



Research article

The development of resin-coating materials for enhancing elemental release of coated glass ionomer cements

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ABSTRACT

This study aimed to develop resin coatings containing monocalcium phosphate monohydrate (MCPM), Sr/F-doped bioactive glass (Sr/F-BAGs), and pre-reacted glass ionomer fillers (SPG) that enhance ion release without detrimentally affecting the mechanical properties of GIC. The objective of this study was to evaluate the degree of monomer conversion (DC), biaxial flexural strength, surface microhardness, and ion release of the GICs coated with experimental coating materials compared to a commercial product (EQUIA Coat, EC). Four experimental resin coating materials containing 10–20 wt% of MCPM with Sr/F-BAGs and 5–10 wt% SPG were prepared. The DC of the coating material was determined using ATR-FTIR. The flexural strength and surface microhardness of the coated GICs were assessed. Fluoride and elemental (Ca,P,Sr,Si,Al) release were measured using fluoride-specific electrodes and ICP-OES. The DC of the experimental coating material (60–69 %) was higher than that of EC (55 %). The strength of GICs coated with experimental materials (35–40 MPa) was comparable to EC (37 MPa). However, their surface microhardness (13–24 VHN) was lower than EC (44 VHN). The experimental coating materials reduced fluoride release by ~43 %, similar to EC (~40 %). However, experimental coating materials promoted higher P and Sr release than EC. In conclusion, GICs coated with the experimental resin coating containing ion-releasing additives exhibited mechanical properties similar to those of the commercial product. The new coating materials promoted a higher level of ion release for GICs. These properties could potentially enhance remineralizing actions for the coated GICs.

1. Introduction

Managing untreated dental caries continues to be a significant financial challenge for the oral health sector. Despite substantial

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efforts being utilized to address this issue, the number of incidents of untreated caries is still high, reported at 3.09 billion cases in permanent teeth and 1.15 billion in deciduous teeth globally in 2019 [1]. The lesions that were left untreated will ultimately progress to cavitated carious lesions, requiring restorations with adhesive materials such as resin composites or glass ionomer cement (GICs). The limitation of resin composites is the risk of secondary caries due to their technique-sensitive placing protocol, particularly in cavities with inadequate moisture isolation [2]. A suitable alternative material is glass ionomer cement (GICs). These water-based materials exhibited attractive properties, including chemical adhesion with tooth structure and fluoride release, which could enhance the remineralization of the carious dentine [3,4].

The primary limitation of GICs is their low physical and mechanical strength compared to resin composites [5]. It is generally recommended that GIC should be predominantly used in small to moderate load-bearing restorations such as cervical restorations or class I cavities [6–8]. Another critical factor to consider with GICs is their sensitivity to moisture during the setting and maturation process. Hence, GIC is usually coated with varnish, petroleum jelly, or resin-based materials to control water loss from the material after finishing. This resin coating offers multiple benefits. For example, it helps control water loss from the material, reduce the microleakage [9], wear [5,10], and color change [11], and enhance the mechanical strength of the materials. Studies have demonstrated that the flexural strength of coated conventional GIC and resin-modified GIC (RMGIC) was 63 % and 49 % higher than that of uncoated restorations, respectively [12]. The proposed mechanisms was that resin coating may seal cracks and porosities to enhance materials' physical and mechanical properties [13]. The application of a rigid, polymerized resin coating over GICs may, however, act as a barrier to ion diffusion. An *in vitro* study demonstrated that a resin coating significantly reduced the fluoride release of GICs from 203 ppm to 66 ppm [14]. Additionally, the use of dental adhesive as a coating material for GICs or other ion-releasing materials resulted in a substantial decrease in ion release by ~307 times [15].

The addition of calcium phosphate compounds such as monocalcium phosphate monohydrate (MCMPM) or tricalcium phosphate to resin-based material promoted the release of Ca and P [16]. Similarly, bioactive glass facilitates the release of multiple ions, such as Ca, P, and Sr, which is expected to encourage multiple actions, such as remineralization and antibacterial properties [17]. These ions were expected to help promote the precipitation of hydroxyapatite, remineralize the demineralized dentine, and prevent bacterial penetration at the marginal leakage [18,19]. Additionally, pre-reacted glass ionomer fillers were also expected to improve the release of essential ions, such as fluoride [20–22]. The addition of these ion-releasing additives to resin-coating materials was expected to improve ion release from coated GICs. However, it is necessary to examine the effects of using hydrophilic components on the mechanical strength of coated GICs. Furthermore, the resin coating currently used for GICs contained a high level of methyl methacrylate monomer (MMA, 25–50 wt%). These monomers may be responsible for the unpleasant odor experienced by patients and dental professionals [23]. Additionally, the direct contact or inhalation of MMA monomers could lead to health concerns due to the risk of skin sensitization and respiratory irritation [24–26].

The aim of this study was to formulate resin-coating materials without the MMA monomer. The materials were added with ion-releasing fillers, including monocalcium phosphate monohydrate (MCMPM), Sr/F-bioactive glass (Sr/F-BAGs), and pre-reacted glass ionomer fillers (SPG) to encourage ion-releasing actions from coated GICs. The first objective was to assess the monomer conversion, flexural strength, surface microhardness, and elemental release compared with a commercial material. Additionally, the second objective was to investigate the effect of increasing the concentrations of Ca/Sr (comprising MCMPM and BAG) and SPG on these tested properties. The null hypothesis was that the prepared resin coating exhibited no significant difference in tested properties compared with the commercial material. Furthermore, it was hypothesized that increasing the concentrations of Ca/Sr and SPG exhibited minimal effect on the physical and mechanical characteristics of the materials in the current study.

2. Materials and methods

2.1. Preparation of ion-releasing fillers

A sol-gel process was used to fabricate the Sr/F-bioactive glass (Sr/F-BAGs) using a method similar to the previous study [21]. Briefly, silica nanoparticles (SiO_2) were synthesized by mixing of ethanol (Merck, Darmstadt, Germany), deionized water, and ammonium hydroxide (Merck, Darmstadt, Germany) in an Erlenmeyer flask and stirred at 600 rpm for 15 min. Then, tetraethyl orthosilicate (TEOS, Sigma-Aldrich, St. Louis, MO, USA) was added and stirred for 16–18 h. The resulting silica nanoparticles were centrifuged, followed by re-suspension in deionized water. For doping, $\text{Ca}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $\text{Sr}(\text{NO}_3)_2$, and NaF (Merck, Darmstadt, Germany) were added to the SiO_2 particles by maintaining a specific molar ratio of $\text{SiO}_2:\text{CaO}:\text{SrO}:\text{NaF}$ at 1.0:0.33:0.98:0.5. Then, the particles were dried, calcined at 680 °C to remove nitrate precursors, and washed with ethanol.

Pre-reacted glass ionomer fillers (SPG) were produced using the method described in a previous study [21]. Fluoroaluminosilicate glass ($\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaF}_2\text{-ZrO}_2$) were synthesized by mixing SiO_2 (Ajax Finechem, Thermo Fisher Scientific, Waltham, MA, USA), Al_2O_3 (Fluka Analytical, Honeywell Fluka, Charlotte, NC, USA), P_2O_5 (ACROS ORGANICS, Thermo Fisher Scientific, Waltham, MA, USA), CaF_2 (Merck, Darmstadt, Germany), ZrO_2 (Sigma-Aldrich, St. Louis, MO, USA) and SrCO_3 (Sigma-Aldrich, St. Louis, MO, USA). These mixtures were then heated at 1450 °C for 2 h in a platinum-rhodium crucible and cooled in water to obtain glass frits. They were ground with a planetary micro mill (Fritsch Pulverisette 7, FRITSCH, Idar-Oberstein, Germany) and refined by ball milling (Mastersizer 2000, Malvern Instruments, Malvern, UK). The resulting glass was mixed with deionized water in a 3:7 wt ratio and 2 wt% of polyacrylic acids to form a slurry. Then, this slurry mixture was spray-dried, producing pre-reacted glass fillers (SPG).

To examine the microscopic characteristics of fillers, they were sputter-coated with gold at 23 mA for 45 s. Then, they were examined using a scanning electron microscope (SEM, JSM 7800F, JOEL, Tokyo, Japan) using an accelerated voltage of 5–15 kV.

2.2. Preparation of resin coating materials

The experimental resin-coating materials were prepared using two components: a monomer phase and a powder phase (Fig. 1). The monomer phase was the mixture of 60 wt% urethane dimethacrylate (UDMA, Sigma–Aldrich, St. Louis, MO, USA), 36 wt% triethylene glycol dimethacrylate (TEGDMA), 2 wt% 10-methacryloyloxydecyl dihydrogen phosphate (10-MDP, Sigma–Aldrich, St. Louis, MO, USA), 1 wt% diphenyl (2,4,6-trimethyl benzoyl) phosphine oxide (TPO, Sigma–Aldrich, St. Louis, MO, USA), and 1 wt% dimethylaminoethyl methacrylate (DMAEMA, Sigma–Aldrich, St. Louis, MO, USA). All chemicals were mixed with a magnetic stirrer in a brown bottle for 24 h at 25 °C until the mixed monomers were clear.

The powder phase of the experimental formulations contained silanated baroaluminasilicate glass (Esstech, Essington, PA, USA), monocalcium phosphate monohydrate (MCPM, Himed, Old Bethpage, NY, USA), Sr/F-doped bioactive glass (Sr/F-BAGs, King Mongkut’s University of Technology Thonburi, Bangkok, Thailand), and pre-reacted glass ionomer fillers (SPG, National Science and Technology Development Agency, Pathum Thani, Thailand).

Five experimental formulations with varying powder phase compositions were prepared (Table 1). Two variables were investigated in the formulation of experimental materials. The first variable was the total concentration of MCPM with Sr/F-BAGs (defined as Ca/Sr), which was either 10 wt% or 20 wt%. The second variable was the concentration of SPG, which was either 5 wt% or 10 wt%. These concentrations were chosen based on the formulation of resin composites for restoration and orthodontic adhesive in the previous studies [27,28].

The powder components (Table 1) were weighed using a 4-digit balance. They were then manually mixed with the prepared monomer phase at a 1:3 powder-to-liquid mass ratio (PLR) or 25 % filler to 75 % monomer liquid in a rubber cup within 20 s to obtain a homogeneous paste of resin coating materials. A low PLR was to enhance the flowability of the resin coating materials. The prepared resin coating was placed in a 5 mL black adhesive bottle and kept at 4 °C. A commercial resin coating material (EQUIA Forte Coat, GC Corporation, Tokyo, Japan) was used for comparison (Table 2).

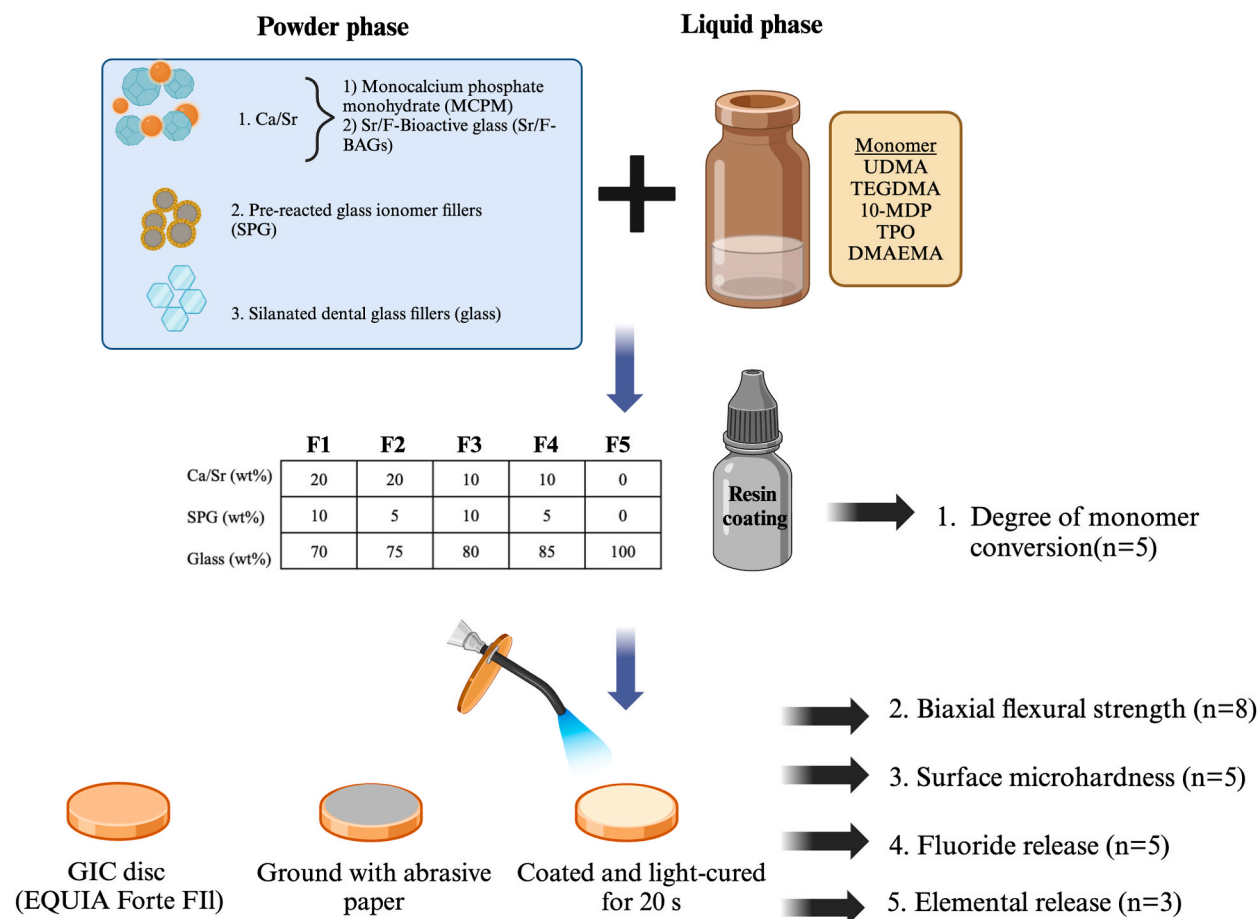


Fig. 1. Experimental setup of the current study (Created with BioRender.com).

Table 1

Composition (wt%) of the experimental resin-coating material in the current study. The powder to liquid ratio was fixed at 1:3 mass ratio (25 wt% fillers and 75 wt% liquid).

Composition of powder phase in each formulation	F1 (wt%)	F2 (wt%)	F3 (wt%)	F4 (wt%)	F5 (wt%)
1) Silanated baroaluminosilicate glass	70	75	80	85	100
2) Ca/Sr Monocalcium phosphate monohydrate (MCPM)	10	10	5	5	0
Sr/F- bioactive glass (Sr/F-BAG)	10	10	5	5	0
3) Pre-reacted glass ionomer fillers (SPG, diameter)	10	5	10	5	0

Table 2

Composition of the commercial material used in the current study.

Materials	Composition	Lot number	Supplier
EQUIA Forte HT Fil (capsule)	Fluoroaluminosilicate glass, polyacrylic acid, surface-treated glass	2302091	GC Corporation, Tokyo, Japan
EQUIA Forte Coat	25–50 % methyl methacrylate (MMA), camphorquinone, colloidal silica, urethane dimethacrylate, phosphoric acid ester monomer, butylated hydroxytoluene	2301231	GC Corporation, Tokyo, Japan

2.3. Degree of monomer conversion (DC)

An attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR, Nicolet is5, Thermo Fisher Scientific, Waltham, NA, USA) was employed to determine the DC of the resin coating materials upon light curing. The experimental resin coatings were placed on the ATR diamond ($n = 5$) and cured using an LED light-curing unit (1500 mW/cm², Eighteen CuringPen, Changzhou Sifary Medical Technology, Jiangsu, China) for 20 s. FTIR spectra in the range of 700–4000 cm⁻¹ at a resolution of 4 cm⁻¹ and eight repetitions were obtained before and after light-curing. The temperature for testing was set at 25 ± 1 °C. The degree of monomer conversion (DC) was then obtained using Equation (1) [29].

$$DC = \frac{100(\Delta A_0 - \Delta A_t)}{\Delta A_0} \quad \text{Equation 1}$$

This formula, ΔA_0 and ΔA_t represents the peak height of C–O stretching vibration of the methacrylate group at 1320 cm⁻¹ above the baseline level at 1335 cm⁻¹, measured before curing and at the time t after curing, respectively [30].

2.4. Biaxial flexural strength (BFS) and biaxial flexural modulus (BFM)

The high-viscosity glass ionomer cement or glass hybrid (EQUIA Forte Fil HT, GC Corporation, Tokyo, Japan) was prepared according to the manufacturer's instructions. The mixed cement was injected into a metal circlip (10 mm in diameter and 1 mm thick). The cement was covered with acetate sheets and glass slides to produce a disc specimen. They were allowed to set at 25 ± 1 °C for approximately 1 h. Then, the specimen was removed from the circlip, and one side was ground with no. 500 abrasive paper for 10 s, followed by 5-s rinse with water. The ground surface was then blot-dried and coated with resin-coating materials using a micro brush dental applicator, then light-cured for 20 s.

Specimens ($n = 8$) were immersed in 5 mL of deionized water and stored in an incubator at 37 °C for 24 h. Then, the thickness of each specimen was measured with a digital vernier caliper. BFS and BFM of the specimens were then tested using a ball-on-ring setup on a mechanical testing frame (AGSX, Shimadzu, Kyoto, Japan). A load of 500 N was applied using a 4 mm diameter spherical ball indenter at a crosshead speed of 1 mm/min. The BFS (Pa) and BFM (Pa) were obtained using Equation (2) and Equation (3), respectively [31].

$$BFS = \frac{F}{d^2} \left\{ (1 + \nu) \left[0.485 \ln \left(\frac{r}{d} \right) + 0.52 \right] + 0.48 \right\} \quad \text{Equation 2}$$

$$BFM = \left(\frac{\Delta H}{\Delta W_c} \right) \times \left(\frac{\beta_c d^2}{q^3} \right) \quad \text{Equation 3}$$

In these equations, F represents the failure load (N), d is the specimen thickness (m), r is the radius of the ring support of the testing jig (m), and ν is Poisson's ratio (0.3) [31]. The term $\frac{\Delta H}{\Delta W_c}$ denotes the rate of load change relative to central deflection (N/m). β_c and q , which are the center deflection function (0.5024) and the ratio of the support radius to the specimen radius, respectively.

A specimen from each group was chosen and placed in a blank tube. The fracture surface was then sputter-coated with gold (Q150R, Quorum Technologies, East Sussex, UK) at 23 mA for 45 s. They were examined under a scanning electron microscope (SEM, JSM 7800F, JOEL, Tokyo, Japan) equipped with an energy-dispersive X-ray spectroscopy (EDX, X-Max 20, Oxford Instruments, Abingdon, UK) at an accelerated voltage of 15 kV. EDX mapping was conducted at the center of the coating resin to analyze the

elemental composition of the material.

2.5. Vickers surface microhardness

Coated glass ionomer cement (GIC) disc specimens were prepared similarly to those in the biaxial flexural strength test ($n = 5$). The Vickers microhardness tester (FM-800, Future-Tech Corp, Kanagawa, Japan) was employed to determine the surface microhardness. Each specimen was loaded with a 300 g load for 10 s. The Vickers hardness number (VHN) obtained from each specimen was averaged from four different areas on the coated surface. The specimens were subsequently immersed in 5 mL of deionized water at 37 °C. The test was performed on the same side of the specimens at 24 h, 1 week, 2 weeks, and 4 weeks.

2.6. Elemental release

Coated-GIC disc specimens were prepared using the same protocol as the biaxial flexural strength test ($n = 5$) to determine fluoride release. The specimens were immersed in 5 mL of deionized water and kept at 37 °C. A calibration curve of fluoride measurement was determined using standard fluoride solutions at concentrations of 1, 10, 100, and 1000 ppm. At 24 h post-immersion, the storage solutions from each specimen were mixed with TISAB II (Orion ionplus, Thermo Scientific, Waltham, MA, USA) in a 1:10 vol ratio. Fluoride concentration was subsequently determined using the fluoride-specific electrode (Orion Versastar Pro, Thermo Scientific, Waltham, MA, USA). Then, the specimens were placed in a tube containing 5 mL of fresh solution. The test was repeated at 1 week, 2 weeks, and 4 weeks.

For analyzing the release of essential elements of GICs, including Aluminum (Al), Phosphorus (P), Calcium (Ca), Silicon (Si), and Strontium (Sr), separate disc specimens were prepared and placed in a tube containing 5 mL of deionized water ($n = 3$). These tubes were then incubated at 37 °C for 4 weeks. After the immersion period, the concentrations of the elements in the storage solution were measured using inductively coupled plasma optical emission spectrometry (ICP-OES, Optima 8300, PerkinElmer, Waltham, MA, USA). The analysis was calibrated using an Environmental standard containing 26 components (CPA Chem, Bogomilovo, Bulgaria). The specific wavelengths and detection ranges used for the measurements were 396 nm (0.1–20 ppm) for Al, 317 nm (0.1–50 ppm) for Ca, 213 nm (0.5–20 ppm) for P, 251 nm (0.1–50 ppm) for Si, and 460 nm (0.1–50 ppm) for Sr.

2.7. Statistical analysis

The numerical results were presented as mean and SD. Statistical analysis was performed using Prism 10 for MacOS (GraphPad Software, Boston, MA, USA). The raw data was provided in supplementary information (See File S1). The distribution of data was assessed with the Shapiro-Wilk test. For data following a normal distribution, analysis was carried out using one-way ANOVA, followed by the Tukey test for post hoc comparisons. The Kruskal-Wallis test, followed by the Dunn test, was employed for non-normally distributed data. Furthermore, one-way repeated ANOVA was employed to compare the fluoride release and surface microhardness within the same group. A significance level was set at $p = 0.05$.

G*Power version 3.1.9.6 (University of Düsseldorf, Düsseldorf, Germany) was utilized to conduct a power analysis [32,33]. Mean and standard deviations from previous studies [12,28,34] were used to calculate the effect size (Cohen's f) and determine the required sample size for each test. It was indicated that sample sizes of $n = 5, 8, 5,$ and 5 would achieve a power exceeding 0.95 at an alpha level of 0.05 for assessing monomer conversion, biaxial flexural strength, surface microhardness, and elemental release, respectively. Additionally, a factorial analysis was conducted to assess the effect of increased concentrations of Ca/Sr (from 10 to 20 wt%) and SPG (from 5 to 10 wt%) on the properties under investigation [27].

3. Results

3.1. SEM analysis of the powder phase

SEM images of MCPM powder revealed particles with irregular shapes (Fig. 2). In contrast, the particles of Sr/F-BAGs and SPG were spherical, with diameters of approximately 0.1–0.2 μm and 10–20 μm , respectively.

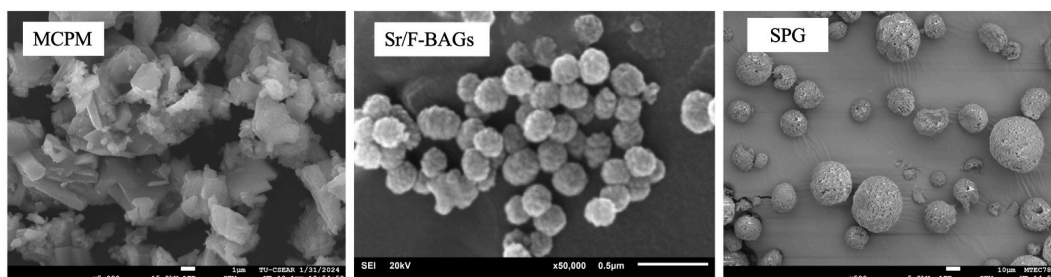


Fig. 2. SEM images of the reactive fillers used in the current study.

3.2. Degree of monomer conversion (DC)

The highest and lowest DC was detected with F4 ($70.2 \pm 1.6\%$) and F2 ($60.2 \pm 0.5\%$), respectively (Fig. 3). The DC of EC ($65.5 \pm 17.3\%$) was comparable to that of the experimental formulations F1 ($69.4 \pm 1.3\%$), F2, F3 ($68.5 \pm 3.3\%$), F4, and F5 ($69.7 \pm 1.8\%$) ($p > 0.05$). Additionally, no significant difference in DC was observed among the experimental materials ($p > 0.05$). Factorial analysis indicated that an increase in the concentrations of Ca/Sr led to a reduction in DC by approximately $7 \pm 1\%$, especially at high concentrations of SPG.

3.3. Biaxial flexural strength (BFS) and biaxial flexural modulus (BFM)

The highest mean BFS was recorded for F5 at 40 ± 6 MPa, while the group without coating showed the lowest mean BFS at 30 ± 5 MPa (Fig. 4A). F5 also exhibited a significantly higher BFS compared to the no-coating group ($p = 0.040$). EC showed a similar BFS (37 ± 6 MPa) to that of F1 (35 ± 4 MPa), F2 (38 ± 7 MPa), F3 (30 ± 5 MPa), F4 (39 ± 7 MPa), F5 (40 ± 6 MPa), and no-coating ($p > 0.05$).

The highest BFM was detected with F2 (2.00 ± 0.53 GPa), while the lowest was recorded for F1 (1.50 ± 0.16 GPa)(Fig. 4B). However, there were no significant differences in BFM among all groups ($p > 0.05$). The factorial analysis showed that the increase in concentrations of both Ca/Sr and SPG showed negligible effects on the strength of materials.

The resin coating layer was observed on the tested specimens in groups F1–F5 and EC (Fig. 5). The thickness of this layer in F3, F4, F5, and EC was approximately $40\text{--}50\ \mu\text{m}$, whereas that of F1 and F2 was $\sim 100\ \mu\text{m}$. EC demonstrated good adaptation between the resin coating and GIC compared to the experimental groups.

3.4. Vickers surface microhardness

In general, the surface microhardness of all groups was reduced after immersion in water (Fig. 6). A significant reduction was detected with F1, F4, F5, and EC ($p < 0.05$). At 4 weeks, the highest surface microhardness was detected with the no-coating group (92 ± 5 VHN), whilst the lowest value was recorded for F2 (15 ± 3 VHN). The surface microhardness of EC (46 ± 11 VHN) was significantly higher than that of F2 ($p = 0.002$). No significant differences were detected among the surface microhardness of experimental materials ($p > 0.05$). Factorial analysis revealed that increasing the concentration of Ca/Sr from 5 to 10 wt% reduced the surface microhardness at the 4-week by approximately $28 \pm 12\%$. The increase in SPG concentration resulted in an increase in surface microhardness by approximately $28 \pm 15\%$.

3.5. Elemental release

All groups exhibited a significant increase in cumulative release upon immersion time ($p < 0.05$)(Fig. 7). The highest mean cumulative fluoride release at 4 weeks was detected with the no-coating group (28.9 ± 5.3 ppm), while the lowest value was obtained from F4 (14.6 ± 1.5 ppm). The fluoride release of EC (17.4 ± 3.2 ppm) was comparable to all experimental groups ($p > 0.05$). A

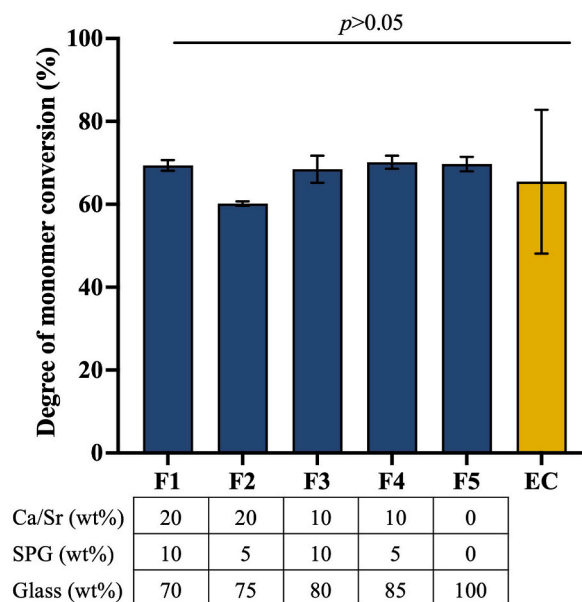


Fig. 3. The degree of monomer conversion of experimental resin-coating materials and the commercial product after light-curing for 20 s. Error bars are SD ($n = 5$). The line indicates the p -value.

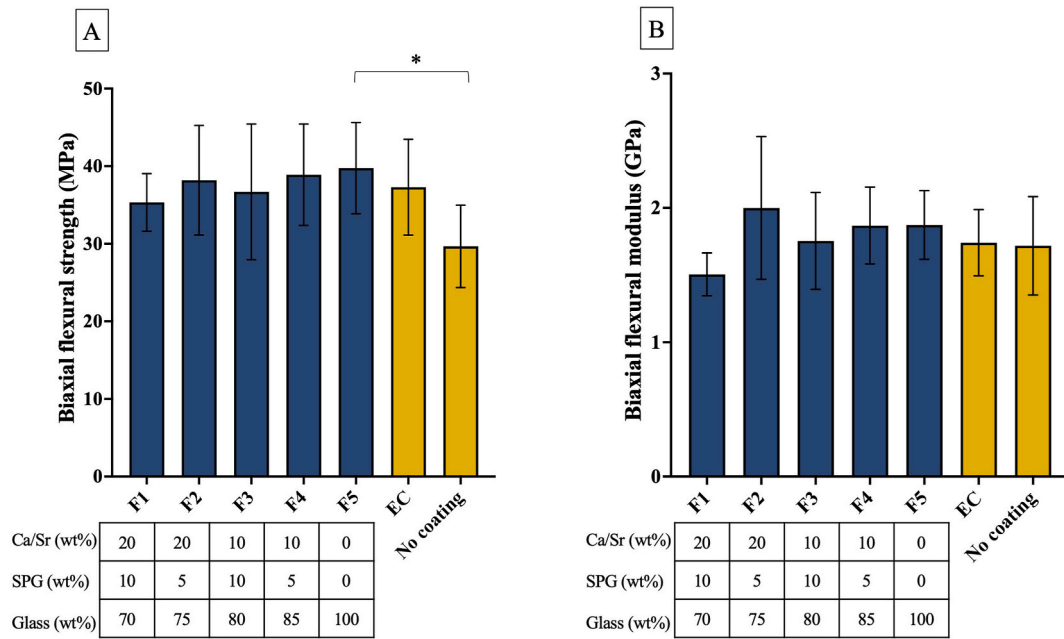


Fig. 4. The (A) biaxial flexural strength and (B) biaxial flexural modulus of glass ionomer cement coated with experimental resin-coating materials (F1–F5), commercial material (EC), and the group with no coating. The test was conducted after 24 h immersion in water. Error bars are SD (n = 8). Star (*) indicated $p < 0.05$.

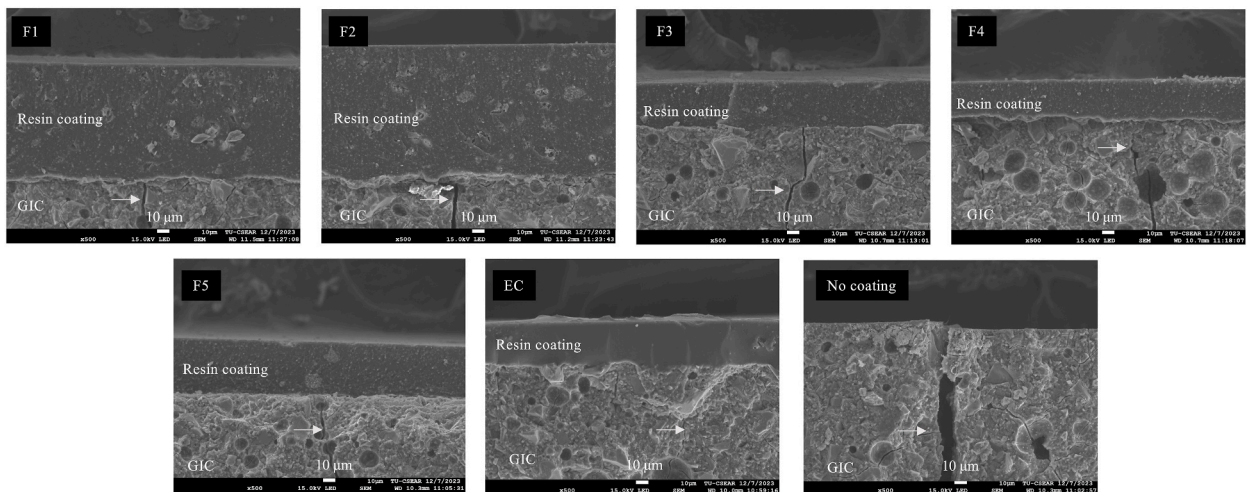


Fig. 5. The SEM image of the fracture surface of the representative sample after BFS testing. The cracks (arrows) were detected within the bulk of the materials.

significant difference was detected between No-coating with F5 (15.9 ± 1.3 ppm)($p = 0.04$) and F4 ($p = 0.003$). No significant difference in fluoride release was observed across various experimental formulations ($p > 0.05$). The factorial analysis indicated that increasing the Ca/Sr concentration from 5 to 10 wt% led to an approximately 18 ± 2 % increase in cumulative fluoride release. On the other hand, changes in SPG concentration exhibit a minimal effect on fluoride release.

The release of Ca among each material was below the detection limit (Table 3). The no-coating group showed the highest release of Al (7.67 ± 0.65 ppm) and Si (12.07 ± 1.17 ppm). The highest P release was observed with F2 (2.14 ± 0.02 ppm), significantly greater than that of the EC group (0.51 ± 0.10 ppm) and the no-coating group (1.20 ± 0.07 ppm). The Si release among the experimental materials and EC was comparable ($p > 0.05$). For Sr release, the highest value was detected with F2 (0.38 ± 0.03 ppm), which was also significantly higher than EC (0.31 ± 0.02 ppm) and no-coating group (0.30 ± 0.01 ppm)($p < 0.05$). The increase in Ca/Sr concentration showed minimal effect on Al and Si release, but it promoted the release of P and Sr by approximately 156 ± 15 % and 15 ± 8 %, respectively.

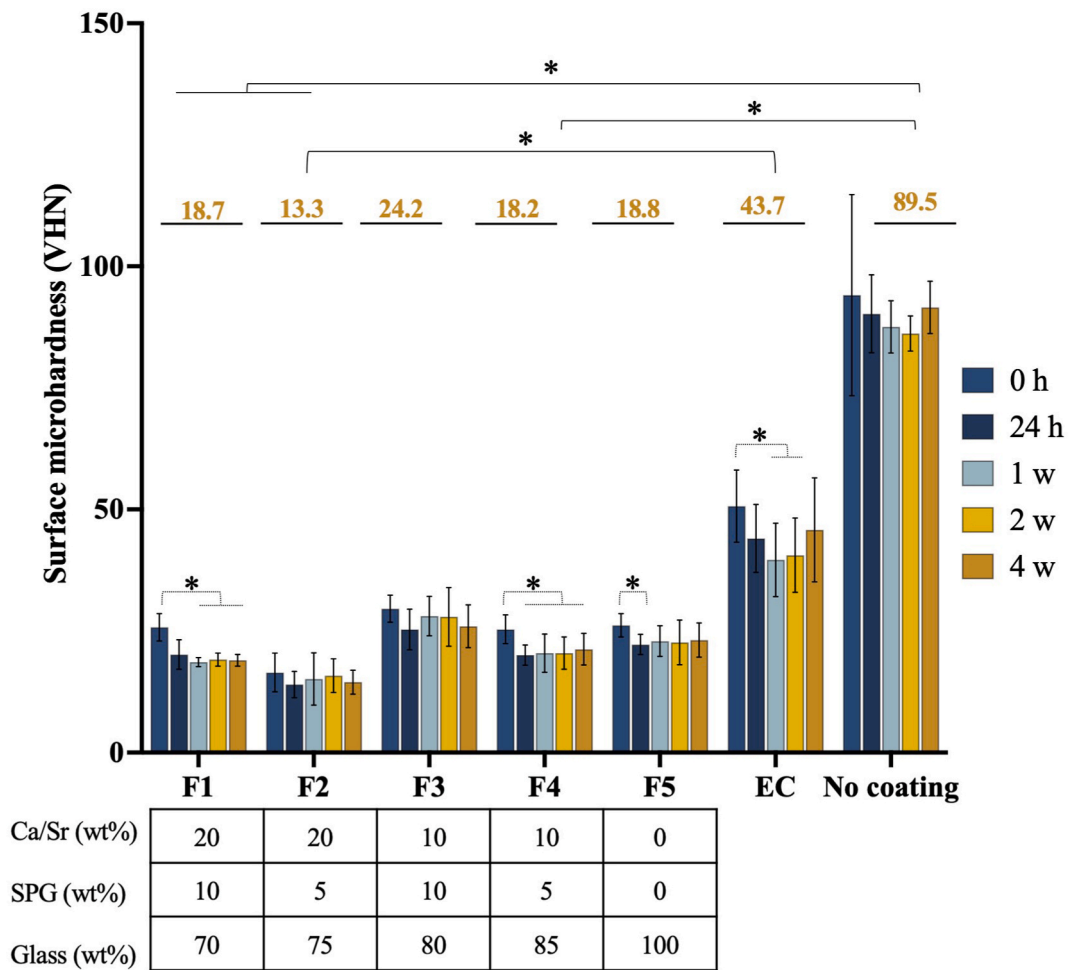


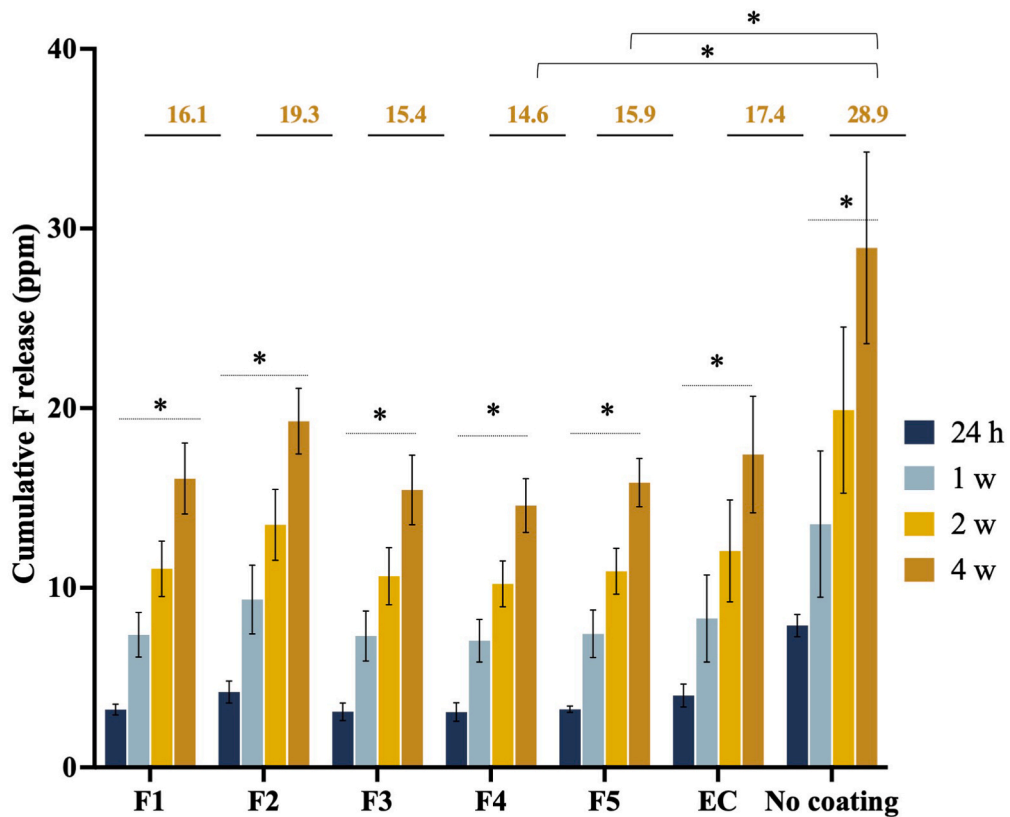
Fig. 6. The Vickers surface microhardness of the specimens before (0 h) and after immersion in deionized water for 24 h, 1, 2, and 4 weeks. The numerical values above each formulation are the surface microhardness at 4 weeks. Error bars are SD (n = 5). The solid lines compare the final surface microhardness value at 4 weeks. The dotted lines compare the values within the same group. Stars (*) indicated $p < 0.05$.

4. Discussion

This study aimed to examine the physical and mechanical properties of conventional GIC when coated with experimental resin coating. The experimental coating contained a mixture of ion-releasing fillers, including Sr/F-bioactive glass, monocalcium phosphate monohydrate, and pre-reacted glass ionomer fillers. The findings revealed that the GIC coated with experimental resin coating materials demonstrated physical and mechanical properties similar to GICs coated with a commercial product (EQUIA Coat), with the exceptions being surface microhardness and elemental release (P and Sr). Consequently, the null hypothesis was partially rejected.

The high degree of monomer conversion (DC) in the coating materials was expected to increase polymer rigidity and reduce the risk of unreacted monomer release [35]. This could potentially reduce the surface loss of GICs [13], particularly in high-risk areas exposed to acids from dysbiotic dental biofilms or mechanical abrasion. The DC of the experimental resin coating was found to be within the range of the commercial product. The addition of additives also exhibited no detrimental effect on the DC. This outcome might be attributed to using a low powder-to-liquid ratio in the formulation, which was intended to improve material flow. Consequently, such a concentration of filler content may not substantially increase the light-scattering effect [36].

A study reported that light-curable MMA-based resin exhibited a DC of 40–60 % [37]. This was in agreement with the result observed with MMA-based material (EC) used in the current study. Using low molecular weight monomers, such as MMA monomers (100.12 g/mol), may reduce viscosity and increase the resin mixture’s flowability [38]. This may facilitate the sealing of irregularities or defects on the surface of GICs and enhance the wear resistance of coated GICs [39]. However, MMA exhibited some limitations, such as an unpleasant odor. Moreover, it was reported to be a respiratory irritant and dermal sensitizer, potentially leading to occupational asthma in some cases [40,41]. The modification of a monomer system without MMA may help mitigate these concerns. This study introduces simple formulations of methacrylate-based resin coating, demonstrating a DC within the range of the commercial material. The di-methylmethacrylate monomers, such as UDMA, contained two polymerizable groups for cross-linking [42], which may lower



Ca/Sr (wt%)	20	20	10	10	0
SPG (wt%)	10	5	10	5	0
Glass (wt%)	70	75	80	85	100

Fig. 7. The cumulative fluoride release at 24 h, 1, 2, and 4 weeks. The numerical values above each formulation are values at 4 weeks. Error bars are SD (n = 5). The solid lines compare the final surface microhardness value at 4 weeks. The dotted lines compare the values within the same group. Stars (*) indicated $p < 0.05$.

Table 3

The concentration (mean and SD) of the released elements (ppm) after immersion in water for 4 weeks. NA represent the value that is under the detection limit of the instrument.

Element/formulations	F1	F2	F3	F4	F5	EC	No-coat
Ca	NA	NA	NA	NA	NA	NA	NA
Al	2.15 (1.11)	2.53 (0.62)	2.00 (0.53)	2.09 (0.09)	1.61 (0.28)	2.51 (1.17)	7.67 (0.65)
P	1.39 (0.30)	2.14 (0.02)	0.79 (0.07)	0.57 (0.08)	0.44 (0.03)	0.51 (0.10)	1.20 (0.07)
Si	5.37 (1.83)	7.08 (0.82)	4.92 (0.46)	4.79 (0.22)	3.99 (0.71)	5.19 (1.61)	12.07 (1.17)
Sr	0.30 (0.02)	0.38 (0.03)	0.31 (0.03)	0.27 (0.02)	0.27 (0.02)	0.31 (0.02)	0.30 (0.01)

the risk of monomer release from the polymer network compared with monomers containing only a single polymerizable group, such as mono methacrylate monomer (MMA). However, further investigations are necessary to assess the leaching of unreacted monomers and the toxicity of these experimental resin-coating materials.

In this study, the surfaces of glass ionomer cement specimens were artificially roughened with no. 500 sandpaper. The aim was to simulate defects that might be due to finishing/polishing steps or abrasions on the surface of GICs. It was hypothesized that the coating resin could improve the mechanical properties of the GICs. This hypothesis aligns with the strength test results, which indicated that most of the coated group exhibited higher BFS than the uncoated group. The result from the current study also correlates with the previous study, which showed that coated GIC exhibited higher flexural strength than uncoated GIC [12]. It was speculated that the beneficial effect of resin coating was not as significant as in the previous study (2–3 times increase) [12]. This study performed a biaxial flexural strength test primarily because of the ease of preparing disc-shaped specimens. This may reduce the variation of results due to

the risk of incorporating flaws or defects in the specimens compared to beam and rod specimens used in 3-point bending and compression tests, respectively [43].

The increase of strength upon coating could be attributed to the low-viscosity resin helping to seal defects or cracks on the surface, potentially delaying the formation of cracks originating from these imperfections. Thus, applying resin-coating material is particularly beneficial for GICs with surface cracks or defects. This is especially relevant in some limited treatment conditions, such as outreach or mobile clinical services, where finishing/polishing procedures may be inadequate due to limited equipment. However, the crack observed in the SEM images of the tested specimen could have occurred due to the drying of specimens after the test or the vacuum process during the SEM analysis.

A higher thickness of the coating layer was observed in the coating resin that contained higher levels of MCPM (diameter $\sim 10\ \mu\text{m}$) and Sr/F-bioactive glass (diameter $\sim 0.2\ \mu\text{m}$). The viscosity of resin-based materials may increase as particle size decreases [44], potentially leading to a thicker coating layer. This aspect is particularly crucial in occlusal restorations, where an excessively thick coating might result in high occlusion, causing discomfort for patients. The air-drying step to thin the coating layer may be required if used in the patient. It could be possible that the different concentrations of additives caused by the variation in thickness of the coated layer may influence the strength and hardness of the materials. This could be the reason for the lack of significant findings among the F1–F4 in mechanical and hardness tests. The high thickness of the resin coating layer could additionally influence the release of ions from the GIC, as the coating layer acts as a barrier to ion diffusion [15]. Future work should characterize the effect of fillers on the rheological properties of the experimental resin coating materials to ensure that the viscosity and thickness of the coating resin could be controlled.

The non-coated group exhibited higher surface microhardness, as was expected. The result was in agreement with the previous study [45]. This could be due to the indenter pressing mainly against the highly rigid remaining fluoroalumininate glass fillers. The average surface microhardness of the GIC coated with experimental materials was lower than that coated with EC. A plausible reason for this could be the hydrophilic characteristics of components, such as MCPM and Sr/F-BAGs, encouraged water sorption. This may consequently increase the flexibility of the polymer network [46,47]. Additionally, using low molecular weight MMA monomers in EC may result in a polymer with a high cross-linking density, thereby increasing its rigidity [48]. The decreased polymer rigidity due to enhanced water sorption in experimental materials is expected to facilitate ion diffusion from GICs underneath the coating resin [49]. However, this could negatively affect other mechanical and physical characteristics of the restorations, such as increased pigment accumulation, leading to reduced color stability of the materials [11]. Future studies should focus on the long-term mechanical strength, wear resistance, and color stability of these materials.

Previous studies indicated that resin-coating on GICs can act as a physical barrier, thus reducing the diffusion of ions from the GICs to the external environment [15,50]. This aligns with the results of the present study, where the coated groups exhibited a 40–43 % reduction in fluoride release compared to the uncoated group. This study aimed to overcome this limitation by incorporating ion-releasing fillers into the resin coating material. The findings suggest that an increase in the concentration of Ca/Sr (MCPM and Sr/F-BAGs) effectively enhanced fluoride release for the experimental materials. The observed effects might be attributed to multiple factors. For instance, the bioactive glass could release fluoride when it reacts with moisture. Additionally, MCPM might promote the hygroscopic expansion of the polymer network, thereby facilitating ion diffusion from both the resin coating and the underlying GIC. Moreover, the dissolution of MCPM could increase the release of phosphate ions [16,51], which may explain the increase in levels of P release observed with rising Ca/Sr concentrations.

The experimental formulations investigated in the current study pave the way for selecting suitable candidates for more extensive *in vitro* or *in vivo* testing. It was observed that the effect of pre-reacted glass ionomer fillers (SPG) was relatively minimal, possibly due to their lower solubility compared with other fillers. Consequently, based on the results of this study, formulations F1 or F2, which contain high levels of reactive fillers, may be more favorable. Increasing the concentration of these additives improved ion release with minimal impact on the material's physical and mechanical properties. Such properties are anticipated to benefit coated GICs, maintaining their ion release capability. This feature is essential in acidic environments and could play a vital role in preventing secondary caries [52]. This should be tested in the future work.

The current study has several limitations that need to be acknowledged. Firstly, being an *in vitro* study, its clinical relevance needs cautious interpretation. The considerable thickness of the experimental resin coating might also be seen as a drawback. Moreover, the influence of additives on the rheological properties of the resin coating needs further investigation to confirm adequate flowability and surface wetting on the GIC. Additionally, wear resistance and fatigue tests are necessary to verify the resin coating's ability to protect the surface and prevent cracking. Finally, the antibacterial properties of the additives within the resin coating remain unexplored and should be examined in future studies.

5. Conclusion

The experimental resin coating containing ion-releasing fillers exhibited physical and mechanical properties in the range of commercial material, except for the surface microhardness. The beneficial effects were detected with increasing Ca/Sr filler concentrations, which enhanced ion release but exhibited negligible effects on physical/mechanical properties. This balance of maintaining key properties while improving ion release highlights the potential of these experimental formulations for further assessment.

Data availability statement

All the relevant data are included in the manuscript and the supplementary document. No separate repository is attached.

Ethic declaration

Review and approval by an ethics committee was not needed for this study because This work does not address the ethical considerations of animal, cell, and human experimentation.

CRedit authorship contribution statement

Piyaphong Panpisut: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Arnit Toneluck:** Project administration, Investigation, Data curation. **Chutikarn Khamsuk:** Project administration, Investigation, Data curation. **Somruethai Channasanon:** Project administration, Investigation, Data curation. **Siriporn Tanodekaew:** Supervision, Resources, Project administration, Funding acquisition, Data curation. **Naruporn Monmaturapoj:** Supervision, Resources, Project administration, Funding acquisition, Data curation. **Parichart Naruphontjirakul:** Supervision, Resources, Project administration, Funding acquisition, Data curation.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Piyaphong Panpisut reports financial support was provided by Thailand Science Research and Innovation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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