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Original Article

Physicochemical Properties, Sealing Ability, Bond Strength and Cytotoxicity of a New Dimethacrylate-based Root Canal Sealer

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Background/Purpose: Resin-based root canal sealer can bond to dentin and establish a hermetic seal. The aim of this study was to evaluate the physicochemical properties, sealing ability, cytotoxicity and bond strength of a new resin-based root canal sealer (NRCS).

Methods: The physicochemical properties were assessed by the flow, setting time, solubility, film thickness, radiopacity and dimensional changes. Sixty premolar root samples were filled with either Resilon/NRCS or Resilon/Epiphany and sectioned perpendicularly at the long axis at 2 mm below the cemento-enamel junction into 1-mm serial slices. The bond strength was tested by a universal testing machine. The glucose microleakage model was used to test the sealing ability. Elutes of NRCS and Epiphany were co-cultured with human periodontal ligament cells to test the cytotoxicity.

Results: All the physicochemical properties of NRCS conformed to ISO 6876:2001(E). The root samples filled with Resilon/NRCS had significantly less leakage ($p < 0.01$) and greater bond strength ($p < 0.001$) than the Resilon/Epiphany group had. Environmental scanning electron microscopy showed that Resilon/NRCS filling material was intimately bonded to the root dentin. Although NRCS was slightly toxic to human periodontal ligament cells, its cytotoxicity was significantly less than that of Epiphany ($p < 0.01$)

Conclusion: NRCS has better physicochemical and sealing properties, as well as lower cytotoxicity and microleakage than Epiphany has.

Key Words: cytotoxicity, dimethacrylate, physicochemical property, root canal sealer, sealing ability

Successful root canal therapy requires a hermetic, three-dimensional obturation of the canal with a non-irritating biomaterial.[1] Ideally, the filling material and/or sealer should chemically adhere to the dentinal surface, thus eliminating any space that could allow penetration of fluids or microorganisms and induce a periapical inflammatory reaction.[2] The most commonly used filling material in the endodontic treatment is gutta-percha (GP) cones in combination with a sealer. Since GP

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does not bond to the dentin walls, a hermetic seal cannot be achieved without a sealer.[3] Moreover, most percolation apparently takes place at the cement–dentinal wall interface or the GP–sealer interface, which implicates the sealer as the weakest link in long-term successful obturation of a root canal.[4] Different types of sealers such as zinc oxide–eugenol cements, epoxy resin sealers, and calcium hydroxide sealers have been used.[5–10] Although adhesive resins have potential as root canal fillings, none meets the requirements of an ideal material.[11–14]

Recently, a new root canal filling material, Resilon with Epiphany sealer, has been introduced.[15] Resilon is based on polymers and contains a bifunctional methacrylate resin, bioactive glass and radiopaque fillers. The constituents of Epiphany are bisphenol-A–glycidyl dimethacrylate (bis-GMA), ethoxylated bis-GMA, urethane dimethacrylate (UDMA), hydrophilic bifunctional methacrylates, and fillers of calcium hydroxide, barium sulfate, barium glass, and silica (Table 1).[11,16] These also are the main constituents of most commercially available composite resins, and bis-GMA is the most frequently used cross-linker in restorative composites.[17] This thermoplastic root canal filling material is capable of coupling to self-etching dentin adhesives and resin cement-type sealers.[15,16,18] The use of dual-curable methacrylate sealer (Epiphany) to bind chemically to the thermoplastic filled polymer (Resilon cones) results in improved apical seals. The efficacy of Resilon/Epiphany has been assessed in many *in vitro* studies. Compared to GP, this material appears to have less apical leakage[15,19] and more resistance to fracture.[16] However, there are no significant differences in sealing ability,[20–25] resistance to fracture, and cytotoxicity[26,27] between Resilon/Epiphany and GP/AH Plus.

The drawbacks of resin monomers are high water susceptibility, low biocompatibility and shrinkage on curing.[28] A volumetric shrinkage of 2% in a composite resin is sufficient to compromise the marginal integrity between restorative materials and tooth structure, and as a result, microleakage and restoration failure can occur.[29] Anic et al[30] used the composite resin as a root filling material and have demonstrated that resin can penetrate into the dentinal tubules, but polymerization contraction prevents complete adhesion. Li et al[31] reported that, in their sensitivity analysis of composite resin, polymerization shrinkage is the most important factor in the development of residual stresses. Attempts are being made to reduce the shrinkage. One method involves the development of new base monomers with higher molecular weight than that of bis-GMA.[32,33] The polymerization shrinkage decreases when the molecular weight is increased.[33,34] To reduce the polymerization shrinkage by increasing the biocompatibility of dimethacrylate-based root canal sealer, we have recently synthesized a new monomer, 9,9′-bis[4-(21-hydroxy-31-methacryloxy-propyloxy)phenyl] fluorine (HPFDM) with a molecular weight of 634, using 9,91-bis(4-hydroxyphenyl)fluorine, epichlorohydrin and methacrylic acid in two steps. The structure of HPFDM has been confirmed by Fourier transform infrared spectroscopy, proton

Table 1. Component of root canal sealer tested

Sealer	Material group	Composition
Epiphany	Multi-methacrylates	Bisphenol-A-GMA, ethoxylate bis-GMA, urethane dimethacrylate resin, silane-treated barium borosilicate glasses, barium sulfate, silica, calcium hydroxide, bismuth oxychloride with amines peroxide, photoinitiator stabilizers, pigments
NRCS	Multi-methacrylates	HPFDM, bisphenol-A-glycidyl methacrylate, triethylene glycol dimethacrylate, barium sulfate, hydroxyapatite, silicon dioxide, zinc oxide, N,N-dimethyl-ethylamine methacrylate, camphorquinone

GMA = glycidyl methacrylate; HPFDM = 9,9′-bis[4-(21-hydroxy-31-methacryloxy-propyloxy)phenyl] fluorine; NRCS = new resin-based root canal sealer.

nuclear magnetic resonance, mass spectroscopy and element analysis.[35] We have also compared the polymerization of the new resin systems HMPF/TEGDMA/CQ/DMAEMA and the conventional product BIS-GMA/TEGDMA/CQ/DMAEMA, with the former being 6.19 ± 0.23% and the later being 1.43 ± 0.06%.[36] Consequently, we developed a new sealer, new resin-based canal sealer (NRCS), which consisted mainly of the resin matrix HPFDM, Bis-GMA, TEGDMA and fillers including barium sulfate, silicon dioxide, and hydroxyapatite ceramic. However, the biological and physiochemical properties of this new sealer have not been completely determined. The aims of this study were to test the physiochemical properties, sealing ability, bond strength, and cytotoxicity of NRCS. We hope that these data provide the basis for future clinical trials.

Methods

Physicochemical properties

The flow, setting time, solubility, film thickness, radiopacity and dimensional changes for NRCS and Epiphany were determined according to the method described by the International Organization for Standardization [ISO 6876:2001 (E)] for dental root canal sealing materials. For each material, the solubility, flow, film thickness and setting time were measured four times, and rationality and dimensional change, were measured three times. The results are presented in Table 2.

Sealing ability test

Sixty extracted single-rooted, straight, non-carious human mandibular premolars from patients aged 18–25 years old were used. They were inspected under a microscope for cracks and fractures. The crowns were removed at the cemento-enamel junction by a diamond saw (Isomet; Buehler, Lake Bluff, IL, USA). The root length was determined by inserting a size 15 Flex-O-File (Dentsply Maillefer, Tulsa, OK, USA) into the root canal until it was visible at the apex. The working length was established by subtracting 1 mm from the real length. All the roots were performed in a crown-down manner with 0.04 taper ProFile nickel–titanium rotary instruments (Dentsply Maillefer). The apical third was prepared with ProFile hand instruments up to size 45, 0.04 taper (Dentsply Maillefer). Each root canal was irrigated with 15 mL 1.25% NaOCl followed by 5 mL 17% EDTA (pH 7.4). After final instrumentation, all teeth received the above irrigation. The canals were then flushed with 10 mL sterile water to remove the remaining NaOCl.

The teeth were divided randomly into two experimental groups of 20 teeth each: one positive and one negative control group of 10 roots each. In group 1, the canals were filled with Resilon/Epiphany using the lateral condensation technique. Those in group 2 were obturated with Resilon/NRCS with the same procedure as in group 1. The coronal portion of the obturation was light-cured for 40 seconds, and radiographs were taken to verify the quality and apical extent of the root fillings. The teeth of the negative control group

Table 2. Physicochemical properties of NRCS and Epiphany

	ISO standards	NRCS	Epiphany
Setting time	When ≤ 30 min, ≤ 110% stated by the manufacturer	75.55	28.03
	When > 30 min < 72 hr, within the range (min)		
Flow	≥ 20 min	37.21	38.22
Film thickness	≤ 50 μm	50.55	27.60
Solubility	≤ 3%	0.50	3.84
Dimensional change	Shrinkage ≤ 1%	0.92	7.56
	Expansion ≤ 0.1%		
Radiopacity	≥ 3 mm aluminum	8.80	9.20

NRCS = new resin-based root canal sealer; ISO = International Organization for Standardization.

were filled as for group 1 with sticky wax on the whole surface. The 10 roots in the positive control group were obturated with Resilon only. All roots were stored for 7 days in gauze dampened with sterile saline.

The apical sealing ability was determined by using a microleakage model based on the filtration of glucose along the root canal fillings as described by Xu et al.[37] The glass bottle contained 0.01% NaN_3 to inhibit the growth of microorganisms, with the tracer substance 1 mol/L glucose solution (pH 7.0 and density 1.09×10^3 g/L). The model was placed in an incubator at 37°C and 100% humidity. On days 1, 4, 15, 20 and 30, 10 μL of the solution was removed by a micropipette. Fresh 0.01% NaN_3 (10 μL) was added to maintain a constant volume of 1.0 mL. The samples were analyzed with a Glucose kit (Diasys, Shanghai, China) and a UV-Vis Recording Spectrophotometer (Schimadzu, Kyoto, Japan).

Bond strength test

A total of 35 extracted human maxillary incisors were used. Each tooth was placed in 1.25% NaOCl for 2 hours for surface disinfection. The middle and coronal thirds were prepared using ISO size 50, 70, 90 and 110 Gates Glidden drills (Produits Dentaires S.A., Vevey, Switzerland), with a low-speed handpiece to a depth of 5 mm. The apical third was prepared with ProFile instruments up to size 45, 0.04 taper. The irrigation was the same as that used for the sealing ability test.

The roots were divided randomly into two experimental groups of 15 roots each and one positive control group of five roots. All roots were obturated as described above for the sealing ability test. After complete setting, each root was sectioned perpendicularly to their long axis at 2 mm below the cemento-enamel junction into three or four 1-mm serial slices with a slow-speed saw (Isomet; Buehler) under water cooling. Each slice was marked to distinguish the coronal from the apical side. We obtained 45 slices for the Resilon/NRCS group, 46 for the Resilon/Epiphany group, and 15 slices for control group. Digitized images of the coronal and apical sides of each slice were captured at 10× magnification using a charge-coupled device camera attached to a stereomicroscope.

The slice thickness was measured by digital caliper. Each slice was subjected to compressive loading via a universal testing machine (WD-10A Instrone, Norwood, USA) equipped with a 1-mm diameter cylindrical plunger. The loading force was applied at a crosshead speed of 0.5 mm/min in an apical to coronal direction. Failure was manifested by extrusion of the intact cone of root filling from the root slice, and confirmed by the appearance of a sharp drop along the load/time curve recorded by the machine.

The circumferences of the coronal (Cc) and apical aspects (Ca) of each slice were measured from the digitized images using image analysis software (Image 4.01; Scion Corp., Frederick, MD, USA). The interfacial area of the root filling was calculated by the following formula: $A = 0.5(Cc + Ca) \times h$, where h is the root slice thickness. Interfacial bond strength was then obtained by dividing the load at failure recorded by the estimated interfacial area (A). Before and after the push-out test, five intact slices and five "empty" slices from each group were randomly selected for examination by environmental scanning electron microscopy (ESEM) (Philips XL-30 ESEM-FEG; Eindhoven, Netherlands). The specimens were fixed in 2.5% glutaraldehyde at 4°C, and dehydrated in ascending ethanol solutions (30% to absolute ethanol) and dried. After sputter-coating the specimens with gold, they were viewed by ESEM.

Cytotoxicity test (MTT assay)

Freshly mixed NRCS and Epiphany were lighted for 40 seconds to set and exposed to UV light for 1 hour in an aseptic environment. Four grams of sealer was added to 20 mL Dulbecco's Modified Eagle Medium (Gibco, Montana, USA) supplemented with 10% fetal bovine serum and incubated for 72 hours at 37°C in an atmosphere of 5% CO_2 and air. The elute was filtered by the Filter Unit (Corning Inc., NY, USA) with a diameter of 0.22 μm. To determine the cytotoxicity of NRCS, human periodontal ligament cells were cultured from healthy premolars for orthodontic purpose, with informed consent of two donors: a 15-year-old woman and a 16-year-old man. The study protocol were approved by the Ethics Committee, Guanghua College of Stomatology, Sun-yat Sen University. The middle thirds of the periodontal ligaments on root surfaces were minced and cultivated in Dulbecco's Modified Eagle's Medium with 20% fetal bovine serum. The cells were grown at 37°C in an atmosphere of 5% CO_2. Cells from passage four were placed into three wells of 96-well plates (Corning, NY, USA), each with 18 samples at 3.0×10^4 cells/mL. After 24 hours incubation, 200 μL of elute of each material was placed in a well of the microplates and incubated for 24 hours, 48 hours and 72 hours at 37°C in a 5% CO_2 atmosphere.

The tetrazolium reduction assay was accomplished as described by Denizot.[30] Cytotoxicity was calculated by the relative growth rates (RGR) and the cytotoxicity grade (CG) was presented as follows: non-cytotoxicity, RGR > 90%, CG = 0; slight cytotoxicity, RGR 60–90%, CG = 1; moderate cytotoxicity, RGR 30–59%, CG = 2; severe cytotoxicity, RGR ≤ 30%, CG = 3.

Statistical analysis

Assays were repeated at least three times to ensure reproducibility. For the physicochemical properties, we evaluated the values according the ISO specifications. The Kruskal–Wallis test and Bonferroni test were used to determine the difference between the Resilon/NRCS and Resilon/Epiphany groups. The data of push-out bond strength were analyzed using one-way analysis of variance and *post hoc* Tukey's test between the Resilon/NRCS and Resilon/Epiphany groups.

Results

Physicochemical properties

As illustrated in Table 1, the setting times, flow, solubility, dimensional change and radiopacity of NRCS conformed to the ISO standard for endodontic filling materials [ISO 6876:2001(E)]. However, the film thickness or NRCS was slightly higher than the ISO specifications. The setting time solubility and dimensional changes of Epiphany did not comply with the ISO requirements.

Sealing ability

The microleakage of the two sealers is summarized in Table 3. There was no difference between the Resilon/NRCS and Resilon/Epiphany groups before day 15. Significant differences were observed at days 20 and 30 ($p < 0.01$) and the teeth in the NRCS group had significantly lower microleakage than those in the Epiphany group. The increase in microleakage with time was slower in the Resilon/NRCS than in the Resilon/Epiphany group.

Bond strength

The mean push-out bond strength for the Resilon/NRCS ($n = 45$), Resilon/Epiphany ($n = 46$) and Resilon only ($n = 15$) groups was 2.0531 ± 1.0214 Mpa, 1.5062 ± 0.5923 Mpa, and 0.5061 ± 0.5914 Mpa, respectively (Table 4). ESEM results are shown in the Figure.

Table 3. Microleakage of glucose in the Resilon/NRCS and Resilon/Epiphany groups ($n = 8$ in each time point)*

Time (d)	Resilon/NRCS (mmol/L)	Resilon/Epiphany (mmol/L)
1	0.05 ± 0.00	0.06 ± 0.01
4	0.21 ± 0.01	0.24 ± 0.06
15	0.90 ± 0.20	0.99 ± 0.42
20	1.31 ± 0.47	2.56 ± 1.11
30	3.16 ± 0.67	4.25 ± 1.77

**Data presented as mean ± standard deviation. NRCS = new resin-based root canal sealer.*

Table 4. Bond strength values for displacement of the root canal filling system from root canal in bond strength test*

	n	Bond strength (MPa)
Resilon/Epiphany	46	1.5062 ± 0.5923
Resilon/NRCS	45	2.0531 ± 1.0214
Resilon	15	0.5061 ± 0.5914

**Data presented as n or mean ± standard deviation.*

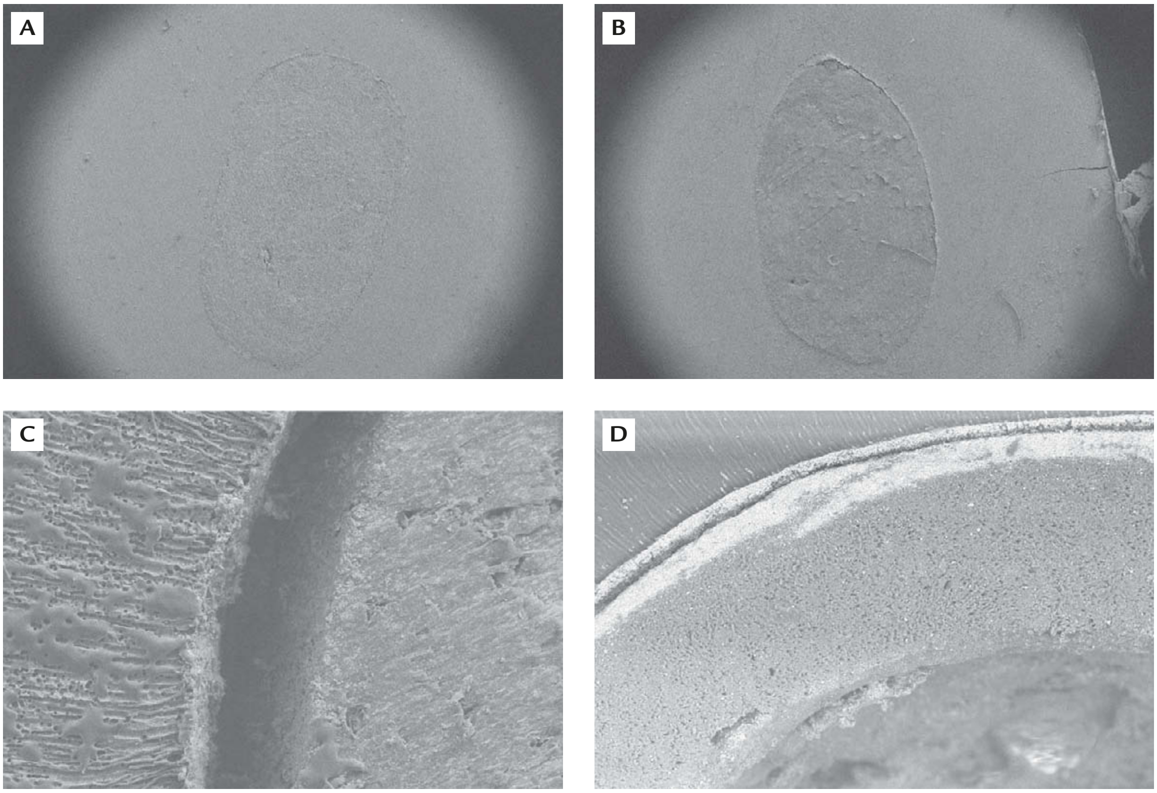

Figure. (A) Dentin/new resin-based root canal sealer (NRCS)/Resilon was uniform. (B) Gaps between the dentin-sealer interface in the Resilon/Epiphany sample. (C) Bond failure occurred along Epiphany and dentinal wall after the push-out test. (D) All bond fractures took place within the Resilon core material in the NRCS group after the push-out test.

Cytotoxicity

After the cells were incubated for 24 hours, 48 hours and 72 hours, there were significant differences in the RGR and the CG between NRCS and Epiphany at all times ($p < 0.05$, Table 5). The NRCS group showed an RGR of 91.24–102.10% at 24 hours and 48 hours, with a CG of 0, but at 72 hours, it showed slight cytotoxicity (RGR 70.01% and CG of 1). For the Epiphany group, it displayed RGR of 60.30% at 24 hours with a CG of 1, and it showed moderate cytotoxicity with an RGR of 36.62–48.23% and CG of 2 at 48 hours and 72 hours.

Table 5. Relative growth rate and cytotoxicity grade of new resin-based root canal sealer and Epiphany*

	24 hr		48 hr		72 hr	
	RGR	CG	RGR	CG	RGR	CG
NRCS ($n = 18$)	1.0210	0	0.9124	0	0.7001	1
Epiphany ($n = 18$)	0.6030	1	0.4823	2	0.3662	2

**RGR is the rate of optical density of the cell co-cultured to the sealer leaching liquor and the optical density of the normal cell. RGR = relative growth rate; CG = cytotoxicity grade; NRCS = new resin-based root canal sealer.*

Discussion

Techniques that are currently used in restorative dentistry involve adhesive bonding to dentin. The new generation of dentin bonding systems have achieved high bonding strength and reduced microleakage by micromechanical bonding or by forming a hybrid layer between the dentin and the resin.[38] Methacrylate monomers have been the most extensively used component of commercially available composite restorative materials. Resin-based filling materials are now accepted for use in root canals. Attempts are being made to improve the clinical performance of the resin-based sealers. The chief approaches include the reduction of polymerization shrinkage to improve adaptation of sealers to the dentinal walls, avoidance of interface gaps, and improvement of biocompatibility by reducing the elution of components. Chemical approaches to improving the efficacy of the sealers have been mainly focused on the development of new base monomers. We have synthesized a novel monomer (HPFDM)[35] by modifying bis-GMA so that the co-polymerization rate is higher. In the present study, a novel sealer (NRCS) with a molecular weight of 634 was developed using the new dimethacrylate-based monomer as one of its constituents, along with bis-GMA, TEGDMA, UDMA and fillers. Although incorporation of a new monomer into a resin-based restorative material is not particularly new,[39] the addition of this novel monomer to the resin-based root canal sealer might affect its properties, by decreasing shrinkage and improving its physicochemical properties.

The flow properties and film thickness represent the ability of a sealer to change form and completely fill the prepared root canal system. It is important for methacrylate-based sealers to set slowly and flow for a long time to compensate for the polymerization stress.[40] NRCS and Epiphany showed good flow properties *in vivo*. However, Tay[40] pointed out that applying an immediate coronal seal after obturation could affect the benefits of a slow, auto-curing process. Epiphany showed high dimensional change and solubility compared to NRCS. This was in conformity with the results obtained in previous studies.[9,41] Although the monomers in the polymerization process undergo cross-linking reactions that decrease the diffusion of water through the network structure, the chemical structure of the monomer remains the determining factor in water sorption.[42] NRCS was synthesized by mixing the new monomer HPFDM with bis-GMA, TEGDMA, UDMA and fillers. Bis-GMA was added because of the possible decrease in the conversion of the C–C double bond of the new monomer with higher molecular weight and volume. This gave increased solubility and excessive leaching of monomer, thus compromising the biocompatibility of the polymer. NRCS satisfied the requirements of the ISO standards for root canal sealing material, except for a slightly high value for linear expansion. This means that the addition of HPFDM appeared to improve the physical and chemical properties compared to those of Epiphany.

Because the glucose had a small molecular size and was nutrient for bacteria, the glucose fluid leakage test was more relevant to clinical outcomes than other models in the apical sealing ability.[37] Degradation of Resilon can occur in an alkaline medium.[20] Preliminary tests were accomplished to find a suitable concentration of NaN_3 that could both inhibit bacterial contamination and not affect Resilon. The Resilon/NRCS showed better sealing ability compared to that of Resilon/Epiphany. It is probable that some unreacted monomer and filler in Epiphany penetrated more into the solution than NRCS, which resulted in a space between the dentin and sealer. Another reason might be that NRCS exhibits less shrinkage during the curing process than does Epiphany, and the gap between the dentin and sealer is smaller with NRCS. This suggests that NRCS has good curing behavior and most probably lower residual bonds in the resin matrix compared to those with Resilon/Epiphany. However, additional studies are needed to determine the amounts of residual double bonds.

In this study, the push-out method was used to compare the bond strength of NRCS and Epiphany to the dentinal wall. The mean bond strength of the Resilon/NRCS was significantly higher than that of Resilon/Epiphany ($p < 0.05$). Studies by Ungor and Skidmore on bond strength using the push-out method have shown similar results with the Resilon/Epiphany combination.[8,43] ESEM allowed examination of fully hydrated specimens in their natural state with no prior preparation. This is important to rule out artifacts caused by vacuum desiccation during conventional SEM. ESEM inspection of the dentinal surface revealed that NRCS showed a better adaptation to the dentinal wall compared to Epiphany (Figure A and B). For Resilon/Epiphany, the bond failure appeared mainly at the dentin/Epiphany junction after the push-out test (Figure C). For Resilon/NRCS, fracture of the bonded specimen occurred within the Resilon core material, with dentin/NRCS/Resilon uniform, and the gap among the dentin/Epiphany/Resilon interface was easily recognized (Figure C and D). Inspection of the dentinal surface demonstrated that NRCS adhered to the surface more efficiently than did Epiphany.

The root-canal filling has long-term contact the periodontal tissue when it is used in endodontic therapy; therefore, the periodontal ligament cells were selected. The co-culture of periodontal ligament with extracts of NRCS and Epiphany, which were used in this study, has several benefits: (1) the extract can be easily prepared; (2) the extract can be completely dissolved in medium and thus have homogeneous contact with cells; (3) the extract can be autoclaved easily; and (4) some impurities can be removed by centrifugation or filtration. Current root-canal filling materials have some cytotoxicity,[44] which can influence the biocompatibility of materials and thus decrease the efficiency of root-canal therapy. The biocompatibility of resin-based filling materials is mainly affected by unpolymerized exudates such as the monomer and formaldehyde. Susini[45] has tested the toxicity of extracts from the Resilon/Epiphany root-canal filling system to mouse L-929 fibroblasts by the MTT assay. Their results showed that the Resilon/Epiphany root-canal filling materials had greatest toxicity at days 1 and 2. This temporary cytotoxicity is due to Epiphany; the results are similar to those of Key et al.[46] Key et al[46] tested cytotoxicity of several root-canal fillings by the trypan blue exclusion assay and have found that Epiphany has greater toxicity than Sealapex after 1 hour and 24 hours exposure. This toxicity might result from the high solubility of the non-cured layer of Epiphany. The results of the present study indicate that NRCS shows only slightly cytotoxicity, with CG values of 0 or 1, which indicates that NRCS has good biocompatibility. It is possible that less unreacted monomer is released into the elute, and less filler, such as zinc oxide, is dissolved into the extract.

NRCS is a new resin-based root-canal sealer made from the methacrylic monomer HPFDM. Its improved physicochemical properties enhance sealing of the root canal and it has good biocompatibility, therefore, NRCS could be valuable for future clinical applications.

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