3):302-13.
95. Yap A, Lee CM. Water sorption and solubility of resin-modified polyalkenoate cements. *J Oral Rehabil* 1997;24(4):310-4.

ABSTRACT

EVALUATION OF WATER SORPTION AND SOLUBILITY  
BEHAVIOR OF NINE DIFFERENT POLYMERIC  
LUTING MATERIALS

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The cementation procedure is the key to long-term success of fixed restorations. The prognosis of prosthetic restoration is largely impacted by the maintenance of the luting cement and the adhesive bond. When exposed to water or saliva, most restorative materials undergo hydrolytic degradation. The purpose of this study is to evaluate the water solubility and water sorption characteristics of newly introduced acidic polymeric luting agents over a 180-day water-storage period.

Nine different luting agents were tested. Fifty-two disc specimens of each material were fabricated using a mold with an internal dimension of  $15\pm 0.1$  mm in diameter and  $1.0\pm 0.1$  mm deep. A constant weight,  $W_0$ , was reached after desiccating the specimens. Then, 13 specimens were assigned randomly to one of the four testing periods

in the water for seven, 30, 90 and 180 days. After each period, the specimens were removed from the water and weighed to get  $W_1$ . A second period of desiccating the samples provided a constant weight  $W_2$ . The water sorption and solubility were determined by the following equations:  $W_{SP}(\%) = (W_1 - W_2) \times 100 / W_0$ ,  $W_{SL}(\%) = (W_0 - W_2) \times 100 / W_0$ .

The resin-modified glass-ionomers showed the highest water sorption/solubility results. The resin luting agents had the lowest sorption/solubility results. The self-adhesives showed a wide range of solubility/sorption; in general, they showed lower results compared with the resin-modified glass-ionomers. All the materials reached some sort of equilibrium after 90-days.

Based on the results of our study, we conclude that self-adhesive luting materials were not all alike. Rely X Unicem was the most comparable to the resin luting materials. The resin luting materials had the lowest solubility and sorption. Resin-modified glass-ionomers showed the highest sorption/solubility results.

APPENDICES

## APPENDIX I

## Water sorption results by time periods

Analysis Variable: wsp Water sorption (%%)								
Time period (days)	Luting agent	N Obs	N	Mean	Std Dev	Minimum	Maximum	Median
7	BC	13	13	6.44	0.38	5.54	7.08	6.34
	BZ	13	13	4.66	0.18	4.40	5.08	4.67
	FC	13	13	15.67	0.55	14.93	16.74	15.69
	FP	13	13	12.87	1.23	10.55	14.86	12.91
	MX	13	13	3.68	0.21	3.43	4.04	3.61
	PF	13	13	1.62	0.21	1.14	1.92	1.63
	RA	13	13	1.87	0.19	1.62	2.20	1.85
	RL	13	13	16.92	0.46	16.19	17.58	16.96
	RU	12	12	1.89	0.16	1.59	2.26	1.88
30	BC	13	13	6.55	0.21	6.26	6.93	6.54
	BZ	13	13	5.31	0.26	4.76	5.68	5.38
	FC	13	13	16.21	0.47	15.25	17.01	16.22
	FP	13	13	12.79	1.02	10.81	14.09	12.94
	MX	13	13	3.88	0.19	3.62	4.22	3.86
	PF	13	13	2.21	0.17	1.96	2.49	2.20
	RA	13	13	1.76	0.08	1.69	1.93	1.74
	RL	13	13	17.12	1.31	13.19	18.41	17.19
	RU	13	13	2.53	0.21	2.10	2.86	2.57
90	BC	13	13	6.69	0.27	5.97	7.03	6.68
	BZ	12	12	5.47	0.17	5.20	5.71	5.46
	FC	13	13	14.26	8.95	-15.21	18.08	16.90
	FP	13	13	14.16	1.91	10.76	16.63	14.26
	MX	8	8	4.14	0.16	3.99	4.41	4.05
	PF	13	13	2.49	0.21	2.10	2.83	2.42
	RA	13	13	1.79	0.06	1.65	1.87	1.81
	RL	13	13	18.32	0.34	17.77	18.95	18.27
	RU	12	12	3.27	0.35	2.69	3.69	3.26
180	BC	13	13	6.59	0.17	6.44	6.96	6.52
	BZ	13	13	5.55	0.17	5.20	5.85	5.59
	FC	12	12	16.97	0.67	15.59	17.92	17.03
	FP	13	13	14.14	1.53	12.38	16.90	14.01
	MX	12	12	4.01	0.25	3.57	4.48	4.00
	PF	13	13	2.70	0.18	2.45	3.13	2.69
	RA	13	13	1.80	0.08	1.71	1.97	1.78
	RL	13	13	15.47	9.04	-14.52	19.08	17.98
	RU	12	12	3.95	0.42	3.64	5.23	3.86

## APPENDIX II

## Water sorption results by luting agents

Luting agent	Time period (days)	N Obs	N	Mean	Std Dev	Minimum	Maximum	Median
BC	7	13	13	6.44	0.38	5.54	7.08	6.34
	30	13	13	6.55	0.21	6.26	6.93	6.54
	90	13	13	6.69	0.27	5.97	7.03	6.68
	180	13	13	6.59	0.17	6.44	6.96	6.52
BZ	7	13	13	4.66	0.18	4.40	5.08	4.67
	30	13	13	5.31	0.26	4.76	5.68	5.38
	90	12	12	5.47	0.17	5.20	5.71	5.46
	180	13	13	5.55	0.17	5.20	5.85	5.59
FC	7	13	13	15.67	0.55	14.93	16.74	15.69
	30	13	13	16.21	0.47	15.25	17.01	16.22
	90	13	13	14.26	8.95	-15.21	18.08	16.90
	180	12	12	16.97	0.67	15.59	17.92	17.03
FP	7	13	13	12.87	1.23	10.55	14.86	12.91
	30	13	13	12.79	1.02	10.81	14.09	12.94
	90	13	13	14.16	1.91	10.76	16.63	14.26
	180	13	13	14.14	1.53	12.38	16.90	14.01
MX	7	13	13	3.68	0.21	3.43	4.04	3.61
	30	13	13	3.88	0.19	3.62	4.22	3.86
	90	8	8	4.14	0.16	3.99	4.41	4.05
	180	12	12	4.01	0.25	3.57	4.48	4.00
PF	7	13	13	1.62	0.21	1.14	1.92	1.63
	30	13	13	2.21	0.17	1.96	2.49	2.20
	90	13	13	2.49	0.21	2.10	2.83	2.42
	180	13	13	2.70	0.18	2.45	3.13	2.69
RA	7	13	13	1.87	0.19	1.62	2.20	1.85
	30	13	13	1.76	0.08	1.69	1.93	1.74
	90	13	13	1.79	0.06	1.65	1.87	1.81
	180	13	13	1.80	0.08	1.71	1.97	1.78
RL	7	13	13	16.92	0.46	16.19	17.58	16.96
	30	13	13	17.12	1.31	13.19	18.41	17.19
	90	13	13	18.32	0.34	17.77	18.95	18.27
	180	13	13	15.47	9.04	-14.52	19.08	17.98
RU	7	12	12	1.89	0.16	1.59	2.26	1.88
	30	13	13	2.53	0.21	2.10	2.86	2.57
	90	12	12	3.27	0.35	2.69	3.69	3.26
	180	12	12	3.95	0.42	3.64	5.23	3.86



## APPENDIX III

## Water solubility results by time periods

Analysis Variable: wsl Water solubility (%)								
Time period (days)	Luting agent	N Obs	N	Mean	Std Dev	Minimum	Maximum	Median
7	BC	13	13	0.57	0.30	-0.13	1.32	0.56
	BZ	13	13	0.75	0.05	0.66	0.82	0.74
	FC	13	13	2.29	0.26	1.61	2.62	2.32
	FP	13	13	-0.18	4.14	-13.95	1.23	0.93
	MX	13	13	0.75	0.10	0.56	0.87	0.78
	PF	13	13	0.43	0.12	0.26	0.65	0.41
	RA	13	13	0.74	0.22	0.43	1.17	0.72
	RL	13	13	1.88	0.30	1.57	2.46	1.85
	RU	12	12	0.06	0.05	0.00	0.18	0.05
30	BC	13	13	0.73	0.07	0.66	0.91	0.71
	BZ	13	13	0.83	0.08	0.69	0.98	0.82
	FC	13	13	3.24	0.79	2.30	5.52	3.14
	FP	13	13	1.07	0.13	0.89	1.26	1.05
	MX	13	13	1.04	0.13	0.82	1.22	1.08
	PF	13	13	0.56	0.16	0.35	0.87	0.54
	RA	13	13	0.55	0.13	0.37	0.86	0.55
	RL	13	13	4.31	7.34	2.02	28.73	2.25
	RU	13	13	0.07	0.03	0.02	0.13	0.07
90	BC	13	13	1.27	0.17	1.04	1.77	1.22
	BZ	12	12	0.99	0.09	0.86	1.15	1.00
	FC	13	13	6.09	11.47	-25.25	30.39	6.63
	FP	13	13	2.81	0.38	2.23	3.27	2.92
	MX	8	8	1.16	0.11	0.97	1.30	1.18
	PF	13	13	0.82	0.14	0.67	1.08	0.78
	RA	13	13	0.31	0.04	0.22	0.36	0.31
	RL	13	13	4.42	0.21	4.13	4.83	4.48
	RU	12	12	0.16	0.05	0.07	0.24	0.16
180	BC	13	13	1.12	0.12	0.92	1.26	1.15
	BZ	13	13	1.15	0.13	0.91	1.35	1.18
	FC	12	12	7.59	0.56	6.65	8.66	7.65
	FP	13	13	3.11	0.38	2.49	3.72	3.13
	MX	12	12	1.55	0.12	1.41	1.71	1.52
	PF	13	13	0.85	0.18	0.64	1.30	0.80
	RA	13	13	0.23	0.02	0.20	0.29	0.23
	RU	12	12	0.22	0.05	0.16	0.30	0.22

## APPENDIX IV

## Water solubility results by luting agent

Luting agent	Time period (days)	N Obs	N	Mean	Std Dev	Minimum	Maximum	Median
BC	7	13	13	0.57	0.30	-0.13	1.32	0.56
	30	13	13	0.73	0.07	0.66	0.91	0.71
	90	13	13	1.27	0.17	1.04	1.77	1.22
	180	13	13	1.12	0.12	0.92	1.26	1.15
BZ	7	13	13	0.75	0.05	0.66	0.82	0.74
	30	13	13	0.83	0.08	0.69	0.98	0.82
	90	12	12	0.99	0.09	0.86	1.15	1.00
	180	13	13	1.15	0.13	0.91	1.35	1.18
FC	7	13	13	2.29	0.26	1.61	2.62	2.32
	30	13	13	3.24	0.79	2.30	5.52	3.14
	90	13	13	6.09	11.47	-25.25	30.39	6.63
	180	12	12	7.59	0.56	6.65	8.66	7.65
FP	7	13	13	-0.18	4.14	-13.95	1.23	0.93
	30	13	13	1.07	0.13	0.89	1.26	1.05
	90	13	13	2.81	0.38	2.23	3.27	2.92
	180	13	13	3.11	0.38	2.49	3.72	3.13
MX	7	13	13	0.75	0.10	0.56	0.87	0.78
	30	13	13	1.04	0.13	0.82	1.22	1.08
	90	8	8	1.16	0.11	0.97	1.30	1.18
	180	12	12	1.55	0.12	1.41	1.71	1.52
PF	7	13	13	0.43	0.12	0.26	0.65	0.41
	30	13	13	0.56	0.16	0.35	0.87	0.54
	90	13	13	0.82	0.14	0.67	1.08	0.78
	180	13	13	0.85	0.18	0.64	1.30	0.80
RA	7	13	13	0.74	0.22	0.43	1.17	0.72
	30	13	13	0.55	0.13	0.37	0.86	0.55
	90	13	13	0.31	0.04	0.22	0.36	0.31
	180	13	13	0.23	0.02	0.20	0.29	0.23
RL	7	13	13	1.88	0.30	1.57	2.46	1.85
	30	13	13	4.31	7.34	2.02	28.73	2.25
	90	13	13	4.42	0.21	4.13	4.83	4.48
	180	13	13	1.87	9.28	-29.00	5.21	4.39
RU	7	12	12	0.06	0.05	0.00	0.18	0.05
	30	13	13	0.07	0.03	0.02	0.13	0.07
	90	12	12	0.16	0.05	0.07	0.24	0.16
	180	12	12	0.22	0.05	0.16	0.30	0.22

## APPENDIX V

## Original data Breeze (BZ)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
BZ 1	364.4	15.08	15.05	15.065	1.09	1.1	1.095	195.084178	1.867911605	380.4	359.8	180
BZ 2	357.9	15.1	15.03	15.065	1.07	1.05	1.06	188.8486106	1.895168828	372.9	353.7	180
BZ 3	373.3	15.05	15.07	15.06	1.1	1.1	1.1	195.8449086	1.906100101	389.8	368.9	180
BZ 4	379.3	15.05	15.07	15.06	1.06	1.1	1.08	192.2840921	1.97260208	395.8	374.6	180
BZ 5	367.6	15.06	15.09	15.075	1.04	1.09	1.065	189.9913839	1.934824582	384.3	363.2	180
BZ 6	348.8	15.02	14.98	15	1.09	1.06	1.075	189.871875	1.837028259	364.7	345.3	180
BZ 7	372.7	15.06	15.05	15.055	1.1	1.09	1.095	194.825274	1.912996155	390.3	368.5	180
BZ 8	370	15.09	14.97	15.03	1.07	1.1	1.085	192.4054441	1.92302251	387.2	366.5	180
BZ 9	373.1	15.04	14.93	14.985	1.1	1.1	1.1	193.8991193	1.924196466	389.1	369.7	180
BZ 10	373.8	15.07	15.02	15.045	1.09	1.1	1.095	194.5665419	1.921193625	390.2	369.4	180
BZ 11	368.6	15.03	15.07	15.05	1.11	1.06	1.085	192.9178418	1.910657908	384.6	364	180
BZ 12	348	15.08	15.03	15.055	1.1	1.01	1.055	187.708369	1.853939715	362.3	343.3	180
BZ 13	374.3	15.05	15.08	15.065	1.09	1.06	1.075	191.5209966	1.954354909	390.2	370.2	180
BZ 14	BROKE											
BZ 15	369	15.05	15.06	15.055	1.05	1.07	1.06	188.5979821	1.956542673	384.5	365.3	90
BZ 16	371.5	15.07	15.04	15.055	1.07	1.05	1.06	188.5979821	1.969798382	388.3	367.3	90
BZ 17	377.9	15.07	15.06	15.065	1.07	1.1	1.085	193.3025873	1.954966073	394.3	374.2	90
BZ 18	375.5	15.07	15	15.035	1.07	1.1	1.085	192.5334796	1.950310153	392.9	371.7	90
BZ 19	381.4	15.05	15.07	15.06	1.1	1.09	1.095	194.9547045	1.956351867	398	377.9	90
BZ 20	360	15.04	15.1	15.07	1.1	1.05	1.075	191.6481475	1.878442368	376.6	356.9	90
BZ 21	362.1	15.06	15.05	15.055	1.06	1.1	1.08	192.1564346	1.884402158	379.1	358.7	90
BZ 22	381.6	15.01	15.03	15.02	1.09	1.1	1.095	193.9204638	1.967817075	398	377.7	90
BZ 23	344.8	15.02	14.99	15.005	1	0.95	0.975	172.3242004	2.000879733	360.9	341.2	90
BZ 24	358.2	15.08	15.01	15.045	1.07	1.04	1.055	187.4590883	1.910816932	374	354.6	90
BZ 25	256.2	15.04	15.02	15.03	1.09	1.01	1.05	186.1988168	1.375948593	371.7	352.1	90
BZ 26	372.4	15.02	15.03	15.025	1.1	1.07	1.085	192.2774511	1.936784568	389.5	369.2	90
BZ 27	359.8	15.01	15.03	15.02	1	1.06	1.03	182.4092034	1.972488193	375.9	357.1	30
BZ 28	368.6	15.03	15.08	15.055	1.01	1.09	1.05	186.8187559	1.9730353	385.6	365.3	30
BZ 29	358.7	15.03	15.03	15.03	1.05	1.01	1.03	182.6521727	1.963841955	375.3	356	30
BZ 30	352.9	15.08	15.08	15.08	1.01	0.99	1	178.514024	1.976875497	366.8	350	30
BZ 31	357.6	15.08	15.04	15.06	1.01	1.06	1.035	184.2722549	1.940606849	374.2	354.4	30
BZ 32	346.1	15.08	15.1	15.09	0.98	1.07	1.025	183.21963	1.888989734	362.3	343.1	30
BZ 33	348.5	15.01	14.97	14.99	1.05	1	1.025	180.799318	1.927551519	364.9	345.1	30
BZ 34	353.4	15.06	15.03	15.045	1.05	0.99	1.02	181.2400664	1.94989997	368.8	350.5	30
BZ 35	375.8	15.08	15.03	15.055	1.07	1.06	1.065	189.4875952	1.983243281	391.9	372.8	30
BZ 36	368.8	15.08	15.08	15.08	1.1	1.01	1.055	188.3322953	1.958240881	385.8	365.8	30
BZ 37	359.3	15.08	15.06	15.07	1.1	1.04	1.07	190.7567608	1.883550541	375.5	356.1	30
BZ 38	370.5	15.1	15.08	15.09	1.02	1	1.01	180.5383671	2.052195364	387.3	367.5	30
BZ 39	361.3	15.1	15.06	15.08	1.01	1.07	1.04	185.654585	1.946087139	376.6	358.8	30

(continued)

## APPENDIX V

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
BZ 40	352.2	15.08	15.04	15.06	1.06	0.99	1.025	182.4918467	1.929949236	366.2	349.7	7
BZ 41	361	15.08	15.06	15.07	1.01	1.07	1.04	185.4084404	1.947052676	375.5	358.1	7
BZ 42	365.3	15.08	15.08	15.08	1.05	1.06	1.055	188.3322953	1.939656708	379.7	362.6	7
BZ 43	362.1	15.08	15.07	15.075	1.06	1.06	1.06	189.0994056	1.914865882	376.1	359.2	7
BZ 44	356.2	15.08	15.1	15.09	1	1.02	1.01	180.5383671	1.972987824	370	353.6	7
BZ 45	349.9	15.03	15.06	15.045	1.01	1.05	1.03	183.0169298	1.911844988	363.3	347.6	7
BZ 46	357.2	15.1	15.08	15.09	1.1	1	1.05	187.6884014	1.903154363	371.3	354.6	7
BZ 47	358.3	15.08	15.03	15.055	1.07	1.05	1.06	188.5979821	1.899808238	373.7	355.5	7
BZ 48	357.6	15.01	15.08	15.045	1.05	1.02	1.035	183.9053615	1.944478383	371.1	355.1	7
BZ 49	363.2	15.03	15.03	15.03	1.06	1.09	1.075	190.632122	1.905240293	376.9	360.4	7
BZ 50	361	15.03	15.06	15.045	1.09	1.02	1.055	187.4590883	1.925753524	374.2	358.3	7
BZ 51	342	14.98	14.96	14.97	0.99	1.06	1.025	180.3171867	1.896657808	355.6	339.2	7
BZ 52	363.3	15.08	15.07	15.075	1.06	1.01	1.035	184.6395139	1.967617832	377.4	360.6	7

## APPENDIX VI

## Original data BisCem (BC)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
BC 1	339.9	15.08	15.02	15.05	1	1.09	1.045	185.8056633	1.829330678	359.3	335.8	180
BC 2	347.6	15.08	15.06	15.07	1.04	1.09	1.065	189.865374	1.830770891	366.6	344.1	180
BC 3	347.8	14.94	15.01	14.98	1.04	1.06	1.05	184.8385777	1.881641833	367.8	344.5	180
BC 4	360.7	15.02	15.03	15.03	1.09	1.07	1.08	191.3913799	1.884619883	379.8	356.5	180
BC 5	356.1	15.08	15.06	15.07	1.04	1.09	1.065	189.865374	1.875539454	374.8	351.6	180
BC 6	335.9	15.03	15.01	15.02	0.99	1.02	1.005	177.9817956	1.887271667	354.7	332.8	180
BC 7	349.3	15.08	15.08	15.08	1.01	1.07	1.04	185.654585	1.881450976	369.5	345.2	180
BC 8	350.1	15.08	15.08	15.08	1.02	1.05	1.035	184.7620148	1.894870005	368.8	345.7	180
BC 9	339.5	15.06	14.63	14.85	0.99	1.04	1.015	175.5885138	1.933497771	357.5	335.6	180
BC 10	352.8	15.04	15.04	15.04	1.04	1.06	1.05	186.4466688	1.892230107	371.6	348.4	180
BC 11	368.5	15.06	15.08	15.07	1.1	1.09	1.095	195.2136944	1.887674946	389.3	365	180
BC 12	349.2	15.1	15.08	15.09	1.01	1.09	1.05	187.6884014	1.860530525	367.9	345.4	180
BC 13	354.8	15.08	15.03	15.06	1.07	1.1	1.085	193.0460477	1.837903465	373.9	350.8	180
BC 14	356.9	15.03	15.04	15.04	1.04	1.07	1.055	187.2099733	1.906415528	373.5	352.2	90
BC 15	354.2	15.03	15.05	15.04	1.05	1.05	1.05	186.4466688	1.899738956	374.5	349.8	90
BC 16	350.4	15.01	15.02	15.02	1.08	1.03	1.055	186.7122401	1.876684677	369.6	346.2	90
BC 17	340.2	15.07	15.1	15.09	0.98	1.04	1.01	180.4187458	1.885613374	358.6	336	90
BC 18	347	15.05	15.02	15.04	1.09	1	1.045	185.4354711	1.871270895	366.6	342.8	90
BC 19	353.4	15.06	15.06	15.06	1.04	1.08	1.06	188.7232756	1.872583013	373.4	348.7	90
BC 20	353.1	15.06	15.04	15.05	1.05	1.04	1.045	185.8056633	1.900372646	372.4	349	90
BC 21	359.7	15	15.08	15.04	1.03	1.1	1.065	189.1101926	1.902065642	380.6	355.3	90
BC 22	355.3	15.08	15.09	15.09	1.06	1.07	1.065	190.243529	1.86760623	374.8	351.1	90
BC 23	352.4	14.98	15.01	15.00	1.04	1.1	1.07	188.8627785	1.865904986	371.7	348.1	90
BC 24	355.3	15.1	15.01	15.06	1.03	1.08	1.055	187.708369	1.892829829	373.8	350.4	90
BC 25	345.1	15.1	15.08	15.09	1.01	1.04	1.025	183.21963	1.883531803	364	341.5	90
BC 26	339.9	14.95	15.02	14.99	0.95	1.1	1.025	180.6787248	1.881239755	356.6	333.9	90
BC 27	341.9	15	15.02	15.01	1.03	1.07	1.05	185.7036074	1.841105861	362.8	339.5	30
BC 28	361	15.03	15.07	15.05	1.1	1.03	1.065	189.3617526	1.906403987	381	358.2	30
BC 29	365.5	15.06	15.1	15.08	1.1	1.05	1.075	191.9025758	1.904612267	386.8	362.9	30
BC 30	379.1	15.05	15.07	15.06	1.1	1.1	1.1	195.8449086	1.935715371	401	376.6	30
BC 31	373.6	15.08	15.08	15.08	1.1	1.08	1.09	194.5802862	1.920030068	394.2	370.8	30
BC 32	375.7	15.07	15.08	15.08	1.09	1.06	1.075	191.7753405	1.959063136	397.1	373.1	30
BC 33	372.8	15.05	15.06	15.06	1.09	1.06	1.075	191.2668215	1.949109611	395.2	370.3	30
BC 34	358.9	15.05	15.06	15.06	1.1	1.09	1.095	194.825274	1.842163456	380.5	356.1	30
BC 35	381.7	15.01	15.03	15.02	1.1	1.11	1.105	195.691427	1.950519785	403.8	378.7	30
BC 36	371.5	14.75	15.09	14.92	1.1	1.05	1.075	187.8519758	1.977620935	392.7	369	30
BC 37	388.5	15.08	15.07	15.08	1.1	1.09	1.095	195.3432539	1.988806843	410.7	385.8	30
BC 38	372.2	15.03	15.08	15.06	1.1	1.06	1.08	192.1564346	1.936963499	394.6	368.8	30
BC 39	385.5	15.06	15.04	15.05	1.09	1.07	1.08	192.0288195	2.007511169	408.1	382.7	30

(continued)

## APPENDIX VI

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
BC 40	368.5	15.1	15.07	15.09	1.05	1.1	1.075	192.0298532	1.918972461	391.3	366.2	7
BC 41	354.5	15.04	15.02	15.03	1	1.1	1.05	186.1988168	1.903878908	375.1	352.2	7
BC 42	372.4	15.07	15.05	15.06	1.1	1.07	1.085	193.1742962	1.92779271	394	370.6	7
BC 43	354.4	15.05	15.06	15.06	1.1	1	1.05	186.8187559	1.897025801	376.2	352.3	7
BC 44	388.1	15.05	15.07	15.06	1.1	1.1	1.1	195.8449086	1.981670102	410.2	386.1	7
BC 45	313.5	15.09	15.1	15.10	1.09	1.05	1.07	191.390188	1.638015006	405.5	381.6	7
BC 46	375.1	15.05	15.03	15.04	1.1	1.09	1.095	194.4372403	1.929157189	396.8	373.1	7
BC 47	384.7	15.01	15.09	15.05	1.1	1.09	1.095	194.6958864	1.975902044	407.2	382.8	7
BC 48	379.3	15.08	15.09	15.09	1.1	1.06	1.08	192.9230154	1.966069208	402.3	377	7
BC 49	383.5	15	15.05	15.03	1.13	1.09	1.11	196.7078071	1.949592168	406.9	381.1	7
BC 50	360.2	15.08	15.05	15.07	1.1	1.07	1.085	193.3025873	1.863399787	380.9	358.2	7
BC 51	377.2	15.08	15.03	15.06	1.1	1.06	1.08	192.1564346	1.962983966	398.6	377.7	7

## APPENDIX VII

## Original data Fuji Plus (FP)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
FP 1	351.1	14.54	14.58	14.56	0.99	1	0.995	165.5829011	2.120388021	389.7	340.4	180
FP 2	378.9	14.7	14.7	14.7	1.01	1.06	1.035	175.5677228	2.158141565	420.4	367.3	180
FP 3	348	14.61	14.64	14.625	1.06	1.01	1.035	173.7807855	2.002522885	387.8	335.9	180
FP 4	358.8	14.63	14.57	14.6	1.04	1.06	1.05	175.69713	2.042150603	394	348.8	180
FP 5	349	14.68	14.63	14.655	1.01	1.05	1.03	173.6514952	2.009772503	390.5	336.8	180
FP 6	359.9	14.06	14.57	14.315	1.07	1.1	1.085	174.5348269	2.062052637	407.7	347.4	180
FP 7	358	14.6	14.57	14.585	1.15	1.06	1.105	184.520576	1.940163031	405.2	344.7	180
FP 8	353.8	14.67	14.68	14.675	1.02	1.03	1.025	173.2805198	2.041775962	390.7	342.4	180
FP 9	374	14.62	13.68	14.15	1.06	1.05	1.055	165.8192689	2.255467669	411	364.7	180
FP 10	351.8	14.6	14.64	14.62	1.02	1	1.01	169.4672475	2.075917353	387.2	342.4	180
FP 11	367.1	14.6	14.61	14.605	1	1	1	167.4452296	2.192358665	404.5	355.3	180
FP 12	370.6	14.68	14.64	14.66	1.05	1.09	1.07	180.5183582	2.052976792	407	361	180
FP 13	354.7	14.66	14.65	14.655	1.02	1.04	1.03	173.6514952	2.042596867	395.5	343.6	180
FP 14	339.7	14.52	14.53	14.525	1.04	1.04	1.04	172.2405003	1.972242298	385	329.4	90
FP 15	358.6	14.58	14.54	14.56	1.07	1.06	1.065	177.2319494	2.023337221	405.7	347	90
FP 16	374.4	14.66	14.65	14.655	1.09	1.06	1.075	181.238211	2.065789537	412.8	365.3	90
FP 17	332.2	14.51	14.54	14.525	0.99	1	0.995	164.7877863	2.015926104	374.1	322.5	90
FP 18	386.4	14.69	14.76	14.725	1.07	1.09	1.08	183.8247649	2.102001873	425.2	377.8	90
FP 19	380	14.62	14.66	14.64	1.05	1.03	1.04	174.9786854	2.171693078	419.1	370.1	90
FP 20	385.7	14.6	14.66	14.63	1.06	1.08	1.07	179.7802942	2.145396423	425.8	375.8	90
FP 21	332.6	14.62	14.69	14.655	1.08	1.09	1.085	182.9241478	1.818239986	370.4	321.8	90
FP 22	361.1	14.59	14.55	14.57	0.97	0.98	0.975	162.4775553	2.222460815	397.7	351.7	90
FP 23	375.8	14.59	14.58	14.585	1.09	1.05	1.07	178.6760329	2.103247951	418.3	364.7	90
FP 24	395.8	14.73	14.72	14.725	1.05	1.01	1.03	175.3143591	2.257658768	429.3	386.7	90
FP 25	332.7	14.47	14.51	14.49	0.99	0.96	0.975	160.6982115	2.070340403	375.4	322.1	90
FP 26	321.1	14.6	14.57	14.585	1	0.91	0.955	159.472534	2.013512872	364	310.6	90
FP 27	364.5	14.57	14.61	14.59	1	0.96	0.98	163.7594293	2.225826027	409.9	360	30
FP 28	379.4	14.6	14.61	14.605	1.05	1	1.025	171.6313604	2.210551727	425.3	375.1	30
FP 29	390.9	14.65	14.56	14.605	1.06	0.99	1.025	171.6313604	2.277555798	431.3	387.2	30
FP 30	363.6	14.51	14.51	14.51	1.1	0.99	1.045	172.7113075	2.10524722	406.6	359.6	30
FP 31	385.2	14.66	14.64	14.65	1.04	0.97	1.005	169.3210558	2.274968096	427.9	381.7	30
FP 32	371.6	14.6	14.56	14.58	1.04	0.99	1.015	169.3755611	2.193941071	416.1	368.3	30
FP 33	381.8	14.63	14.59	14.61	1.01	1.06	1.035	173.4244949	2.201534449	423.2	377.8	30
FP 34	371.6	14.64	14.61	14.625	1	1.02	1.01	169.583182	2.191255026	419.1	366.9	30
FP 35	356.9	14.55	14.5	14.525	1.07	0.97	1.02	168.9281829	2.112732131	398.8	352.6	30
FP 36	381.4	14.61	14.57	14.59	1.06	0.98	1.02	170.4434877	2.237691831	428	377.6	30
FP 37	380.3	14.63	14.63	14.63	1.01	0.96	0.985	165.498682	2.297903496	416.9	375.8	30
FP 38	359.8	14.61	14.61	14.61	1.06	0.99	1.025	171.748896	2.094918852	407.1	356.4	30
FP 39	386.7	14.64	14.6	14.62	1.09	1.01	1.05	176.1788217	2.194928972	434	382.7	30

(continued)

## APPENDIX VII

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
FP 40	375.5	14.63	14.63	14.63	1.07	1.01	1.04	174.7397252	2.148910327	423	371.9	7
FP 41	364.9	14.55	14.59	14.57	1.09	0.9	0.995	165.8104283	2.200705974	410.7	361.5	7
FP 42	354	14.57	14.55	14.56	1.06	0.97	1.015	168.9112006	2.095775761	402.4	349.8	7
FP 43	398.5	14.65	14.65	14.65	1.09	1	1.045	176.0602023	2.26343032	440.6	395.2	7
FP 44	355.9	14.57	14.59	14.58	1.06	0.96	1.01	168.5411987	2.111649868	404.1	352.6	7
FP 45	381.5	14.57	14.56	14.565	1.02	1.06	1.04	173.1904633	2.202777178	424.9	378.1	7
FP 46	406.8	14.63	14.65	14.64	1.06	1.01	1.035	174.1374418	2.336085772	446.2	403.3	7
FP 47	373.8	14.52	14.63	14.575	1.06	0.99	1.025	170.9269916	2.186898607	419	370	7
FP 48	379.7	14.63	14.61	14.62	1.05	0.99	1.02	171.1451411	2.21858475	421.8	376	7
FP 49	383.3	14.6	14.56	14.58	1.06	1.02	1.04	173.547373	2.208618854	429.2	379.7	7
FP 50	339.1	14.65	14.61	14.63	1.07	1.01	1.04	174.7397252	1.940600511	433.5	386.4	7
FP 51	381.9	14.64	14.63	14.635	1.07	1	1.035	174.0185157	2.194594055	422.6	377.2	7
FP 52	387.7	14.61	14.6	14.605	1.09	1.01	1.05	175.8174911	2.205127587	433.7	384.1	7



## APPENDIX VIII

## Original data FujiCem (FC)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
FC 1	328.7	14.52	14.51	14.515	1.06	1.01	1.035	171.1764782	1.920240465	361.7	306.1	180
FC 2	330.3	14.51	14.55	14.53	1.06	1.01	1.035	171.5304532	1.925605592	362.8	304.7	180
FC 3	331.6	14.52	14.55	14.535	1.06	1.01	1.035	171.6485262	1.93185463	357.9	306.2	180
FC 4	311.7	14.47	14.52	14.495	0.99	0.96	0.975	160.8091335	1.938322738	339.2	286.8	180
FC 5	316.4	14.52	14.51	14.515	1.05	0.99	1.02	168.6956597	1.875566927	343.8	290.5	180
FC 6	333.3	14.47	14.5	14.485	1.06	0.99	1.025	168.8225754	1.974262027	364.3	307.4	180
FC 7	313.9	14.49	14.5	14.495	1.06	0.99	1.025	169.0557557	1.856783868	343.4	290.1	180
FC 8	307.9	14.47	14.42	14.445	1.01	0.96	0.985	161.3396014	1.90839693	338.5	284.4	180
FC 9	325.3	14.52	14.56	14.54	1.05	0.99	1.02	169.2772681	1.921699255	360.5	302.2	180
FC 10	333	14.47	14.52	14.495	1.06	1.01	1.035	170.7050802	1.950732806	lost		180
FC 11	318.7	14.47	14.42	14.445	1.04	1.06	1.05	171.9863771	1.853053744	346.2	291.1	180
FC 12	324.8	14.55	14.5	14.525	1.06	1.02	1.04	172.2405003	1.88573535	354.8	303.2	180
FC 13	335.6	14.55	14.56	14.555	1	1.09	1.045	173.7842311	1.931130332	368.9	311.2	180
FC 14	315.7	14.5	14.49	14.495	1.02	0.99	1.005	165.7571068	1.904594053	347.4	294.2	90
FC 15	318.4	14.52	14.52	14.52	1.01	0.99	1	165.501864	1.923845402	351	298.7	90
FC 16	320.6	14.47	14.52	14.495	1.02	1.01	1.015	167.4064313	1.915099662	352.2	298.6	90
FC 17	329.6	14.57	14.55	14.56	1	1.01	1.005	167.2470509	1.970737291	362.6	309.4	90
FC 18	325.7	14.57	14.47	14.52	1.01	1.04	1.025	169.6394106	1.919954796	360	301.1	90
FC 19	325.4	14.56	14.6	14.58	1.06	0.97	1.015	169.3755611	1.921174447	360.5	304.6	90
FC 20	315.2	14.52	14.52	14.52	1.06	0.96	1.01	167.1568826	1.885653735	346.4	293.1	90
FC 21	306.1	14.6	14.57	14.585	1	0.96	0.98	163.6472077	1.870487155	340.9	286.1	90
FC 22	394.6	14.55	14.54	14.545	0.96	0.92	0.94	156.1079287	2.527738361	325	274.7	90
FC 23	326.7	14.5	14.5	14.5	1.06	1.04	1.05	173.2985625	1.885185862	360.7	305.5	90
FC 24	330.4	14.55	14.56	14.555	1.09	1	1.045	173.7842311	1.90120817	365.8	308.5	90
FC 25	312.9	14.57	14.51	14.54	1.05	0.92	0.985	163.4687344	1.9141275	344.3	391.9	90
FC 26	307.7	14.49	14.52	14.505	1.01	0.96	0.985	162.6826932	1.891412012	338.5	285	90
FC 27	329.8	14.55	14.52	14.535	1.1	1	1.05	174.136186	1.89391997	368.1	317.8	30
FC 28	322.6	14.47	14.62	14.545	0.99	1.07	1.03	171.0544326	1.885949374	357.4	304.8	30
FC 29	304	14.39	14.45	14.42	1.06	0.98	1.02	166.4946755	1.825884216	348.3	296.6	30
FC 30	354.7	14.5	14.44	14.47	1.1	1.07	1.085	178.3349471	1.988953965	401.4	343	30
FC 31	325.6	14.58	14.63	14.605	1.05	0.97	1.01	169.1196819	1.925263791	366.5	314.8	30
FC 32	334.1	14.5	14.47	14.485	1.05	0.99	1.02	167.9990507	1.988701714	379.5	323.6	30
FC 33	339	14.47	14.45	14.46	1.09	1.01	1.05	172.3437513	1.966999079	383	329.4	30
FC 34	326.3	14.52	14.52	14.52	1.07	0.93	1	165.501864	1.971579003	368.9	316.7	30
FC 35	339.4	14.52	14.47	14.495	1.07	1.01	1.04	171.5297424	1.978665596	385.5	331.6	30
FC 36	306.5	14.54	14.56	14.55	1.01	0.97	0.99	164.5245979	1.862943316	346.7	297	30
FC 37	333.1	14.55	14.52	14.535	1.09	1.01	1.05	174.136186	1.912870654	375.8	321.3	30
FC 38	342.3	14.54	14.57	14.555	1.07	1.02	1.045	173.7842311	1.969683888	387.7	332.7	30
FC 39	328.9	14.42	14.47	14.445	1.09	0.98	1.035	169.5294289	1.94007614	373.5	318.4	30

(continued)

## APPENDIX VIII

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
FC 40	342.3	14.55	14.57	14.56	1.02	1.09	1.055	175.5677997	1.949674147	387	335.4	7
FC 41	327.7	14.42	14.43	14.425	1.01	1.04	1.025	167.4268729	1.957272416	368.8	319.2	7
FC 42	320.8	14.42	14.47	14.445	1.05	0.99	1.02	167.0724806	1.92012472	366.1	312.4	7
FC 43	342.9	14.45	14.46	14.455	1.09	1.02	1.055	173.0447024	1.981568896	386.8	335.2	7
FC 44	342.1	14.52	14.54	14.53	1.01	1.06	1.035	171.5304532	1.994398042	390.2	336.6	7
FC 45	338.1	14.56	14.54	14.55	1.06	1.04	1.05	174.4957856	1.937582611	384.4	330.2	7
FC 46	349.9	14.52	14.43	14.475	1.1	1.02	1.06	174.3462726	2.006925613	398.4	342.1	7
FC 47	334.7	14.42	14.5	14.46	1.06	1.04	1.05	172.3437513	1.942048943	380.1	326.6	7
FC 48	336.9	14.55	14.59	14.57	1.06	1.01	1.035	172.4761741	1.953313272	382.3	328.9	7
FC 49	340.4	14.45	14.52	14.485	1.07	1.09	1.08	177.8813478	1.913635152	385.9	332.5	7
FC 50	341.5	14.56	14.57	14.565	1.01	1.09	1.05	174.8557562	1.953038364	384.1	333.1	7
FC 51	344.3	14.55	14.47	14.51	1.1	1.01	1.055	174.3640473	1.974604314	388.8	336.3	7
FC 52	333.4	14.22	14.37	14.295	1.07	1.02	1.045	167.6309733	1.988892586	380	326.1	7

## APPENDIX IX

## Original data Maxcem Elite (MX)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
MX 1	389.9	14.96	15.01	14.985	1.1	1.07	1.085	191.2550404	2.038639082	399.5	384.4	180
MX 2	389.7	15.03	15.01	15.02	1.09	1.06	1.075	190.3785376	2.046974438	400.1	383.3	180
MX 3	397.3	15.08	15.03	15.055	1.1	1.09	1.095	194.825274	2.03926314	406.5	390.6	180
MX 4	381.8	15.01	14.99	15	1.09	1.1	1.095	193.404375	1.974102189	390.2	375.4	180
MX 5	394	14.9	15.01	14.955	1.09	1.09	1.09	191.3678552	2.058861973	404.1	388.2	180
MX 6	396	15.08	15.03	15.055	1.09	1.07	1.08	192.1564346	2.060820918	406.2	390.4	180
MX 7	402	15.02	15.03	15.025	1.1	1.09	1.095	194.0495935	2.071635363	410.9	396.1	180
MX 8	400.2	15.01	15.03	15.02	1.07	1.1	1.085	192.1495007	2.082753265	408.8	394.5	180
MX 9	379.8	14.99	15.01	15	1.01	1.04	1.025	181.040625	2.097871679	389.5	373.3	180
MX 10	395	14.99	15.01	15	1.09	1.09	1.09	192.52125	2.051721563	404.7	388.9	180
MX 11	393.6	15.08	15.06	15.07	1.07	1.09	1.08	192.5395342	2.044255491	403.6	387.7	180
MX 12	370.8	14.93	15.03	14.98	1	1.07	1.035	182.319715	2.033789928	381.2	364.6	180
MX 13	396.1	15.06	15.01	15.035	1.09	1.06	1.075	190.7589775	2.076442248	lost		90
MX 14	407.6	15.08	15.07	15.075	1.1	1.1	1.1	196.2352322	2.077098977	lost		90
MX 15	386.4	14.96	14.69	14.825	1.04	1.1	1.07	184.604736	2.093120732	lost		90
MX 16	391.3	15.01	15.01	15.01	1.07	1.04	1.055	186.5879103	2.0971348	lost		90
MX 17		15.07	15.03	15.05	1.09	1.09	1.09	193.8068641	0			0
MX 18		14.99	14.98	14.985	1.09	1.06	1.075	189.4923211	0			0
MX 19	392.8	15	15.04	15.02	1.1	1.06	1.08	191.2640191	2.053705667	404.5	387.7	90
MX 20	371.9	15.08	15.03	15.055	1.09	0.99	1.04	185.0395296	2.00984082	382.3	367.2	90
MX 21	384	15.03	15.03	15.03	1.1	1.04	1.07	189.745461	2.023763826	396.4	379.9	90
MX 22	390.2	15.06	15.08	15.07	1.1	1.07	1.085	193.430921	2.017257624	402.2	386.4	90
MX 23	388.2	15.01	15.04	15.025	1.06	1.03	1.045	185.1888815	2.096238159	399.1	383.6	90
MX 24	372.4	15.02	15.02	15.02	1.05	1.02	1.035	183.294685	2.031701028	382.9	367.9	90
MX 25	376.1	15.03	15.05	15.04	1.09	1.01	1.05	186.4466688	2.017198818	388.3	371.7	90
MX 26	386.2	15.07	15.01	15.04	1.1	1.04	1.07	189.9980339	2.032652612	397.5	381.9	90
MX 27	411.4	15.03	15.01	15.02	1.1	1.07	1.085	192.1495007	2.141041213	422.2	406.4	30
MX 28	409.3	14.99	15.01	15	1.09	1.01	1.05	185.45625	2.206989519	420.3	405.5	30
MX 29	383.2	14.96	14.99	14.975	1.06	0.99	1.025	180.4376591	2.123725179	393.8	379	30
MX 30	407.1	15.02	14.96	14.99	1.1	1.06	1.08	190.5007448	2.136999519	418.5	403.3	30
MX 31	411.7	15.03	15.03	15.03	1.1	1.06	1.08	191.518783	2.149658605	422.6	407.4	30
MX 32	398.9	14.98	14.94	14.96	1.06	1.05	1.055	185.3468901	2.152180702	410.3	394.4	30
MX 33	381.7	15.03	14.96	14.995	1.04	1.01	1.025	180.9199514	2.109772842	392.3	378.4	30
MX 34	407	15.04	15.03	15.035	1.1	1.05	1.075	190.7589775	2.133582416	419.1	402.1	30
MX 35	388.3	15.04	15.03	15.035	1.09	0.97	1.03	182.773718	2.124484878	399.8	385.1	30
MX 36	418.1	15.06	15.03	15.045	1.1	1.06	1.08	191.9012468	2.178724771	430	413.6	30
MX 37	408.9	15.03	14.98	15.005	1.1	1.06	1.08	190.8821912	2.14215898	421.2	405	30
MX 38	391.5	15.03	15.06	15.045	1.1	1.06	1.08	191.9012468	2.04011181	402.8	387.1	30
MX 39	386.3	15.03	15.01	15.02	1.09	0.97	1.03	182.4092034	2.11776595	398.3	382	30

(continued)

## APPENDIX IX

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
MX 40	409.7	15.05	15.07	15.06	1.1	1.07	1.085	193.1742962	2.120882581	423	406.5	7
MX 41	412.2	15.06	15.09	15.075	1.1	1	1.05	187.3154489	2.200565957	424.8	409.9	7
MX 42	397.1	15.03	15.01	15.02	1.1	1.05	1.075	190.3785376	2.085844366	407.9	393.9	7
MX 43	401.2	15.05	15.07	15.06	1.1	1.05	1.075	191.393888	2.09620069	412.7	397.9	7
MX 44	396	15.04	15.02	15.03	1.1	1.04	1.07	189.745461	2.087006445	408.1	392.8	7
MX 45	394.1	15.08	15.01	15.045	1.08	1.02	1.05	186.5706566	2.112336458	405.4	391.8	7
MX 46	391.6	15	15	15	0.1	1.05	0.575	101.559375	3.855872488	402.4	388.3	7
MX 47	402.8	15.03	15.01	15.02	1.1	1.06	1.08	191.2640191	2.105989416	414.7	399.3	7
MX 48	387.8	15.03	15.04	15.035	1.01	1.07	1.04	184.5482201	2.10134782	398.5	385.2	7
MX 49	411.9	15.05	15.08	15.065	1.1	1.06	1.08	192.411792	2.140721189	423.7	408.4	7
MX 50	387.5	15.01	15.01	15.01	1.1	1	1.05	185.7036074	2.086658441	398	384.5	7
MX 51	398.5	15.03	14.6	14.815	1.06	1	1.03	177.4639701	2.245526231	410	395.8	7
MX 52	403.1	15.04	15.1	15.07	1.09	1.06	1.075	191.6481475	2.103333663	416.5	400.2	7

## APPENDIX X

## Original data Panavia F (PF)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
PF 1	416.5	15.1	15.08	15.09	1.1	1.09	1.095	195.7321901	2.127907524	423.4	411.1	180
PF 2	422.5	15.1	15.1	15.1	1.11	1.11	1.11	198.6765135	2.12657245	430.5	419.1	180
PF 3	387.6	14.98	15.03	15.005	1.09	1.01	1.05	185.5799081	2.088588166	394.5	383.8	180
PF 4	402.6	15.03	15.08	15.055	1.09	1.09	1.09	193.9356608	2.075946209	409.7	399.5	180
PF 5	395.2	15.04	15.01	15.025	1.01	1.09	1.05	186.0749527	2.123875322	402.3	392	180
PF 6	415.5	15.08	15.1	15.09	1.1	1.1	1.1	196.6259444	2.113149419	422.6	412.4	180
PF 7	390.1	15.01	15.03	15.02	1.01	1.1	1.055	186.8366113	2.087920549	397.6	387.1	180
PF 8	419.4	15.08	15.04	15.06	1.1	1.1	1.1	195.8449086	2.141490443	427.3	416.7	180
PF 9	402.8	15.1	15.08	15.09	1.1	1.09	1.095	195.7321901	2.057913928	410.6	399.9	180
PF 10	413.5	15.08	15.03	15.055	1.13	1.09	1.11	197.4941133	2.093733292	420.8	409.9	180
PF 11	390.9	15.06	15.08	15.07	1.09	1.07	1.08	192.5395342	2.030232397	398.7	388.1	180
PF 12	390.3	15.08	15.03	15.055	1.1	1.06	1.08	192.1564346	2.031157587	398.2	386	180
PF 13	422.1	15.06	15.07	15.065	1.13	1.1	1.115	198.6473593	2.124870935	430.2	418.6	180
PF 14	403.1	15.07	15.08	15.075	1.07	1.06	1.065	189.9913839	2.121675161	410	400.4	90
PF 15	386.1	15.03	15.03	15.03	1.06	1.09	1.075	190.632122	2.025366953	393.3	382.8	90
PF 16	361.1	15.03	15.06	15.045	1.01	0.99	1	177.6863396	2.032232758	367.4	357.2	90
PF 17	411.2	15.1	15.08	15.09	1.1	1.09	1.095	195.7321901	2.10082971	418.1	408.3	90
PF 18	383.2	15.08	15.03	15.055	1.06	1.09	1.075	191.2668215	2.003483913	389.5	379.4	90
PF 19	397.6	15.08	15.08	15.08	1.06	1.09	1.075	191.9025758	2.071884644	404.6	394.7	90
PF 20	400.9	15.06	15.06	15.06	1.09	1.09	1.09	194.0645003	2.065808014	406.6	398.2	90
PF 21	381.6	15.03	15.08	15.055	1.01	1.06	1.035	184.1499165	2.07222467	387.5	378.6	90
PF 22	388.8	15.08	15.09	15.085	1.02	1.07	1.045	186.6708806	2.082810124	395.2	385.8	90
PF 23	382.5	15.1	15.08	15.09	1.01	1.09	1.05	187.6884014	2.03795225	388.5	379.5	90
PF 24	397.2	15.06	15.08	15.07	1.06	1.09	1.075	191.6481475	2.072548079	403.4	393.5	90
PF 25	385.6	15.08	15.07	15.075	1.01	1.06	1.035	184.6395139	2.088393713	392.6	381.7	90
PF 26	396.6	15.03	15.04	15.035	1.09	1.04	1.065	188.9844754	2.098585078	403.2	393.8	90
PF 27	399.5	15.1	15.12	15.11	1.06	1.04	1.05	188.1862484	2.122896882	406.4	398.1	30
PF 28	402.4	15.11	15.08	15.095	1.06	1	1.03	184.2354147	2.184162045	408.6	400.3	30
PF 29	394.5	15.07	15.08	15.075	1.06	1.04	1.05	187.3154489	2.10607295	400.1	392.1	30
PF 30	390.3	15.08	15.08	15.08	1.06	1	1.03	183.8694447	2.122701793	396.1	386.9	30
PF 31	394.6	15.1	15.08	15.09	1	1.09	1.045	186.7946471	2.112480235	401.5	392.8	30
PF 32	390.3	15.06	15.06	15.06	1.09	0.99	1.04	185.162459	2.107878681	397.4	387.7	30
PF 33	401.3	15.1	15.08	15.09	1.1	1.06	1.08	193.0509272	2.078726095	407.9	399.4	30
PF 34	403.6	15.1	14.98	15.04	1.09	1.05	1.07	189.9980339	2.124232507	409.9	402	30
PF 35	409.6	15.01	15.08	15.045	1.09	1.05	1.07	190.1243834	2.154379111	416.2	407	30
PF 36	490.3	15.06	15.08	15.07	1.07	1.04	1.055	188.0826006	2.606833373	397.2	388.8	30
PF 37	390.9	15.04	15.04	15.04	1.09	1.05	1.07	189.9980339	2.05738971	397.3	387.6	30
PF 38	419.5	15.08	15.08	15.08	1.1	1.1	1.1	196.3654264	2.136323118	426.6	417.1	30
PF 39	408.3	15.08	15.1	15.09	1.09	1.06	1.075	192.1571729	2.124823101	415.5	406.1	30

(continued)

## APPENDIX X

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
PF 40	417.2	15.08	15.08	15.08	1.1	1.07	1.085	193.687716	2.153982754	421.8	415.8	7
PF 41	411.9	15.08	15.08	15.08	1.01	1.09	1.05	187.4397252	2.197506423	417.2	409.8	7
PF 42	411.4	15.07	15.04	15.055	1.11	1.04	1.075	191.2668215	2.150921926	416.3	409.7	7
PF 43	409.8	15.06	15.06	15.06	1.11	1.04	1.075	191.393888	2.141134204	414.8	408.1	7
PF 44	417	15.06	15.06	15.06	1.1	1.1	1.1	195.8449086	2.129235848	422.6	415.7	7
PF 45	406	15.01	15.03	15.02	1.9	1.02	1.46	258.5606184	1.570231393	411	403.8	7
PF 46	414.4	15.03	15.03	15.03	1.1	1.05	1.075	190.632122	2.173820423	419.2	412.7	7
PF 47	400.6	15.03	15.08	15.055	1.05	1.09	1.07	190.3772083	2.104243483	405.7	398	7
PF 48	407.3	15.08	15.08	15.08	1.1	1.06	1.08	192.7951459	2.112605056	412.1	406	7
PF 49	420.6	15.06	15.04	15.05	1.1	1.07	1.085	192.9178418	2.180202702	424.3	419.5	7
PF 50	411	15.03	15.03	15.03	1.09	1.04	1.065	188.8587999	2.176229014	415.7	409.8	7
PF 51	382.6	15.02	14.99	15.005	1.07	0.98	1.025	181.1613389	2.111929634	387.7	380.5	7
PF 52	416.2	15.1	15.08	15.09	1.1	1.01	1.055	188.5821557	2.206995664	420.8	413.8	7

## APPENDIX XI

## Original data Rely X ARC (RA)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
RA 1	367.7	15.03	15.08	15.06	1.1	1.1	1.063461538	189.2138681	1.943303647	373.3	366.8	180
RA 2	362.2	15.08	15.06	15.07	1.09	1.04	1.063461538	189.5911012	1.910427218	367.8	361.4	180
RA 3	338.9	15.08	15.07	15.08	1.02	1.01	1.063461538	189.716929	1.786345593	344.5	338.1	180
RA 4	322.1	15.1	15.08	15.09	1.05	1.02	1.063461538	190.094663	1.694418954	327.3	321.4	180
RA 5	324.2	15.04	15.08	15.06	1.01	1.06	1.063461538	189.3395707	1.712267535	329.4	323.4	180
RA 6	354.3	15.06	14.98	15.02	1.06	1.09	1.063461538	188.3351185	1.881221106	359.8	353.6	180
RA 7	370.9	15.08	15.1	15.09	1.09	1.07	1.063461538	190.094663	1.951133157	376.5	370.1	180
RA 8	349.2	15.02	15.08	15.05	1.06	1.02	1.063461538	189.0882072	1.846757157	354.6	348.4	180
RA 9	349.9	15.03	15.03	15.03	1.04	1.05	1.063461538	188.5859811	1.855387118	355.5	349.1	180
RA 10	339.7	15.03	15.04	15.04	1.06	1.09	1.063461538	188.7114751	1.80010251	345.4	338.7	180
RA 11	362.6	15.1	15.08	15.09	1.1	1.07	1.063461538	190.094663	1.907470701	368	361.8	180
RA 12	336.1	15.08	15.1	15.09	1.04	1.07	1.063461538	190.094663	1.768066471	341.3	335.3	180
RA 13	363	15.03	15.06	15.05	1.07	1.1	1.063461538	188.9625881	1.921015179	368.3	362.1	180
RA 14	336.8	15.08	15.1	15.09	0.98	1.07	1.063461538	190.094663	1.771748847	341.7	335.6	90
RA 15	337.6	15.08	15.04	15.06	1.07	1.01	1.063461538	189.3395707	1.783039851	342.7	336.6	90
RA 16	349.6	15.08	15.04	15.06	1.06	1.04	1.063461538	189.3395707	1.846418045	354.9	348.6	90
RA 17	330.3	15.07	15.04	15.06	1.05	1.09	1.063461538	189.2138681	1.745643717	335.2	329.1	90
RA 18	362.9	15.08	15.11	15.10	1.07	1.09	1.063461538	190.2206578	1.907784382	368.2	362.1	90
RA 19	357.8	15.1	15.08	15.09	1.05	1.09	1.063461538	190.094663	1.882220123	362.8	356.9	90
RA 20	351	15.1	15.08	15.09	1.09	1	1.063461538	190.094663	1.846448472	356.3	350	90
RA 21	355.2	15.01	15.04	15.03	1.06	1.09	1.063461538	188.460529	1.884744789	360.5	354	90
RA 22	364.4	15.06	15.03	15.05	1.1	1.07	1.063461538	188.9625881	1.928424053	369.9	363.1	90
RA 23	351.5	15.07	15.08	15.08	1.1	1.06	1.063461538	189.716929	1.85276033	356.8	350.4	90
RA 24	350.1	15.06	15.08	15.07	1.09	1.09	1.063461538	189.5911012	1.846605657	355.5	349.1	90
RA 25	357	15.01	15.06	15.04	1.01	1.05	1.063461538	188.7114751	1.891776851	362.1	355.8	90
RA 26	345.6	15.06	15.06	15.06	1.01	1.07	1.063461538	189.3395707	1.82529198	350.4	344.4	90
RA 27	373.6	15.08	15.08	15.08	1.1	1.09	1.063461538	189.8427986	1.967944019	378.5	372.2	30
RA 28	353.5	15.03	15.06	15.05	1.1	1.01	1.063461538	188.9625881	1.870740677	357.5	351.2	30
RA 29	379.6	15.03	15.1	15.07	1.1	1.09	1.063461538	189.4653151	2.003532941	384.5	378.1	30
RA 30	333.8	15.03	15.03	15.03	1.1	0.95	1.063461538	188.5859811	1.770014918	338.1	331.9	30
RA 31	337.4	15.1	15.03	15.07	1.09	1.1	1.063461538	189.4653151	1.780800881	341	334.5	30
RA 32	372.7	15.08	15.1	15.09	1.09	1.07	1.063461538	190.094663	1.960602124	377.4	371.1	30
RA 33	366	15.03	15.1	15.07	1.05	1.07	1.063461538	189.4653151	1.931751993	370.8	364.4	30
RA 34	357.4	15.06	15.08	15.07	1.07	1.04	1.063461538	189.5911012	1.885109574	361.5	355.4	30
RA 35	367.4	15.06	15.08	15.07	1.1	1.05	1.063461538	189.5911012	1.937854666	371.7	365.4	30
RA 36	362.9	15.03	15.06	15.05	1.01	1.09	1.063461538	188.9625881	1.920485974	367.2	360.9	30
RA 37	348.4	15.08	15.1	15.09	1.1	0.96	1.063461538	190.094663	1.832771076	352.6	346.6	30
RA 38	376.1	15.07	15.06	15.07	1.1	1.1	1.063461538	189.4653151	1.985059903	380.6	373.6	30
RA 39	364.4	15.07	15.1	15.09	1.09	1	1.063461538	189.9687099	1.918210637	368.9	362.3	30

(continued)

## APPENDIX XI

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
RA 40	349.7	15.03	15.03	15.03	1.0 4	1.07	1.063461538	188.5859811	1.854326594	353.5	346.3	7
RA 41	340.6	15.03	15.03	15.03	1.1	1.05	1.063461538	188.5859811	1.806072742	344.6	337.1	7
RA 42	376.7	15.07	15.06	15.07	1.1	1.1	1.063461538	189.4653151	1.98822671	381.1	373.4	7
RA 43	358.1	15.08	15.01	15.05	1.0 5	1.01	1.063461538	188.9625881	1.89508412	362	356.2	7
RA 44	370.2	15.03	15.06	15.05	1.0 9	1.09	1.063461538	188.9625881	1.959117959	374.6	368.6	7
RA 45	350.7	15.03	15.02	15.03	1.0 9	1.02	1.063461538	188.460529	1.860867111	353.9	346.6	7
RA 46	369.2	15.03	15.06	15.05	1.1	1.05	1.063461538	188.9625881	1.953825907	373.5	366.5	7
RA 47	354.2	15.08	15.06	15.07	1.0 5	0.99	1.063461538	189.5911012	1.868231145	358.4	351.5	7
RA 48	372.1	15.06	15.04	15.05	1.1	1.05	1.063461538	189.0882072	1.967864657	376.5	369.8	7
RA 49	378.3	15.08	15.03	15.06	1.1	1.04	1.063461538	189.2138681	1.999324911	381.9	375.7	7
RA 50	362.6	15.03	15.03	15.03	1.1	1.06	1.063461538	188.5859811	1.922730406	366.7	360	7
RA 51	380.6	15.1	15.1	15.10	1.1	1.05	1.063461538	190.3466943	1.999509376	385.2	378.7	7
RA 52	358.2	15.03	15.03	15.03	1.1	1.04	1.063461538	188.5859811	1.899398873	362.4	356	7



## APPENDIX XII

## Original data Rely X Luting Plus (RL)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
RL 1	317.6	14.36	14.31	14.335	1.02	1	1.01	162.9245106	1.94936906	361.2	304.1	180
RL 2	290	14.33	14.31	14.32	1.01	0.91	0.96	154.5350246	1.876597235	332	374.1	180
RL 3	305.6	14.27	14.27	14.27	1.09	0.95	1.02	163.048863	1.874284765	349.1	292.3	180
RL 4	298.2	14.32	14.32	14.32	1.01	0.95	0.98	157.7545043	1.890278831	339.4	284.9	180
RL 5	325.6	14.36	14.35	14.355	1.06	1.04	1.05	169.8499211	1.916986466	369.7	311.3	180
RL 6	309.7	14.38	14.36	14.37	1	1.05	1.025	166.1525682	1.863949522	353.8	294.7	180
RL 7	307.7	14.36	14.37	14.365	1	0.97	0.985	159.5574724	1.928458726	351	293.9	180
RL 8	298.8	14.4	14.42	14.41	1	0.96	0.98	159.7436833	1.870496496	339.8	286.4	180
RL 9	322.1	14.32	14.3	14.31	1.01	1.07	1.04	167.179208	1.926674996	367.9	309.5	180
RL 10	295.6	14.38	14.37	14.375	1.01	0.93	0.97	157.3465039	1.878656295	330.7	280.2	180
RL 11	312.9	14.37	14.34	14.355	1.07	0.99	1.03	166.6146845	1.87798573	348.4	298.3	180
RL 12	324.2	14.4	14.41	14.405	1.01	1.05	1.03	167.7773794	1.932322469	367.7	310.6	180
RL 13	298.4	14.4	14.41	14.405	0.9	1.05	0.975	158.8183931	1.878875577	340.3	285.1	180
RL 14	326.7	14.36	14.37	14.365	1.05	1.05	1.05	170.0866457	1.920785719	371.6	311.9	90
RL 15	303.7	14.35	14.36	14.355	1.02	0.98	1	161.7618296	1.877451564	345.9	289.6	90
RL 16	312.1	14.34	14.34	14.34	1.09	1.01	1.05	169.4951433	1.841350696	354.9	299.2	90
RL 17	322.4	14.43	14.44	14.435	1.06	0.99	1.025	167.6590877	1.922949746	367.2	307.8	90
RL 18	314.9	14.35	14.37	14.36	1.05	1.02	1.035	167.5401448	1.879549528	358.7	300.8	90
RL 19	323.5	14.42	14.38	14.4	1.05	1.01	1.03	167.660928	1.929489499	368.8	310	90
RL 20	310.3	14.39	14.37	14.38	1.05	1.01	1.03	167.1955266	1.855910898	354.1	295.3	90
RL 21	324.5	14.38	14.44	14.41	1.05	1.03	1.04	169.5239088	1.914184272	369.7	310.5	90
RL 22	321.4	14.47	14.45	14.46	1.09	0.99	1.04	170.7023822	1.882809108	364.6	307.5	90
RL 23	316.8	14.47	14.45	14.46	1.04	0.96	1	164.136906	1.930096087	361.7	302.5	90
RL 24	316.2	14.4	14.38	14.39	1.09	1	1.045	169.8664204	1.861462667	361	302	90
RL 25	302.9	14.35	14.39	14.37	0.96	1.03	0.995	161.2895662	1.877988807	344.3	289.5	90
RL 26	312.6	14.36	14.37	14.365	1.02	0.94	0.98	158.747536	1.969164422	356.4	299.7	90
RL 27	306.2	14.29	14.3	14.295	1	0.97	0.985	158.0062284	1.937898291	353.1	299.5	30
RL 28	328.7	14.39	14.34	14.365	1.06	1	1.03	166.8469001	1.970069566	377.2	321.3	30
RL 29	313.2	14.3	14.3	14.3	1.05	0.97	1.01	162.1298965	1.93178437	362.9	305.5	30
RL 30	373.8	14.33	14.27	14.3	1.08	0.95	1.015	162.9325198	2.294201308	315.7	266.4	30
RL 31	312.9	14.35	14.36	14.355	1.01	1.06	1.035	167.4234937	1.868913336	356.9	304.6	30
RL 32	317.7	14.28	14.26	14.27	1.06	1	1.03	164.6473813	1.929578214	366.7	310.2	30
RL 33	319.2	14.36	14.36	14.36	1.07	1	1.035	167.5401448	1.905215019	366	312.6	30
RL 34	325.2	14.33	14.34	14.335	1.06	1	1.03	166.1507385	1.957258829	374	318.1	30
RL 35	308.9	14.37	14.3	14.335	1	0.99	0.995	160.5048396	1.92455256	357.3	301.5	30
RL 36	326.4	14.31	14.34	14.325	1.02	1.04	1.03	165.9190081	1.967224875	375.7	319.8	30
RL 37	314	14.32	14.27	14.295	1.01	1.08	1.045	167.6309733	1.873162184	362.6	307.4	30
RL 38	317.9	14.38	14.4	14.39	1.01	0.99	1	162.5515985	1.955686705	365	311.3	30
RL 39	306.3	14.33	14.26	14.295	1.05	0.96	1.005	161.2144767	1.899953443	354.8	298.4	30

(continued)

## APPENDIX XII

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
RL 40	327.9	14.39	14.33	14.36	1.06	1	1.03	166.7307721	1.966643565	375.7	322.6	7
RL 41	330.5	14.48	14.45	14.465	1.06	1.03	1.045	171.6417063	1.92552269	378.8	325.3	7
RL 42	331.9	14.35	14.43	14.39	1.07	1.02	1.045	169.8664204	1.953888233	381.1	326.5	7
RL 43	304.9	14.24	14.25	14.245	1	1	1	159.2922196	1.914092231	353.1	300	7
RL 44	312.6	14.33	14.3	14.315	1.04	1	1.02	164.0788235	1.905181872	362.1	307.6	7
RL 45	321.9	14.42	14.38	14.4	1.08	0.97	1.025	166.84704	1.929312021	371.1	316.6	7
RL 46	317.1	14.3	14.25	14.275	1.05	1	1.025	163.9629623	1.933973354	365.7	311.2	7
RL 47	303.1	14.3	14.31	14.305	0.97	1.05	1.01	162.2432939	1.868181992	348.7	296.6	7
RL 48	310	14.3	14.3	14.3	1.04	0.98	1.01	162.1298965	1.91204711	355.9	302.8	7
RL 49	286.2	14.39	14.38	14.385	1.01	0.9	0.955	155.1289171	1.844917153	328.7	280.9	7
RL 50	306.6	14.28	14.3	14.29	1.01	0.97	0.99	158.6972163	1.931980958	352.7	300.7	7
RL 51	311.2	14.4	14.4	14.4	1.06	1	1.03	167.660928	1.856127147	356.2	304.5	7
RL 52	312.9	14.37	14.37	14.37	1.06	0.93	0.995	161.2895662	1.939989098	360.2	305.2	7

## APPENDIX XIII

## Original data Rely X Unicem (RU)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
RU 1	382.3	15.08	15.07	15.08	1.1	1.09	1.095	195.3432539	1.957067841	396.5	381.4	180
RU 2	361.5	15.01	14.93	14.97	1.06	1	1.03	181.1967827	1.995068536	374.1	360.9	180
RU 3	373.8	15.08	15.07	15.08	1.06	1.01	1.035	184.6395139	2.024485399	386.8	373.2	180
RU 4	362.3	15.03	14.99	15.01	0.96	0.99	0.975	172.439064	2.101032049	376	361.2	180
RU 5	400.3	15.08	15.07	15.08	1.1	1.06	1.08	192.6673189	2.077674628	415.4	399.6	180
RU 6	401.2	15.08	15.08	15.08	1.09	1.05	1.07	191.0100057	2.100413528	416	400.4	180
RU 7	368.4	15.03	15.08	15.06	1.04	0.99	1.015	180.591464	2.039963528	381.4	367.4	180
RU 8	397.4	15.06	15.08	15.07	1.09	1.05	1.07	190.7567608	2.083281339	411.7	396.5	180
RU 9	389.7	15.08	15.08	15.08	1.1	0.99	1.045	186.5471551	2.089016044	403.4	388.7	180
RU 10	392.4	15.08	15.03	15.06	1.09	1.06	1.075	191.2668215	2.051584258	406.3	391.7	180
RU 11	385.8	15.08	15.08	15.08	1.07	0.99	1.03	183.8694447	2.098227906	400.1	385	180
RU 12	374.7	15.04	15.01	15.03	1.06	1	1.03	182.5306678	2.052805725	393.3	373.7	180
RU 13	382.6	15.03	15.03	15.03	1.04	1	1.02	180.8788506	2.115227948	395	381.7	90
RU 14	410.1	15.09	15.06	15.08	1.1	1.06	1.08	192.6673189	2.128539507	422.9	409.5	90
RU 15	409.2	15.03	15.1	15.07	1.09	1.04	1.065	189.739406	2.156642148	419.9	408.9	90
RU 16	401.7	15.07	15.02	15.05	1.1	1.04	1.07	190.1243834	2.112827365	414.1	401	90
RU 17	383.7	15.03	15.03	15.03	1	1.09	1.045	185.3121558	2.070560338	397	382.9	90
RU 18	402.2	15.1	15.09	15.10	1.1	1.04	1.07	191.390188	2.101466142	415.6	401.4	90
RU 19	405.4	15.08	15.05	15.07	1.1	1.07	1.085	193.3025873	2.097230077	418	404.8	90
RU 20	398.4	15.1	15.05	15.08	1.09	1.04	1.065	189.9913839	2.096937197	412.2	397.5	90
RU 21	414.8	15.04	15.03	15.04	1.1	1.09	1.095	194.3079817	2.134755332	426.8	414.3	90
RU 22	407.4	15.1	15.1	15.10	1.1	1.04	1.07	191.5169995	2.127226309	418.8	406.9	90
RU 23	385.9	15.08	15.06	15.07	1.05	1.03	1.04	185.4084404	2.081350769	399.2	385.1	90
RU 24	355.8	15.08	15.08	15.08	1.07	0.97	1.02	182.0843045	1.954039921	365.4	355.5	90
RU 25												
RU 26												
RU 27	388.6	15.13	15.15	15.14	1.1	1.01	1.055	189.8339422	2.047052258	398.6	388.3	30
RU 28	409.8	15.09	15.05	15.07	1.1	1.03	1.065	189.865374	2.158371436	418.3	409.7	30
RU 29	417.2	15.12	15.12	15.12	1.09	1.04	1.065	191.1273538	2.182837735	427.9	416.8	30
RU 30	399.6	15.06	15.07	15.07	1.1	1.03	1.065	189.739406	2.106046438	409.2	399.4	30
RU 31	412.7	15.1	15.09	15.10	1.1	1.01	1.055	188.707148	2.186986578	423.2	412.6	30
RU 32	427.1	15.1	15.09	15.10	1.1	1.07	1.085	194.0732281	2.2007157	437.4	426.8	30
RU 33	378.4	15.1	15.07	15.09	1.08	1.06	1.07	191.1366911	1.979735014	388	377.9	30
RU 34	406	15.1	15.07	15.09	1.1	1.01	1.055	188.4572048	2.154335253	416.2	405.9	30
RU 35	394.7	15.1	15.06	15.08	1.1	0.96	1.03	183.8694447	2.146631816	404.4	394.4	30
RU 36	392	15.07	15.05	15.06	1.1	1.06	1.08	192.2840921	2.038650186	401.9	391.6	30
RU 37	414.4	15.1	15.09	15.10	1.1	1.06	1.08	193.1788814	2.145162023	423	414.2	30
RU 38	366.9	15.08	15.09	15.09	1.1	1.03	1.065	190.243529	1.928580708	377	366.5	30
RU 39	417.1	15.07	15.05	15.06	1.1	1.05	1.075	191.393888	2.179275443	427.5	416.8	30

(continued)

## APPENDIX XIII

(continued)

	W0	D1	D2	D	T1	T2	T	V	P	W1	W2	St. period
RU 40	413.8	15.1	15.09	15.10	1.1	1.09	1.095	195.8619214	2.112712859	421.7	413.6	7
RU 41	397.8	15.14	15.14	15.14	1.1	1.02	1.06	190.7336292	2.085631159	405.2	397.6	7
RU 42	405.1	15.12	15	15.06	1.1	1.08	1.09	194.0645003	2.087450303	412.5	405	7
RU 43	409.5	15.17	15.16	15.17	1.1	1	1.05	189.5587277	2.160280378	417.1	409.5	7
RU 44	406.1	15	15.02	15.01	1.1	1.05	1.075	190.1251219	2.135961813	413.5	405.8	7
RU 45	433.7	15.13	15.12	15.13	1.1	1.1	1.1	197.5391172	2.19551452	441.3	433.4	7
RU 46	420.8	15.05	15.07	15.06	1.1	1.05	1.075	191.393888	2.198607304	427.4	420.7	7
RU 47	401.4	15.1	15.1	15.10	1.1	1.05	1.075	192.4119388	2.086149137	408.7	401.3	7
RU 48	397.3	15.07	15.08	15.08	1.1	0.96	1.03	183.7475356	2.16220587	404.7	396.6	7
RU 49	314.4	15.04	15.05	15.05	1.0 9	0.98	1.035	183.9053615	1.709574954	321.1	314	7
RU 50	411.4	15.01	15.02	15.02	1.1	1	1.05	185.827348	2.213882965	419.2	411.3	7
RU 51	421.6	15.21	14.35	14.78	1.1	1.1	1.1	188.6301934	2.23506106	428.8	421.3	7
RU 40	413.8	15.1	15.09	15.10	1.1	1.09	1.095	195.8619214	2.112712859	421.7	413.6	7