Article

Impact of Water Solubility on Chemical Composition and Surface Structure of Two Generations of Bioceramic Root Canal Sealers

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Abstract: Aimed to evaluate the effect of water solubility on chemical properties and surface structure of bioceramic-based (BC-HiFlow and BC-EndoSequence) compared with resin-based (Adseal) root canal sealers. Fresh mix was inserted into polyethylene mold ($n = 10$) and subjected to Vicat needle to evaluate the setting time. The set discs were analyzed by Fourier transform infrared (FTIR) spectroscopy then immersed in deionized water for 1, 7, 14 and 28 days. The solubility%, pH changes, released calcium (Ca^{2+}), phosphate (PO_4^{3-}) and silicon (Si^{4+}) ions were evaluated after each immersion period. The discs were analyzed by scanning electron microscopy/Energy dispersed X-ray (SEM/EDX) before and after solubility test. Although FTIR detected similar composition of both bioceramic-sealers, BC-EndoSequence determined the prolonged setting times. At the end of solubility test, both bioceramic-sealers exhibited significant greater solubility ($>3\%$), alkaline pH (>11) at $p < 0.001$. Adseal displayed the significant greatest Ca^{2+} and PO_4^{3-} released, while BC-HiFlow displayed the significant greatest Si^{4+} release ($p < 0.001$). SEM revealed voids and pores on the surface of all tested sealers with the greatest value on Adseal surface. In conclusion, although both bioceramic-sealers had high solubility, BC-HiFlow complied the ISO standard regarding setting time and least surface micropores better than that of BC-EndoSequence.

Keywords: BC-sealers; setting time; solubility; pH; chemical properties

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1. Introduction

Root canal sealers are used during endodontic obturation to achieve a hermetic seal throughout the root canal system including minor discrepancies and canal irregularities [1]. Therefore, ideal endodontic sealers should prevent leakage, minimize the risk of bacterial invasion from the periapical tissues, and resolve the periapical lesion [1,2]. In addition, root canal sealer should be biocompatible, dimensionally stable, set slowly to ensure sufficient working time, provide an excellent seal when set and provide an adequate adhesion to the canal walls. It also should be insoluble to tissue fluids [3].

Solubility is defined as material mass loss during certain immersion time in aqueous environment [4]. It is an undesirable physical property for root canal sealers. It can induce sealer degradation; creating gaps at sealer/dentin or sealer/gutta-percha interface which compromise the apical seal of the obturation system [5] and enhance bacterial leakage [5]. Solubility behavior is influenced by the setting time and pH of the sealer [6]. In fact, the degraded byproducts and the released particles induce changes in pH of the media and in the sealer surface morphology which may affect the biological property of the sealer [7,8].

Nowadays, various endodontic sealers are available in the market, including sealers based on glass ionomer, calcium hydroxide, resin, zinc oxide eugenol, silicone, and bioceramic (BC)-based root canal sealers. BC-EndoSequence sealer (Brasseler USA, Savannah, GA, USA) was the first bioceramic-based root canal sealer introduced in 2009 [9]. It is a

premixed paste sealer composed of calcium, phosphate, and silicate. It is categorized as hydraulic calcium silicate cement as it sets in the presence of a moist environment [10]. It has the ability to form calcium hydroxide as a byproduct of hydration reaction [11], and is responsible for its alkaline pH inducing antibacterial properties [12]. It is a biocompatible, bioactive sealer and enhances the osteogenic potential [10,13,14]. Its nanoparticle size allows an excellent flow to the dentin wall and improve the sealing ability [9]. In addition, it is available in injectable syringe enhances easy method of delivery and application. Previous studies reported high solubility of EndoSequence sealer [4,6,8] that exceeds the acceptable level of ISO [15].

Recently, a new BC- HiFlow sealer (Brasseler USA, Savannah, GA, USA) launched in the market, and it is compatible with heat generated during warm vertical compaction technique. Few studies were recently published on the effect of heat on its physico-chemical properties [16–18]. It claimed to be less soluble due to its fast setting [16]. More information about chemical changes of BC- HiFlow sealer when immersed in aqueous media are required. The purpose of this study was to evaluate the effect of water solubility on the chemical properties (setting time, pH and released calcium (Ca^{2+}), phosphate (PO_4^{3-}) and silicon (Si^{4+}) ions) and surface structure of recent generation BC-HiFlow versus old generation BC-EndoSequence bioceramic-based root canal sealers. The null hypothesis was that there was no significant difference between both sealers.

2. Materials and Method

The materials used in this study were pre-injectable syringe of EndoSequence BC- and HiFlow BC- Bioceramic-based root canal sealers (Brasseler USA, Savannah, GA, USA). Dual syringe paste/paste injectable resin-based root canal sealer—Adseal (META Biomed Co., Chungbuk, Korea) was used as a control.

2.1. Setting Time

The initial and final setting times of all sealers were measured using a Vicat needle apparatus (Jin-Ching-Her, Taiwan). Sealers were placed inside polyethylene molds of 10 mm diameter and 3 mm height ($n = 10$). The premixed BC-sealers were injected directly into the mold, while the Adseal was mixed according to the manufacture instructions and inserted inside the mold. The surface of freshly mixed sealer was subjected to Vicat needle of 10 mm diameter, 50 mm length and 100 gm weight, at room temperature ($23 \pm 1^\circ\text{C}$). The initial and final setting times were recorded until no indentation of the needle [19].

2.2. Solubility Test

Polyethylene molds of 10 mm diameter and 2 mm height were filled with freshly mixed sealers ($n = 10$). The procedure was performed according to specification number 57 of the American Dental Association (ADA) for root canal filling [15]. The molds were held between two glass slabs and stored in an incubator at 37°C and 100% humidity until sealer set. After complete setting, the original dry weight of each sample (W_0) was measured using an electric balance (Model #ZSA210, Scientech Inc., Boulder, CO, USA). Each sample was then stored in a vial containing 10 mL deionized water at 37°C . At an interval incubation periods (1, 7, 14 and 28 days), the samples were removed from the vial, dried with filter paper overnight and reweighed (W_t). The percent of material loss (solubility %) was calculated according to the following equation [20]:

$$\text{The solubility \%} = \frac{W_0 - W_t}{W_0} \times 100$$

2.3. pH Changes

After calibration of the pH meter (JENWAY-3510 pH meter, Bibby Scientific Ltd., Stone, Staffs, UK) with standard solutions at pH 4.0 and 7.0 at a constant temperature

(25 °C) [12], the pH of each solution at each immersion interval period (1, 7, 14, and 28 days) was evaluated.

2.4. Releasing Elements

The releasing Ca^{2+} , PO_4^{3-} and Si^{4+} ions were determined in the immersing solution of each solubility interval period (1, 7, 14 and 28 days). For Ca^{2+} release, the solution was analyzed by EDTA titration method [21]. For PO_4^{3-} release, the colorimetric method with spectrophotometer (JENWAY- 6705 UV/Vis Spectrophotometer, Stone, Staffs, UK) [22] was used. For Si^{4+} release, Inductively Couple Plasma Optical Emission Spectrometer (ICP-OES, Agilent 5100, Santa Clara, CA, USA) was used.

2.5. Fourier Transform Infra-Red (FTIR) Analysis

The set sealers were subjected to FTIR spectroscopy (Vertex 80v, Bruker, Germany) to analyze their chemical composition. The spectra were scanned at $4000\text{--}400\text{ cm}^{-1}$ and 1 cm^{-1} resolution. To verify the data, each spectrum was scanned three times.

2.6. Scanning Electron Microscope (SEM) and Energy Dispersive X-ray (EDX) Analysis

The surface morphology and composition of each sealer were determined by examining the discs of the set sealer using SEM/EDX (Octane pro, Model 7.2/15252, EDAX. Ametek Material and analysis division, Mahwah, NJ, USA). The changes in surface morphology and composition, as a result of solubility test, were also evaluated by examining the samples at the end of solubility test (28 days).

2.7. Statistical Analysis

To compare between the investigate sealers, the data of setting time, solubility %, pH changes, releasing elements and EDX analysis was statistically analysed by One-Way ANOVA and Post-hoc Tukey HSD tests using SPSS software (Version 16.0; SPSS, Inc., Chicago, IL, USA) at significance level of 5%.

3. Results

3.1. Setting Time

Prolonged initial setting time was significantly recorded by BC-EndoSequence at $p < 0.001$, while shortest initial setting time was recorded by Adseal with no significant difference with BC-HiFlow at $p = 0.194$. In regard to the final setting time, Adseal recorded the significant fastest setting time followed by BC-HiFlow ($p < 0.001$). BC-EndoSequence showed 0.5 mm indentation after 123 ± 10.3 hrs that persisted for 3 months (Table 1).

Table 1. Represents the means $\pm$ Standard deviation values of initial and final setting times of the three tested sealers.

		BC-HiFlow	BC-EndoSequence	Adseal
Setting Time	Initial (hrs)	$4.12 \pm 0.12 \neq$	$44.2 \pm 6.3 *$	$1.22 \pm 0.19 \neq$
	Final (hrs)	37 ± 5.46	$<123 \pm 10.3 *$	$4.7 \pm 0.44 \dagger$

(*) is the significant greatest value while (†) is the significant lowest value at $p < 0.001$. ($\neq$) indicates no significant difference between the sealers of the same symbol at $p > 0.05$.

3.2. Solubility Test

The BC-Hiflow showed the significant lowest solubility % in all experimental immersion times, while the BC-EndoSequence showed the highest values that increases with time ($p < 0.001$). On the contrary, Adseal displayed gradual gain weight that declined after 21 days at $p < 0.001$ (Figure 1A).

Figure 1. Represents the mean ± standard deviation values of (A) solubility %, (B) pH changes, (C) calcium, (D) phosphate and (E) silicon ions released of the investigated sealers among the experiment immersion times. (*) is the significant greatest value while (†) is the significant lowest value at $p < 0.001$. (≠) indicates no significant difference between the sealers of the same symbol at $p > 0.05$.

3.3. pH Analysis

The result of the pH analysis for all tested sealers in all time periods was shown in (Figure 1B). Both BC-sealers determined significant high alkaline pH (range between 10.6 and 12.6), as compared to Adseal (≈ 8.5), at $p < 0.001$. The pH of BC-HiFlow increased from day 1 to day 7 then gradually decreased up to day 28. BC-EndoSequence showed continuous increasing in the pH from day 1 up to day 28. Adseal exhibited the significant weak alkaline pH throughout the experimental periods.

3.4. Releasing Elements

3.4.1. Calcium Ion Release

Adseal exhibited the significant high amount of Ca^{2+} release compared to the BC-sealers ($p < 0.001$). Meanwhile, BC-EndoSequence recorded the significant lowest values of Ca^{2+} release, with no significant difference versus BC-HiFlow within the first 7 days ($p > 0.05$), while a significant difference was detected after the 7th day ($p < 0.001$) (Figure 1C).

3.4.2. Phosphate Ion Release

BC-HiFlow and BC-EndoSequence sealers showed gradual reduction in the amount of PO_4^{3-} release from day 1 up to day 28, whereas a gradual increase was recorded by the Adseal (Figure 1D). The significant greatest value was recorded by BC-EndoSequence at day 1 and 7, whereas Adseal showed the significant greatest values at day 14 and 28 at $p < 0.001$. The significant lowest value was obtained by BC-HiFlow among all experimental immersion times at $p < 0.001$.

3.4.3. Silicon Ion Release

In all the three sealers, the amount of Si^{4+} release was gradually decreased by time. The significant amount of releasing Si^{4+} , from day 1 to day 28, was obtained by BC-HiFlow followed by BC-EndoSequence at $p < 0.001$, while the significant lowest values were obtained by Adseal at $p < 0.001$ (Figure 1E).

3.5. FTIR Spectroscopy

The spectra of BC-HiFlow (Figure 2A,B) and BC-EndoSequence (Figure 2C,D) revealed similar composition of both BC-sealers with variable band intensity. They detected bands of calcium hydroxide $\approx 3640 \text{ cm}^{-1}$ [11,23–26], OH of absorbent water associated with hydration reaction of sealer $\approx 3400 \text{ cm}^{-1}$ [24,25,27], C-H and OH- of organic filler (polyethylene glycol) at 2980–2800 and $\approx 1635 \text{ cm}^{-1}$ [23,25,27], carbonate (CO_3^{2-}) at 1420 and 874 cm^{-1} [23,24,28], phosphate bands of $\nu_3\text{PO}_4^{3-} \approx 1066$ [27,29,30] and at 974 cm^{-1} [28], $\nu_4\text{PO}_4^{3-}$ at 665 cm^{-1} [27,29,30], SiO_4^{4-} of calcium silicate incorporated with $\nu_3\text{PO}_4^{3-}$, calcite at 974 and 874 cm^{-1} , respectively [28,31], $\nu_4\text{SiO}_4 \approx 750 \text{ cm}^{-1}$, SiO_4^{4-} bending mode of tricalcium silicate (C_3S) $\approx 570 \text{ cm}^{-1}$ and dicalcium silicate (C_2S) at 502 and 494 cm^{-1} [25], Si-O- stretching mode of calcium silicate hydrate (CSH) at 450 cm^{-1} [25,28,32].

The spectra of Adseal showed different composition. It detected intense bands of C=O of organic filler $\approx 1635 \text{ cm}^{-1}$ [23,25] carbonate band (CO_3^{2-}) at 1458 cm^{-1} , silicate group (Si-O) at 1113 cm^{-1} [25], phosphate bands (ν_3 and $\nu_4\text{PO}$) at 1035 and 680–560 cm^{-1} [30], respectively, and polymerized silicon (SiO) at 420 cm^{-1} [25], (Figure 2E,F).

3.6. SEM/EDX

The surfaces of both BC-HiFlow (Figure 3A) and BC-EndoSequence (Figure 3B) had a nearly similar structure before immersion in water. They showed a homogenous structure of granular mass. The surface of Adseal showed uniform, homogenous structure of irregular small particles (Figure 3C). The bright particles of radio-opacifiers (ZrO_2) were distributed over the surface of all sealers. The intra-granular voids and pores were detected in all sealers where more prominent in Adseal surface.

Figure 2. FTIR spectra at 4000–2000 and 2000–400 cm^{-1} of BC-HiFlow (A,B), BC-EndoSequence (C,D) and Adseal (E,F) respectively showed their composition.

The elemental analysis determined that all tested sealers contain carbon (C), oxygen (O), silicon (Si), phosphorous (P) and calcium (Ca) peaks with different wt.% (Figure 3D–F). Before immersing (day 0), the surface of the set sealers contained high C peak and low O and Ca peaks ($p < 0.001$). The significant greater C, P peaks were recorded by Adseal, while the significant high Ca peak was recorded by BC-EndoSequence ($p < 0.001$). The Si peak was significantly high in BC-HiFlow and low in BC-EndoSequence. The P peak was significantly high in Adseal at $p < 0.001$, while low in both BC-sealers with no significant difference between them ($p = 0.751$). As a radio-opacifier, BC-HiFlow contains significant higher zirconium (Zr) peak than BC-EndoSequence, while Adseal contained bismuth (Bi) peak ($p < 0.001$).

The development of porous hierarchical structure (calcium carbonate) was shown on BC-HiFlow surface. The spheroidal and rod-like (aragonite) crystalline structure was detected over the BC-EndoSequence surface with the development of cubic shaped crystals (calcium hydroxide) (Figure 3K), while rectangular shaped-crystals (calcium phosphate) were shown on the Adseal surface after water immersion (Figure 3G–I). The voids and pores were predominant on the surfaces of all tested sealers with different distribution at the end of water immersion time (at day 28), (Figure 3J–L). The greatest value was observed on the Adseal surface (Figure 3L).

After immersion in water (day 28), the elemental analysis (EDX) of BC-HiFlow (Figure 3M) showed decrease in C, Si and Zr, while significantly increase in P and Ca peaks ($p < 0.001$). The spectra of BC-EndoSequence (Figure 3N) showed marked decrease in C, but significant increase in Si, P, Zr and Ca. The spectra of Adseal (Figure 3O) showed significant increase in Si and P peak, while decrease in Bi ($p < 0.001$), and no significant changes in Ca peak ($p > 0.05$).

Figure 3. SEM/EDX analysis of the three sealers before (A–F) and after 28 days (G–O) immersion in water. (*) is the significant greatest value while (†) is the significant lowest value at $p < 0.001$. (≠) indicates there is no significant difference between sealers with the same symbol at $p > 0.05$.

4. Discussion

Recently, a new formula of bioceramic (BC-HiFlow) sealer was manufactured to resist the high temperature subjected during warm vertical compaction technique. It is important for clinician to understand the behaviour and performance of root canal sealer under clinical situation. The sealer solubility has an impact on the longevity of endodontic treatment.

It could induce sealer degradation; compromising the apical seal, and enhance bacterial leakage [5].

The current study investigated the impact of water solubility on the chemical properties of the first (BC-EndoSequence) versus the recent (BC-HiFlow) generation of bioceramic sealers. Although both sealers have nearly similar composition [10], their chemical behaviour and surface morphology (particularly after water immersion) have been currently reported to be different. Hence, the null hypothesis was rejected.

Both BC-sealers displayed solubility% with the significant greater values obtained by BC-EndoSequence greater than acceptable level of ISO [18], within 21 days (Figure 1A). The high solubility was previously reported by both BC-sealers in other studies which was either in accordance with acceptable limit (3% mass fraction) [33] or more exaggerated than the recommended with ISO [4,6,8,12,34]. The present result recorded the solubility values within an acceptable limit within the first 14 or 21 days for BC-Endosequence or BC-HiFlow, respectively (Figure 1A). This discrepancy of current versus previous findings may be due to the solubility test was performed after one month until maximum material setting. Conversely, Adseal underwent to gain weight that gradually increased then declined after 21 days. This finding is in accordance with previous study [35]. Adseal resin-based sealer was used as a control sealer due to its availability. It is also composed of calcium phosphate comparable to bioceramic based sealers. Moreover, several studies reported good solubility property similar to other resin based sealer such as AH-plus [36].

The sealer solubility seems to be influenced by multiple factors, setting time and composition. In turn, it may induce greater release of elements and pH changes. According to Grossmann, slow initial setting without further prolonged final setting and alkaline pH are essential properties required for an ideal root canal sealer [3,37]. BC-EndoSequence displayed the significant prolonged initial setting time and failed to complete set until 3 months (Table 1). However, BC-HiFlow recorded 4.12 and 37 hrs for initial and final setting time, respectively. The prolonged setting time of BC-Endosequence was previously reported, where it was exceeding one month [38]. It was suggested that the BC-EndoSequence, as a hydraulic sealer, did not set in >95% humid incubator [35] and it set in a moist environment similar to root canal system that contains fluid inside dentinal tubules [4]. There were few studies related to BC-HiFlow. The current finding was nearly similar to two studies which recorded 4–6 and 31–33 hrs for initial and final setting, respectively [16,39]. In addition, they showed no significant difference between setting time of BC-EndoSequence and BC-HiFlow after heat application [16,39]. Thus, heat seems to accelerate the hydration reaction [18,39].

Reciprocal reaction was noted between solubility, released by-products, and pH. Sealer with high alkaline pH and Ca^{2+} release consider as favourable root canal sealers to enhance antibacterial and osteogenic activity [7,13,33,40–42]. Both BC-sealers exhibit high alkaline pH range between 10.5 and 12, with the maximum value at 21 days for BC-HiFlow, while maintained continuous increased up to 28 days for BC-EndoSequence (Figure 1B). Similar pH values of BC-EndoSequence [18,33,43] and BC-HiFlow sealers [39] were previously reported. However, one study reported lower value for BC-HiFlow that did not exceed 8.5 among 28 days [16]. Their alkalinity was increased consequent to increase the solubility and increasing the releasing Ca^{2+} among the successive experimental periods.

The pH value seems to be associated with the setting time. Although both BC-sealers display high alkaline pH among all the experimental periods, gradual decline of this value by BC-HiFlow after 7 days was noted and may be attributed to its fast setting within this time. However, BC-EndoSequence represents further increase in pH up to 28 days that may be attributed to its prolonged final setting time which extended over 3 months.

On the other hand, Silva et al. 2021 reported that high alkaline environment reduced the solubility of the BC-EndoSequence sealer [6]. As a result of the hydration reaction of the calcium silicate-based sealers, calcium hydroxide was formed and dissociated into hydroxyl ions (OH^-). With high alkaline medium, supersaturation of OH^- may reduce further ion dissociation and thus reduce sealer solubility [6]. However, Adseal displayed weak alkaline pH (≈ 8.5) among all experimental periods. It was suggested that the low alkalinity was

favourable for epoxy resin sealer as it eliminates the corrosion and degradation of sealer particles and in turn reduce its solubility [6].

Assuming the similar composition of both BC-sealers, FTIR analysis revealed similar composition of both sealers with different band intensity indicating the variable concentration of their constituents, which related to their different chemical properties. The high solubility may be attributed to the $\text{Ca}(\text{OH})_2$ content [38] that was detected with high band intensity in FTIR spectra of BC-HiFlow (Figure 2A). Furthermore, the high intensity of CH- of organic filler indicates an increase in the setting accelerating agent while the decrease of OH- (at 3400 and $\approx 1650\text{ cm}^{-1}$) bands indicates the consumption of water during hydration reaction. The high intense $\nu_4\text{SiO}_4 \approx 750$ and 450 cm^{-1} indicates the formation of polymerized calcium silicate hydrate (CSH) [25,28,32]. These findings explained the faster setting of BC-HiFlow compared with spectra of BC-Endosequence (Figure 2B). However, the spectra of BC-Endosequence detected high intensity of $\nu_4\text{SiO}_4$ of C_2S at 874 cm^{-1} indicating greater content of unreacted particles [25], which may be related to its prolonged setting time and possibility of higher solubility.

On the contrast, Adseal contained insoluble resin matrix that formed during polymerization reaction of amine group of epoxide groups having covalent cross-linked bond that resist the solubility. Furthermore, gain weight was initially obtained by Adseal that may be attributed to initial water sorption of resinous matrix during polymerization expansion [5,35]. However, Adseal exhibited delay weight loss after 21 days and that may be attributed to degradation of unreacted particles [44].

The solubility is considered as prerequisite for leaching particles. Since calcium silicate is the main constituent in BC-sealer, and calcium phosphate in Adseal, the leachable Si^{4+} and PO_4^{3-} are respectively critical for these sealers. It confirmed by the current study where significant greater released of Si^{4+} was detected in BC-HiFlow followed by BC-EndoSequence. BC-HiFlow contained greater wt.% of Si than BC-EndoSequence (Figure 3D,E). Meanwhile, Adseal released greater PO_4^{3-} that attributed to its greater P wt.% (Figure 3F). These leachable particles are correlated to the formation of micropores over the sealer surface at the end of the solubility test. Although the greater Si^{4+} released by BC-sealers, the micropores were showed with lower values. It may be attributed to the crystalline phases detected on their surface when immersed in water (Figure 3G,H).

5. Conclusions

Upon the circumstances of this study, both BC-sealers presented greater solubility with significant greater values obtained by BC-EndoSequence that may be correlated with its prolonged setting time. Furthermore, BC-EndoSequence did not compete with the ISO standard related to solubility% that exceeds the acceptable limit within 21 days. It did not completely set at 100% humid incubator in up to 3 months. Yet, BC-HiFlow presented solubility% nearly within acceptable ISO limit. In addition, surface crystallization of BC-sealers after water immersion seems to resist surface solubility.

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