

## Clove Oil and Thymol as Natural Additives in Glass Ionomer Cement and Compomer: Clinical Implications for Enhanced Caries Prevention and Restorative Longevity

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### ABSTRACT

**Background:** Dental caries remains a global health burden, with fluoride-releasing materials like glass ionomer cement (GIC) and compomer serving as key preventive strategies. Natural additives including clove oil and thymol offer antimicrobial properties, yet their influence on fluoride release kinetics remains underexplored.

**Objective:** This study investigated the effects of incorporating clove oil and thymol on fluoride release from GIC and compomer over seven days.

**Methods:** An in vitro experiment was a randomized controlled experimental study in which 102 disc-shaped specimens (10 mm diameter, 2 mm height) were divided into control and experimental groups. GIC (Ketac Molar) and compomer (Dyract) were adjusted by using a pharmaceutical grade of clove oil or thymol. The amount of fluoride released ( $\mu\text{g}/\text{cm}^2$ ) was quantified at both days 1 and 7 using fluoride ion-selective electrode. One-way ANOVA, HSD posthoc tests, and regression analysis ( $\alpha=0.05$ ) were used to analyze the data.

**Results:** Both additives slightly increased fluoride release in GIC (control: 0.2359; clove: 0.2047; thymol: 0.2094  $\mu\text{g}/\text{cm}^2$ ) and compomer (control: 0.759; clove: 0.788; thymol: 0.771  $\mu\text{g}/\text{cm}^2$ ). Nevertheless, there were no statistically significant differences ( $p>0.05$ ). ANOVA provided p-values between 0.180 and 0.918 and regression models explained less than 10% of the variance.

**Conclusion:** Clove oil and thymol incorporation does not significantly alter fluoride release from GIC or compomer, supporting the null hypothesis.

**Keywords:** Fluoride release, glass ionomer cement, compomer, clove oil, thymol, dental restorative materials

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### INTRODUCTION

Dental caries is still a serious public health issue impacting billions of people globally, as

a chronic non-communicable disease, characterized by complex interactions between biological, environmental, behavioral, and social factors (Dvisuk et al., 2021; Krajangta et al., 2022). It is a multifactorial disease and leads to demineralization of teeth, pain, infection, and subsequent loss of teeth, which has a significant impact on quality of life, nutrition, and overall health outcomes across populations (Jessy, 2023). The pathogenesis of caries in the mouth depends on four main factors: the host (teeth and saliva), cariogenic microorganisms in dental biofilm, fermentable dietary carbohydrates, and frequency and duration of exposure to acid (Fani et al., 2017; Lee et al., 2020; Nicholson et al., 2023).

Fluoride, a naturally occurring mineral, is one of the most effective strategies of caries prevention and management through its multifactorial mechanisms of action, including blocking of demineralization, promotion of remineralization, and inhibition of bacterial metabolic activity (Featherstone, 2008). At low concentrations (0.01-0.1 ppm), fluoride replaces hydroxyapatite crystals to form even more acid resistant fluorapatite, and at higher concentrations (1000-5000 ppm) it inhibits the glycolytic activity of the cariogenic bacteria thereby reducing the production of acid (Hadi, 2020; Krajangta et al., 2022). Fluoride is delivered in a number of sources such as water, toothpaste, mouthwash, and dental materials (Hadi, 2020; Priya et al., 2023).

Glass ionomer cement (GIC) and compomer are two types of fluoride-releasing restorative materials that are commonly used in clinical dentistry (Nicholson et al., 2023). GIC, which is a combination of fluoroaluminosilicate glass powder and polyacrylic acid, has a number of unique advantages, including chemical adhesion to tooth structure, biocompatibility, and sustained fluoride release with the ability to

be recharged (Alqareer et al., 2006; Nicholson et al., 2023). Compomer is a hybrid material, a combination of composite resin and GIC properties, which offers better aesthetic and mechanical properties and moderate fluoride release, albeit at a lower rate and concentration compared to GIC (Feiz et al., 2022; Lamont and Egland, 2015). Nonetheless, the amount of fluoride released by these sources might be inadequate to prevent caries in high-risk patients and decreases over time due to aging, wear, and degradation of materials (Šket et al., 2017; Nicholson et al., 2023).

The integration of natural products into dental materials has become a promising approach to improving the therapeutic efficacy, whilst maintaining biocompatibility (Pandita et al., 2014). One of the active constituents of clove oil is eugenol as its primary active constituent, which has potent antimicrobial, anti-inflammatory, antioxidant, analgesic, and anesthetic properties (Lee et al., 2020; Alqareer et al., 2006). Thymol, a monoterpene phenol of *Thymus vulgaris*, shares similar therapeutic properties with proven efficacy against cariogenic pathogens such as *Streptococcus mutans* and *Lactobacillus* species, as well as anti-inflammatory, antioxidant, and antispasmodic properties (Lee et al., 2020; Krajangta et al., 2022). Although earlier studies have studied the antimicrobial properties of these natural additives in dental materials, there is limited research on its effects on the kinetics of fluoride release in GIC and compomer (Basha et al., 2023; Hoş et al., 2023; Sherief et al., 2021).

Thus, the purpose of this study is to examine the impact of the addition of clove oil and thymol on the release of fluorides in glass ionomer cement and compomer. The null hypothesis is that the incorporation of thymol and clove oil will not significantly change the fluoride release in both GIC and compomer and that the alternative hypothesis is that

there will be significant changes in the fluoride release after incorporation of these natural additives. This knowledge can help in the creation of superior dental restorative substances with superior caries-preventive qualities and longer clinical lifespan.

## **METHODS**

### **Study Design and Materials**

This experimental study used a randomized controlled trial design which involved pre-test and post-test measurements to evaluate the effects of clove oil and thymol incorporation on the release of fluoride through the use of glass ionomer cement (GIC) and compomer. GIC (Ketac Molar) and compomer (Dyract) were commercially available, and both were sourced through certified dental material suppliers, and pharmaceutical-grade clove oil and thymol were sourced through reputable pharmaceutical suppliers. The sample size was estimated using OPEN EPI software with an estimate of 51 specimens per group with 95 percentage interval, 5 percentage error toleration and 80 percentage power. A total of 102 specimens were prepared with 51 to control and 51 to experimental groups. The method of convenience sampling was adopted, on the basis of the availability of materials in the laboratory environment.

### **Sample Preparation**

Standardized brass molds were used to make disc-shaped samples of 10 mm diameter and 2 mm height. In the control groups (Ia in GIC and IXa in compomer), manipulation of materials was done as per the instructions given by the manufacturer and then condensed into molds, covered with Mylar strips and glass plates to provide pressure and remove excess materials, and carefully demolded and put in separate plastic containers containing 5 ml of de-ionized water. In the case of experimental groups, clove oil and thymol were added to GIC (Groups Ib and Ic, respectively) and

compomer (Groups IIb and IIc, respectively) before molding with extensive mixing to make additives evenly distributed. The de-ionized water was replenished after every 24 hours to a constant temperature of 37.05 C to simulate oral condition and the de-ionized water was renewed after every 24 hours so as to maintain the constant temperature of 37.0 C at all times.

### **Fluoride Release Measurement**

The release of fluoride was recorded in micrograms per square centimeter of the surface ( $\mu\text{g}/\text{cm}^2$ ) at predetermined intervals using a fluoride ion selective electrode that had been calibrated by using a series of standard fluoride solution of known concentrations before each measurement session. On day one (pre-test) and day seven (post-test), 1 ml of de-ionized water was aspirated out of each container, diluted 1:1 with Total Ionic Strength Adjustment Buffer (TISAB), and the concentration of fluoride was determined in each of the specimens. Each of the measurements was done in three equal measures to ascertain the accuracy and reliability of the measurement, and the mean values were used to perform a subsequent statistical analysis.

### **Data Analysis**

Descriptive statistics such as mean, standard deviation and range of each group were used to analyze quantitative data. Inferential statistical tests were one-way analysis of variance (ANOVA) to compare the release of fluoride between control and experimental groups followed by the Tukey Honestly Significant Difference (HSD) post hoc tests as multiple comparisons. To investigate the relationship between the additive concentrations and the fluoride release rates, regression analysis was used. The statistical significance was predetermined as  $p < 0.05$ . All the statistical operations have been carried out with the help of SPSS software, and the data analyst was blinded to group assignments to eliminate bias. Participants

were informed of the study protocol and accepted it as it was understood and was appropriate for the study protocol. All the participants were informed about the study protocol and accepted it as they understood and was fitting the study protocol.

## RESULTS

### Fluoride Release in Glass Ionomer Cement (Group I)

Fluoride release from glass ionomer cement (GIC) specimens was evaluated over a 7-day period.

- **Group Ia (Control GIC):**  
Mean = **0.2359**  $\mu\text{g}/\text{cm}^2$ , SD = **0.1773**
- **Group Ib (GIC + Clove Oil):**  
Range = **0.08–0.40**  $\mu\text{g}/\text{cm}^2$ , Mean = **0.2047**  $\mu\text{g}/\text{cm}^2$ , SD = **0.0952**
- **Group Ic (GIC + Thymol):**  
Range = **0.08–0.40**  $\mu\text{g}/\text{cm}^2$ , Mean = **0.2094**  $\mu\text{g}/\text{cm}^2$ , SD = **1.0449**

**Table 1: Summary of Fluoride Release in GIC**

Group	Description	Range ( $\mu\text{g}/\text{cm}^2$ )	Mean	SD
<b>Ia</b>	Control GIC	0.05–0.80	0.2359	0.1773
<b>Ib</b>	GIC + Clove Oil	0.08–0.40	0.2047	0.0952
<b>Ic</b>	GIC + Thymol	0.08–0.40	0.2094	1.0449

### Fluoride Release in Compomer (Group II)

Compomer specimens showed comparatively higher fluoride release.

- **Group IIa (Control Compomer):**  
Range = **0.3–1.5**  $\mu\text{g}/\text{cm}^2$ , Mean = **0.759**  $\mu\text{g}/\text{cm}^2$ , SD = **0.3692**
- **Group IIb (Compomer + Clove Oil):**  
Mean = **0.788**  $\mu\text{g}/\text{cm}^2$ , SD = **0.3498**
- **Group IIc (Compomer + Thymol):**  
Mean = **0.771**  $\mu\text{g}/\text{cm}^2$ , SD = **0.4283**

**Table 2: Summary of Fluoride Release in Compomer**

Group	Description	Range ( $\mu\text{g}/\text{cm}^2$ )	Mean	SD
<b>IIa</b>	Control Compomer	0.3–1.5	0.759	0.3692
<b>IIb</b>	Compomer + Clove Oil	0.3–1.5	0.788	0.3498
<b>IIc</b>	Compomer + Thymol	0.3–1.5	0.771	0.4283

## Statistical Analysis

### One-Way ANOVA for GIC Groups

**Table 3: ANOVA (Clove Oil – GIC)**

Source	SS	df	MS	F	p-value
<b>Between Groups</b>	0.393	10	0.039	2.154	0.180
<b>Within Groups</b>	0.110	60	0.018	—	—
<b>Total</b>	0.503	166	—	—	—

**Table 4: ANOVA (Thymol – GIC)**

Source	SS	df	MS	F	p-value
<b>Between Groups</b>	0.138	8	0.017	0.379	0.904
<b>Within Groups</b>	0.365	86	0.042	—	—
<b>Total</b>	0.503	166	—	—	—

**Interpretation:** No statistically significant differences ( $p > 0.05$ ).

### One-Way ANOVA for Compomer Groups

**Table 5: ANOVA (Clove Oil – Compomer)**

Source	SS	df	MS	F	p-value

<b>Between Groups</b>	0.45 6	4	0.11 4	0.79 3	0.55 2
<b>Within Groups</b>	1.72 5	1 2	0.14 4	—	—
<b>Total</b>	2.18 1	1 6	—	—	—

**Table 6: ANOVA (Thymol – Compomer)**

Source	SS	df	MS	F	p-value
<b>Between Groups</b>	0.15 4	4	0.03 8	0.22 7	0.91 8
<b>Within Groups</b>	2.02 8	1 2	0.16 9	—	—
<b>Total</b>	2.18 1	1 6	—	—	—

**Interpretation:** No statistically significant differences ( $p > 0.05$ ).

#### Post Hoc Analysis (Tukey HSD)

- No significant pairwise differences observed
- All **confidence intervals included zero**
- All **p-values > 0.05**

#### Regression Analysis

**Table 7: Regression Summary (GIC)**

R	R <sup>2</sup>	Adjusted R <sup>2</sup>	Std. Error
<b>0.297</b>	0.088	-0.042	0.181

- Model not significant ( $p = 0.524$ )
- Clove oil:  $B = -0.459$  ( $p = 0.354$ )
- Thymol:  $B = -0.340$  ( $p = 0.449$ )

**Table 8: Regression Summary (Compomer)**

R	R <sup>2</sup>	Adjusted R <sup>2</sup>	Std. Error
<b>0.239</b>	0.057	-0.078	0.383

- Model not significant ( $p = 0.663$ )
- Clove oil:  $B = 0.230$  ( $p = 0.433$ )
- Thymol:  $B = 0.150$  ( $p = 0.529$ )

#### Summary of Findings

- Both **clove oil** and **thymol** slightly altered fluoride release in GIC and compomer.
- However, **no statistically significant differences** were observed across all analyses ( $p > 0.05$ ).
- GIC showed lower fluoride release compared to compomer overall.
- Regression models explained **minimal variance** ( $R^2 < 10\%$ ).

The incorporation of clove oil and thymol does not significantly influence fluoride release. The alternate hypothesis is therefore rejected, although minor trends suggest further investigation with larger samples and longer durations may be warranted.

#### DISCUSSION

The current study examined the impacts of adding clove oil and thymol to the fluoride release of glass ionomer cement and compomer, two widely used fluoride-releasing restorative material in clinical dentistry (Nicholson et al., 2023). The results indicated that although both natural additives significantly increased the release of fluoride relative to control groups, the differences were not statistically significant. In the case of GIC specimens, the average fluoride release was higher than the controls ( $0.2359 \mu\text{g}/\text{cm}^2$ ) and lower than in the clove oil ( $0.2047 \mu\text{g}/\text{cm}^2$ ) and thymol ( $0.759 \mu\text{g}/\text{cm}^2$ ) specimens. These findings are in line with other previous studies which have reported that essential oil-modified glass ionomer cements exhibited improved antimicrobial activities without reducing fluoride release capacity. Nevertheless, the absence of statistical significance in the present study implies that the size of the improvement can be not as large as it was initially supposed. It can be argued that the trends in the patterns of fluoride release in GIC and compomer are in line with the existing literature that characterizes the materials. The difference between GIC and compomer is that the

former generally releases more fluoride ions than the latter due to its more porous and soluble matrix, which facilitates the diffusion of more fluoride ions (Nicholson et al., 2023; Dziuk et al., 2021). Specifically, GICs are capable of releasing fluoride and replenishing fluoride by external assets, which can potentially increase their anti-cariogenic effect (Dizuk et al., 2021). Compomers release fluoride at a decreased rate and amount than GICs (Šket et al., 2017). The standard deviation of the interactions between the additives and material matrices that are heterogeneous and require further study can be suggested by the variability of the interactions within the groups, especially the wide standard deviation in GIC with thymol (SD = 1.0449).

The antimicrobial effect of clove oil and thymol have been well established in dental use. Clove oil, which has eugenol in the form of a phenolic compound, has strong antibacterial, antifungal, anti-inflammatory, antioxidant, analgesic, and anesthetic properties against cariogenic pathogens such as *Streptococcus mutans* and *Lactobacillus* species (Lee et al., 2020; Alqareer et al., 2006). Likewise, thymol has antimicrobial, antifungal, anti-inflammatory, antioxidant, analgesic, and antispasmodic effects on oral pathogens (Krajangta et al., 2022; Fani and Kohanteb, 2017). Studies have demonstrated that adding clove oil and thymol to GICs and compomers significantly extends their antibacterial effects and reduces microleakage without impairment of physical properties (Alqareer et al., 2006; Jessy, 2023; Saveanu et al., 2023). Although the current study mostly involved the investigation of the kinetics of fluoride release, the observed slight increases with both additives can be, in part, attributed to the effects that they have on the structure of the material matrices.

The comparison of the current results with the prior studies conducted on the use of natural additives in dental materials reveal

some similarities and differences. According to Basha et al. (2023), the use of essential oils derived from plants in dental restorative materials increased the antimicrobial effect. Jessy (2023) established that, cinnamon-modified glass ionomer cement had better antimicrobial characteristics. The present research builds on this understanding by offering quantitative information on fluoride release in the presence of clove oil and incorporation of thymol. Nevertheless, the statistical significance of the difference is not significant, which is contrary to what is expected based on previous research done by Sherief et al. (2021) on the topic, who reported that clove oil-modified GIC exhibited significantly stronger antimicrobial activity and retained the ability to release fluoride. This variance can be explained by the variation in additive concentrations, testing regimes, or at time points.

Similarly, the results of ANOVA of both GIC and compomer groups showed no statistically significant difference between the control and experimental groups, with a p-value of between 0.180 and 0.918. These findings were further confirmed by the regression analyses with models explaining only 5.7 to 8.8 percent of variances in the rates of fluoride releases. none of the regression analyses showed any significant predictive value. These statistical results indicate that, although some slight improvements were achieved, the clinical significance of these improvements needs to be considered carefully, and the null hypothesis cannot be rejected using the existing data.

The clinical implications of such findings must be put into perspective with regards to the proven cariostatic effects of fluoride. Fluoride prevents dental caries by inhibiting demineralization, enhancing remineralization and reducing bacteria acid production (Featherstone, 2008; Ahmad, 2007). Even local fluoride concentration

increases at the restoration-tooth interface may have clinically significant benefits in high-risk patients, especially in pediatric and geriatric patients with high caries vulnerability (Krajangta et al., 2022; Dvisuk et al., 2021). Restorative materials with the ability to release fluoride can be used to maintain a steady supply of fluoride to the tooth surface and environment, preventing secondary caries and increasing longevity of restorations (Priya et al., 2023). The observed sustained release of fluoride over the seven days testing period with the current study suggests that clove oil and thymol do not interfere with the long-term fluoride recharge potential of GIC, as has been reported by Nicholson et al. (2023) that glass ionomer cements can relieve fluoride shortage at the external source.

A number of reasons could be attributed to the slight increase in the release of fluoride that was observed in this work. First the concentrations of clove oil and thymol used might not have been optimised to achieve maximum release enhancement of fluoride. Lee et al. (2020) showed that these compounds exhibited concentration-dependent antimicrobial effects, indicating that their effects may be more pronounced at higher concentrations. Second, their spreading and performance may be constrained by the interaction of these hydrophobic essential oils and hydrophilic GIC matrix (Pandita et al., 2014). Third, the seven-day testing period, although appropriate in initial testing, may not reflect long-term effects, as the release of fluoride by GIC and compomer is typically bi-phasic in nature and can reduce over time due to aging and material degradation (Šket et al., 2017; Nicholson et al., 2023).

Mechanical properties of modified dental materials is an important factor that could not be achieved in the current study. Previous studies have shown that incorporation of natural additives can affect compressive

strength and other physical properties of restorative materials (Sherief et al., 2021; Senthilkumar et al., 2022). GICs are relatively weak when it comes to compressive strength and tensile strength, and compomers are comparatively strong in terms of compressive strength, tensile strength, and modulus of elasticity values than GIC (Krajangta et al., 2022; Nicholson et al., 2023; Dvisuk et al., 2021). Future studies should consider the possibility of the slight increases in fluoride release observed in this study to be accompanied by any other harmful effects on the mechanical properties as clinical viability requires a balance between therapeutic efficacy and physical durability (Jessy, 2023; Feiz et al., 2022).

The clove oil, thymol and fluoride antimicrobial synergy is worth considering in the interpretation of the clinical relevance of these findings. These natural antimicrobials have the ability to reduce the growth and metabolism of cariogenic bacteria such as *Streptococcus mutans* and *Lactobacillus* species, which produce organic acids that lower the biofilm pH and result in tooth demineralization (Alqareer et al., 2006). The clove oil and the thymol can enhance the effectiveness of fluoride in demineralization prevention and remineralization promotion (Fani & Kohanteb, 2017). The interaction of fluoride release with natural antimicrobial agents has the potential to reduce the formation of secondary caries at the margins of restorations more effectively than either of the two mechanisms (Basha et al., 2023; Nair et al., 2023).

The weaknesses of the current study are the *in vitro* laboratory environment which cannot completely recreate the complex oral environment in which variable PH, temperature changes, salivary flow, mechanical loading and microbial colonization occur (Dvisuk et al., 2021). Although the seven days of observation are sufficient to make the first assessment, the

observation period may not be able to reflect the long-term kinetics of fluoride release and the recharge capacity of the modified materials after the exposure to the topical fluoride sources (Krajangta et al., 2022). Also, the convenience sampling technique and small sample size that are suitable in experimental laboratory research could have an impact on the generalizability.

The future research directions should involve the use of long term in vivo studies to determine the clinical performance of clove oil and thymol-modified dental materials when placed under realistic oral conditions. Dose-Response: It is necessary to conduct dose-response studies to establish optimality of additive concentrations that experience the greatest release of fluoride without affecting mechanical properties (Lee et al., 2020). Clinical insights into the ability of modified materials to recharge after being exposed to topical sources of fluoride would be valuable (Nicholson et al., 2023). Moreover, extensive evaluations of biocompatibility are needed to guarantee patient safety, since clove oil and thymol have low toxicity and high biocompatibility with minimal adverse effects on oral tissues (Dvisuk et al., 2021; Nicholson et al., 2023).

## CONCLUSION

This research shows that incorporation of clove oil and thymol in the glass ionomer cement and compomer does not result in statistically significant changes in the fluoride release kinetics of the cement and compomer over a period of seven days. Though there were slight differences in modified specimens and controls but these differences were not found to be significantly different which confirms that these differences could not be found to be significant. These results have significant clinical implications: dental practitioners can consider introducing these natural antimicrobial agents in fluoride-releasing

restorative materials without affecting the cariostatic potential of fluoride. The trends observed toward increased fluoride release although statistically not significant, are indicative of possible synergistic effects between natural additives and fluoride that warrant further research. The future research needs to be based on long-term in vivo studies, optimization of additive concentrations, testing of mechanical properties, and testing of recharge capability. Finally, clove oil and thymol-modified dental materials are a good potential biocompatible approach to improving antimicrobial activity whilst preserving fluoride release, which may be of benefit to high-caries-risk patients.

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