

# Apical Sealing Ability of Calcite-Synthesized Hydroxyapatite as a Filler of Epoxy Resin-Based Root Canal Sealer

## Abstract

**Background:** The success of root canal treatment is influenced by hermetic root canal obturation. This study was conducted to analyze the apical sealing ability after the addition of calcite-synthesized hydroxyapatite (HA) as an epoxy resin sealer filler. **Methods:** Calcite-synthesized HA powder was prepared using the microwave hydrothermal process. HA resin sealer powder and epoxy resin paste (3:1) were mixed, and concentrations of 10%, 20%, 30%, 40%, and 50% were prepared. A sample of thirty maxillary incisors were prepared in the root canal and then, the crown was cut to leave 13 mm of the root and a working length of 12 mm. The root canal was prepared using the crown-down technique and irrigated using 2.5% sodium hypochlorite and 17% ethylenediaminetetraacetic acid alternately. The samples were divided into six groups, with each group consisting of five roots. Group I was obturated with gutta percha using an epoxy resin sealer without HA (HA-0%) as a control group. In each of the Groups II, III, IV, V, and VI, 10% HA resin sealer, 20% HA, 30% HA, 40% HA, and 50% HA were used. All the samples were incubated in a 10-ml simulated body fluid solution at 37°C for 4 weeks. Apical closure density measurement was done using a scanning electron microscope, and the results were analyzed using the Kruskal–Wallis and Mann–Whitney U-tests. **Results:** A significant increase in the apical sealing ability was observed in the HA-20% sealer group and the HA-30% and HA-40% groups compared to that in the control group. However, the HA-50% sealer group showed a decrease in the apical sealing ability, whereas the HA-10% sealer group showed no difference. The HA-30% had the highest sealing ability than other concentrations. **Conclusion:** The addition of calcite-synthesized HA as a filler at concentrations of 20%, 30%, and 40% increased the apical sealing ability of the epoxy resin sealer.

**Keywords:** Apical sealing ability, calcite-synthesized hydroxyapatite, root canal sealer

## Introduction

The aim of root canal treatment is to eliminate infection in the root canal and close the entire root canal system to prevent the entry of tissue fluids and microorganisms into the root canal.<sup>[1,2]</sup> The goal of obturation of the root canal is hermetic closure along the root canal system, especially in the apical region.<sup>[2]</sup> The primary parameter of an obturation material is the apical sealing ability to prevent apical leakage. If apical leakage occurs, it indicates the failure of the root canal treatment. Apical leakage allows the entry of periapical tissue fluids, microorganisms, and antigens.<sup>[3,4]</sup>

Gutta percha is a root canal obturation material, and it must be added to the sealer to enable adherence to the canal wall. The sealer material is used to fill the gaps between the gutta percha and the root canal

wall. The root canal obturation material is in direct contact with the periapical tissue whose conditions are similar to those of the accessory canal or the raminal tubule ramification of the periapical tissue. The area that is in maximum and direct contact with the periapical tissue is the apical end of the obturation material located in the apical foramen. An ideal obturation material is hermetic and is not affected by the moisture in the periapical tissue.<sup>[3,4]</sup>

Hydroxyapatite (HA) has been used as a filler for several dental restoration materials such as composite resins and adhesive systems.<sup>[5,6]</sup> Synthesis of HA calcite is done from local mining in Indonesia. Previous studies have mentioned that HA calcite synthesis is not toxic.<sup>[7,8]</sup> Another study had analyzed the physical properties by the addition of up to 50% synthesized HA calcite as a filler in the epoxy resin root canal sealer and reported no effect on the contact angle and the film thickness, but the

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hardness of the sealer could be increased by the addition of a maximum of 30% HA calcite synthesis.<sup>[9]</sup> This study was conducted to analyze the apical sealing ability after the addition of calcite-synthesized HA as an epoxy resin sealer filler.

## Methods

This was an experimental, laboratory-based study, and the procedure was approved by the Ethics Committee of the Faculty of Dentistry, Universitas Gadjah Mada (UGM), in accordance with the Ethical clearance No. 290/KKEP/FKG-UGM/EC/2012. HA synthetic calcite<sup>[9]</sup> powder was prepared using the microwave hydrothermal process at the Bioceramics Laboratory, Department of Mechanical and Industrial Engineering, Faculty of Engineering, UGM. The synthesis was carried out by reacting natural calcite powder (PT. Omya, Mojokerto, East Java, Indonesia) with a solution of diammonium hydrogen phosphate  $(\text{NH}_4)_2\text{HPO}_4$ . The material in this study complied with ISO6876/2012 and ADA/ANSI 57 specifications.

Calcite-synthesized HA sealer powder was prepared from AH-26 silver-free epoxy resin (Dentsply, Germany) epoxy resin powder using HA calcite synthesis powder. HA resin sealer powder was stirred with an epoxy resin paste (3:1) on a mixing pad for 30 Seconds using a metal spatula. The mixing of the resin sealer powder with the calcite synthesis HA powder was done at the Integrated Research Laboratory, Faculty of Dentistry, UGM. Calcite-synthesized HA sealer powder was prepared at various concentrations using HA as the filler, that is, 10%, 20%, 30%, 40%, and 50% (in weight).

A sample of thirty maxillary incisors were prepared at the root canal, and the crown was cut using a diamond disc bur to leave 13 mm of the root. K-file number 15 was inserted up to the apical foramen section with a working length of 12 mm. The root canals were prepared by the crown-down technique using ProTaper in clockwise reaming movements. The ProTaper consists of shaping files (S1 and S2) and finishing files (F1, F2, and F3). The preparation was started using S1 file and then S2 file with a working length of 8 mm, followed by S1, S2, F1, and F2 files and completed using F3 file until a working length of 12 mm was reached. The completion was recapitulated using the same K-file numbers, that is, 20, 25, and 30. Each root canal file was irrigated with a maximum of 2 ml of 2.5% sodium hypochlorite irrigation solution using a Max-I-Probe irrigation needle (DENTSPLY, Maillefer, North America) and 17% ethylenediaminetetraacetic acid (EDTA) alternately. To equalize the diameter of the apical foramen, all the root canals were penetrated using the K-file number 25 1 mm out of the apical foramen.<sup>[10]</sup> Subsequently, the root canals were flooded with sodium hypochlorite for 5 min and 17% EDTA for 1 min and then irrigated again using saline and dried using 5 paper points.<sup>[11]</sup>

The samples were divided into six groups, with each group consisting of five roots. Group I (control) was obturated

with gutta percha using epoxy resin sealer without HA (HA-0%), and Groups II to VI were obturated using 10% HA resin sealer, 20% HA, 30% HA, 40% HA, and 50% HA, respectively. The sealer was inserted into the root canal using a Lentulo until the entire wall was coated with the sealer. Next, the gutta percha was inserted into the root canal according to its working length. The coronal part was closed using glass ionomer cement. Radiographs of the teeth were taken to ensure hermetic root canal obturation.

A simulated body fluid (SBF) solution was prepared in Unit II LPPT Laboratory and the Pharmacy Analysis Laboratory, Department of Chemistry, Faculty of Pharmacy, UGM.<sup>[12]</sup> A total of thirty plastic bottles were used, each containing cotton soaked in 10 ml of the SBF solution. Each root was inserted into the bottle according to the group with the root coronal position at the top. The samples were incubated at 37°C for 4 weeks.

The apical sealing ability was evaluated using a scanning electron microscope (SEM) at  $\times 100$  magnification. SEM photographs were taken to evaluate the apical sealing ability using the AutoCAD program by determining the root canal wall boundary and the root canal obturation boundary; then, the root canal cross-sectional (area I) and cross-sectional area of obturation material (area II) can be obtained between area II and area I in percentage.

Apical closure density ratio =

$$\frac{\text{Cross-sectional area of obturation material}}{\text{Root canal cross-sectional area}} \times 100\%$$

Data were analyzed using IBM SPSS Statistics version 22 for Windows (IBM SPSS Inc., IBM Software Group, Chicago, USA), and the significance level was set at  $P < 0.05$ . Normality and homogeneity of variance were calculated using the Shapiro–Wilk and the Levene’s tests, respectively. Statistical calculations were performed using the Kruskal–Wallis and Mann–Whitney U-tests.

## Results

The measurement of the apical closure density of the epoxy resin sealer was done using the AutoCAD program under the SEM. The results revealed a gap between the obturation material and the root canal wall [Figure 1] at various HA concentrations of calcite synthesis in all groups. The gap was the source of the apical leakage that could reduce the apical sealing ability.

The mean and standard deviation of the percentage of apical sealing ability using the HA sealer at various concentrations are depicted in Figure 2. The density of the HA sealer at various concentrations ranged from 82.0463%  $\pm 4.9708\%$  to 93.2050%  $\pm 2.1117\%$ .

The results of the normality test conducted using the Shapiro–Wilk test revealed normal data ( $P > 0.05$ ) and nonhomogeneous variance (Levene’s test showed  $P < 0.05$ ).

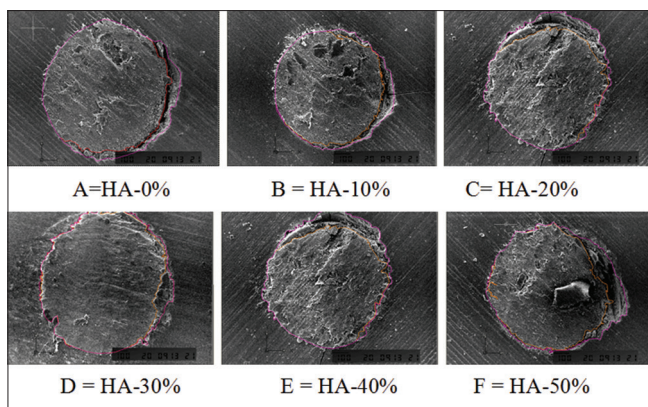


Figure 1: Apical sealing ability in root canal obturation using hydroxyapatite sealer at various concentrations

The Kruskal–Wallis test resulted in a Chi-square value of 20.277 with  $P = 0.001$ , which indicated a statistically significant difference in the apical sealing ability in all the treatment groups ( $P < 0.05$ ). The Mann–Whitney U-test [Table 1] showed that there were nine pairs of groups showing significant differences, whereas six pairs of groups showed no differences in the apical sealing ability.

Furthermore, the apical sealing ability increased in the HA-20% sealer group and the HA-30% and HA-40% groups compared to that in the control group. However, the apical sealing ability decreased in the HA-50% sealer group, whereas the HA-10% sealer group showed no difference. The HA-30% had the highest sealing ability than other concentrations, although it did not have a significant difference compared to HA-20%.

### Discussion

The results of this study [Figure 1] revealed apical leakage in all the experimental samples. This finding is consistent with the opinion of Kim *et al.*,<sup>[13]</sup> who stated that till date, there is no sealer material that can be firmly and perfectly attached to the gutta percha and the root canal wall. Furthermore, the resin sealer that has the best sealing ability compared to that of other sealers such as zinc oxide eugenol, calcium hydroxide, and glass ionomer cement also does not result in a perfect apical sealing ability. In addition, the phenomenon of polymerization contraction has been believed to result in a gap between the resin and the adherent surface. Apical leakage can also occur because the root canal of the tooth apical area is anatomically the most difficult part to produce density compared with the coronal part. This is due to the increasingly narrow anatomical shape in the apical region, the decrease in the density of the dentinal tubules, and the more sclerotic dentin that covers the dentinal tubules and comes into direct contact with the moist periodontal tissue due to the presence of the apical foramen.<sup>[13]</sup>

As shown in Figure 2, the apical sealing ability achieved using HA-20%, HA-30%, and HA-40% sealers was higher

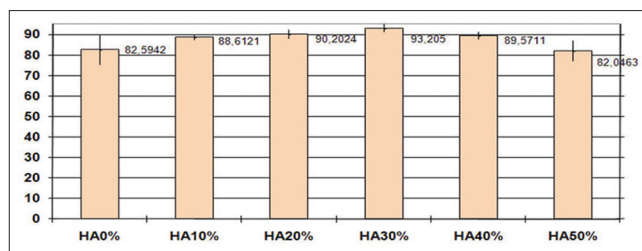


Figure 2: The mean and standard deviation of the percentage of apical closure density using hydroxyapatite sealer at various concentrations (%)

Table 1: Results of the Mann-Whitney U-test of apical sealing ability using hydroxyapatite sealer at various concentrations

Group	Mann-Whitney	P
HA-0%-HA-10%	5.000	0.117
HA-0%-HA-20%	2.000	0.028*
HA-0%-HA-30%	0.000	0.009*
HA-0%-HA-40%	2.000	0.028*
HA-0%-HA-50%	11.000	0.754
HA-10%-HA-20%	6.000	0.175
HA-10%-HA-30%	1.000	0.016
HA-10%-HA-40%	7.000	0.251
HA-10%-HA-50%	1.000	0.016*
HA-20%-HA-30%	4.000	0.076
HA-20%-HA-40%	11.000	0.754
HA-20%-HA-50%	0.000	0.009*
HA-30%-HA-40%	2.000	0.028*
HA-30%-HA-50%	0.000	0.009*
HA-40%-HA-50%	1.000	0.016*

\*Significantly different ( $P < 0.016$ ). HA: Hydroxyapatite

than that obtained using HA-0% and HA-10% sealers. This finding indicated that the addition of calcite-synthesized HA as a resin filler material had an effect on the apical sealing ability. Factors that affect the apical sealing ability of a resin sealer include adhesion of the material, shrinkage of polymerization of the resin, environmental conditions, and the shape of the cavity that affects the configuration factor or the C-factor.

The C-factor is the ratio between the surfaces of the adhesive material, which, in this case, is the resin that binds to the tooth structure (bonded surface) with the free surface (unbonded surface) of a cavity. The higher the C-factor value, the smaller the resin flow power to compensate for the reduced volume due to shrinkage of polymerization, so that the contraction pressure also becomes greater, thereby reducing the adhesion of the resin to the dentine. During obturation of the root canal, the cavity to be filled by the sealer is relatively closed, and almost the entire surface of the sealer binds to the adherent structure of the root canal wall, the gutta percha, or a small portion of the glass ionomer cement used as a base material in the coronal area. It can be stated that there is no free surface during obturation of the root canal so that the

C-factor value is extremely high. According to Schwartz,<sup>[14]</sup> the C-factor in the root canal can reach 100:1. This may cause all the samples to have apical leakage, although the results shown in Figure 1 in this study indicate that apical leakage occurs only in a small part of the root canal wall. These results indicated that the addition of calcite-synthesized HA could compensate for the C-factor through the flow properties of the material and its setting period that reaches 15 h.

The apical sealing ability of a sealer is the primary parameter that determines the success of obturation of the root canal in a laboratory. Root canal sealing should ideally form a monoblock, that is, the space in the root canal should be completely filled without any gaps by a solid mass that occurs due to the bonding between the root canal and the sealer and between the sealer and the obturation material.<sup>[13]</sup> The formation of the monoblock has an impact on the apical sealing ability or fluid-tight seal in the apical region. During obturation done using gutta percha, it was extremely difficult to achieve the formation of the monoblock. Gutta percha that contains the primary component polyisoprene has no chemical bonds with various types of sealers such as zinc oxide eugenol, glass ionomer cement, and resin sealer,<sup>[3]</sup> so that in the main root canal obturation system, the main apical sealability is the attachment of the sealer to the root canal wall.

The results of this study support those of a previous study<sup>[9]</sup> that epoxy resin sealer with the addition of calcite HA as a filler has good wettability and film thickness. The apical density increased with the addition of HA calcite synthesis because this material does not react with resins but interacts with epoxy resins as well as other fillers. Bisphenol A-diglycidyl epoxy resin cross-links with hexamethylenetetramine to form quaternary ammonium. In addition [Figure 3], the phosphate groups formed from HA calcite synthesis and bismuth oxide as the radiopaque material may interact with quaternary ammonium from the reaction of epoxy resin with hexamethylenetetramine,

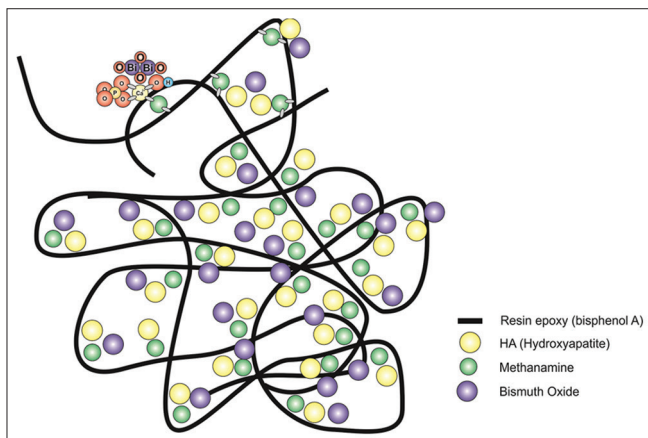


Figure 3: Interaction between hydroxyapatite, bismuth oxide, methanamine, and resin epoxy (Bisphenol A)

which forms a composite. The N group of quaternary ammonium interacts with the O atom of HA. There may also be an interaction between calcium in calcite HA and O atoms in the epoxy resin so that calcite HA can penetrate and be trapped in the resin sealer [Figure 4]. The presence of HA that was penetrated and integrated into the mass of the composite reduces the shrinkage of polymerization. The shortening of the distance between epoxy monomers that occurs during polymerization, as a result of the shrinkage of polymerization, is inhibited by the presence of penetrating HA and interaction with the A-diglycidyl epoxy resin molecules and other fillers, including both hexamethylenetetramine and bismuth oxide.

As shown in Table 1, the addition of 10% HA resulted in the same apical sealing ability as that in the control group. This could imply that the addition of calcite-synthesized HA was not sufficient to inhibit the shrinkage of polymerization. Shrinkage of polymerization results in a high contraction pressure, which could release epoxy resin bonds in the root canal wall that results in gaps or leakage.

After the addition of HA-50%, the apical sealing ability was actually decreased compared to that observed with the addition of HA-10%, HA-20%, HA-30%, or HA-40% [Figure 2]. This was probably due to the microhardness of the resin sealer (7 × 24 h after stirring), which was lower than that in the other groups. Low hardness indicates that the degree of polymerization of the resin sealer is not perfect or imperfect. The hardness of the sealer should not be directly related to density, but it could be highly influential when associated with environmental conditions. When the epoxy resin sealer is not completely polymerized, it will increase the sensitivity of the sealer to moisture, which is more easily dissolved in the periodontal tissue. Under humid conditions, monomers that are not or have not been polymerized could be released faster than the polymer resin so that it affects the physical properties of

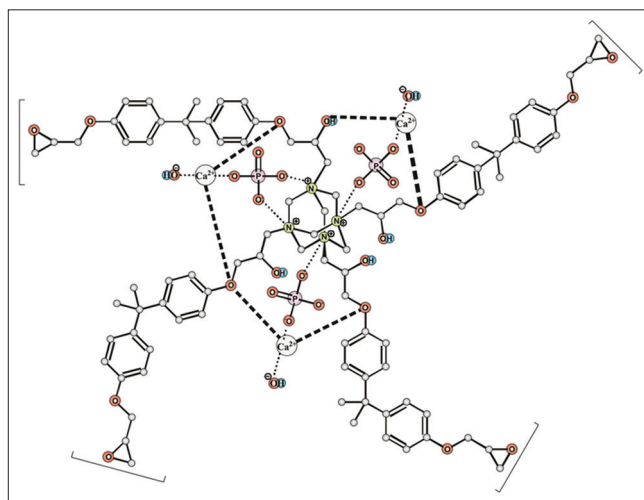


Figure 4: Interaction between hydroxyapatite, bismuth oxide, and quaternary ammonium

the sealer, that is, reducing the adaptation of the sealer to the root canal wall, thereby resulting in apical leakage.<sup>[15,16]</sup>

The HA-30% had the highest sealing ability than other concentrations. This study supported to the result of previous study<sup>[9]</sup> that the concentration of 30% as the maximum concentration of addition calcite synthesized HA to produce sealer hardness. The highest hardness indicated the most perfect polymerization compared to that of other groups. The perfect polymerization could reduce polymerization shrinkage, so leakage was minimized. Other factors to minimize shrinkage were environment and low solubility level.<sup>[17]</sup>

## Conclusion

The addition of calcite-synthesized HA as a filler at concentrations of 20%, 30%, and 40% increased the apical sealing ability of the epoxy resin sealer, but the addition of HA at 50% concentration decreased the apical sealing ability.

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Nil.

## Conflicts of interest

There are no conflicts of interest.

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