Monomer conversion, dimensional stability, biaxial flexural strength, and fluoride release of resin-based restorative material containing alkaline fillers

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The aim of this study was to assess monomer conversion, dimensional stability (mass and volume changes), biaxial flexural strength (BFS), and fluoride release of recently developed resin composites containing alkaline fillers (Cention N; CN) compared with resin-modified glass ionomer cements (RMGICs: Riva LC; RL and Fuji II LC; FL), and conventional composite (Z350). FL showed highest monomer conversion ($88\pm2\%$) followed by RL ($73\pm10\%$), CN ($59\pm2\%$), and Z350 ($50\pm2\%$). RL exhibited highest mass and volume increase (10.22 ± 0.04 wt% and 19.4 ± 0.2 vol%). CN exhibited higher BFS (180 ± 20 MPa) than RMGICs but lower than Z350 (248 ± 27 MPa). The highest cumulative fluoride release at 6 weeks was observed with RL (136 ± 22 ppm) followed by CN (36 ± 4 ppm) and FL (30 ± 3 ppm). CN exhibited monomer conversion higher than the composite. CN also released fluoride in the range of that observed with RMGICs but with higher flexural strength.

Keywords: Monomer conversion, Dimensional stability, Biaxial flexural strength, Fluoride release, Alkaline fillers

INTRODUCTION

Untreated dental caries is the most prevalent chronic disease affecting 2.4 billion adult and 621 million children worldwide¹. In 2014, the cost for management of caries in the US was estimated to be 122 billion USD. The commonly used restorative materials include dental amalgam, resin composites, and glass ionomer cements. Dental amalgam is a cost-effective material but it has been phased down due to the Minamata Convention on Mercury, which is a global treaty to reduce the risk of anthropogenic mercury release to environment². Resin composites showed high mechanical strength³ with superior esthetics compared to dental amalgam. Most of the commercially available composites exhibit no remineralizing effects and require complicated placement techniques. This could increase the risk of bacterial microleakage at the tooth-composite interface leading to secondary caries⁴ which is the major cause of composite restoration failure. A recently published study demonstrated that secondary caries found in 3.6% of all restorations and one fifth of the patients experienced at least one secondary caries lesion⁵. Additionally, the prevalence of secondary caries in composite restorations was almost twice of that in dental amalgam restorations.

GICs exhibit attractive properties such as the ability to release fluoride which could potentially help to promote tooth remineralization and reduce the risk of secondary caries⁶. Resin-modified glass ionomer cements (RMGICs) has been developed to enable the command setting by photo-polymerization in addition to acid-base neutralization reaction⁷. However, it has been demonstrated that the polymerization of methacrylate monomers may be affected by the restricted radicals mobility from cement hardening through acid-base reaction⁸. This may reduce early strength and increase

the risk of toxic monomer release from RMGICs⁹. Additionally, low molecular weight and hydrophilic 2-hydroxyethyl methacrylate (HEMA) is used as base monomer in RMGICs. The high flexibility of HEMA polymer may lead to excessive water sorption¹⁰ and subsequently reduce mechanical properties of the materials¹¹.

Recently, a new dual-cured resin based composite containing alkaline fillers (alkasite) has been introduced (Cention N, CN, Ivoclar Vivadent, Schaan, Liechtenstein). The filler phase of this material contains ion-releasing glass fillers (alkaline filler). The liquid phase consists of dimethacrylate monomers which requires hand mixing with filler phase. This material can be cured by light or chemical activated polymerization to facilitate bulk placement.

High monomer conversion of resin-based restorative materials usually associates with high physical/mechanical properties¹². The volumetric expansion upon water sorption, *i.e.* hygroscopic expansion, could potentially help to compensate polymerization shrinkage and relieve shrinkage stress of resin composites^{13,14}. This may subsequently help to reduce the stress generated at tooth-restoration interface that could lead to gap formation and bacterial microleakage. Additionally, water sorption also enabled the release of active components (drugs/ions) from the reactive fillers^{15,16}. Hence, the water sorption may promote the dissolution and diffusion of fluoride from CN which could potentially help to promote remineralization of tooth structure. The concern was that the absorbed water may cause plasticization of polymer network and detrimentally affect mechanical properties of the material¹⁷.

The aim of this study was therefore to assess monomer conversion, dimensional stability (hygroscopic expansion), biaxial flexural strength, and fluoride release of the recently developed resin composite containing

alkaline fillers compared with RMGICs and conventional resin composite.

MATERIALS AND METHODS

Commercial materials and their compositions used in the study are provided in Table 1^{8,18-20}. Materials that require mixing were prepared according to their manufacturer instructions.

Monomer conversion

An attenuated total reflection-Fourier transform IR (ATR-FTIR, Nicolet i5, Thermo Fisher Scientific, Waltham, MA, USA) was employed to assess degree of monomer conversion (D_c) of all materials. The materials were prepared according to their manufacturer instructions ($n=5$) and were placed in a metal circlip (10 mm in diameter and 1 mm thickness) on the ATR diamond. The specimens were covered by an acetate sheet and light activated by the LED light curing unit (light intensity of 900–1,000 mW/cm², DEEP BLUE, SMACO, Hangzhou Zhejiang, China) on the top surface. FTIR spectra in a region of 700–1,800 cm⁻¹ at the bottom surface of the specimens were recorded for 5 min. D_c of the sample was then calculated using the following equation. The test was performed at room temperature (25±1°C).

$$D_c(\%) = \frac{100(\Delta B_0 - \Delta B_t)}{\Delta B_0} \quad \text{Equation 1}$$

Where ΔB_0 and ΔB_t were the absorbance of the C-O peak (1,320 cm⁻¹) above background level at 1,335 cm⁻¹ initially and after time t^{21} . Peak of C=C stretch (1,637

cm⁻¹) of methacrylate group was not used to calculate degree of monomer conversion because this peak also represents O-H stretch of absorbed water in the setting RMGICs²².

Mass and volume changes

Gravimetric studies were used to assess mass and volume changes of all materials ($n=3$)^{14,21}. Disc specimens were prepared¹⁴. Materials were prepared according to their manufacturer instruction at room temperature (25±1°C). They were placed in a metal circlip (10 mm in diameter and 1 mm in thickness) and covered with an acetate sheet. They were light activated by the LED curing (DEEP BLUE, 900–1,000 mW/cm², SMACO) for 40 s on both top and bottom surfaces and were incubated in an incubator at the controlled temperature of 37°C for 1 h. Then, they were removed, trimmed all excess, and placed in a tube containing 10 mL of deionized water. The tubes were incubated at 37°C for up to 4 weeks. At each time points (0, 2, 6, 24 h, 3, 4, 7 days, and 2–4 weeks), the samples were removed and blotted dry. Mass and volume of specimens were measured using a four-figure balance equipped with a density kit (MS-DNY-43, METTLER TOLEDO, Columbus, OH, USA). The samples were then placed in the same tube. Mass and volume changes upon immersion time were calculated using the following equations.

$$\Delta M(\%) = \frac{100 [M_t - M_0]}{M_0} \quad \text{Equation 2}$$

$$\Delta V(\%) = \frac{100 [V_t - V_0]}{V_0} \quad \text{Equation 3}$$

Table 1 Materials and their compositions used in this study

Materials	Composition	Lot number	Manufacturer
RIVA Light Cure (RMGIC; RL)	Liquid: Poly(acrylic acid), Tartaric acid, 2-Hydroxyethyl methacrylate, Dimethacrylate crosslinker, Acidic monomer Powder: fluoroalumino silicate glass Recommended powder to liquid ratio: 3:1 (mass ratio)	1096490 (shade A3)	SDI, Bayswater, Victoria, Australia
Fuji II LC (RMGIC; FL)	Liquid: Polyacrylic acid, 2-Hydroxyethyl methacrylate (HEMA), Proprietary ingredient, 2,2,4,-Trimethyl hexamethylene decarbonate, Triethylene glycol dimethacrylate Powder: fluoroalumino silicate glass Recommended powder to liquid ratio: 3.2:1(mass ratio)	1511141 (shade A3)	GC, Tokyo, Japan
Cention N (Alkasite; CN)	Liquid: Urethane dimethacrylate, tricyclodecan-dimethanol dimethacrylate, polyethylene glycol dimethacrylate Powder: Inorganic fillers (Ba-Al-Ca-Ba-Al-F silicate glass, Ca-F-silicate glass, YtF3) and customized fillers Recommended powder to liquid ratio: 4.6:1 (mass ratio)	W44058 (shade A2)	Ivoclar Vivadent, Schaan, Liechtenstein
Filtek 350 XT (Resin composite; Z350)	Urethane dimethacrylate, Bis-GMA, PEGDMA, Bis-EMA, TEGDMA, zirconia, silica	N834734 (shade A2)	3M ESPE, St. Pual, MN, USA

Additionally, a modified Fick's equation (equation 4) was employed to calculate a diffusion coefficient (D , cm^2s^{-1}) of water sorption induced mass change.

$$\frac{\Delta M_t}{\Delta M_{t \rightarrow \infty}} = 2\sqrt{\frac{D_t}{\pi d^2}} \quad \text{Equation 4}$$

Where ΔM_t and $\Delta M_{t \rightarrow \infty}$ are mass increase at time t and maximum increase at time infinity, d is sample thickness (m).

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

The ball-on-ring testing jig were used to measured BFS and BFM of the tested materials ($n=8$)²¹. Disc specimens were prepared according to section 2.2 and immersed in a tube containing 10 mL of deionized water. The tubes were incubated at 37°C for 24 h prior the test. Thicknesses of the specimens were measured using a digital vernier calliper and placed on a ring support (8 mm in diameter) of the ball-on-ring testing jig. BFS of the specimens was determined at room temperature ($25 \pm 1^\circ\text{C}$) using mechanical testing frame (AGSX, Shimadzu, Kyoto, Japan) using 500 N load cell with a crosshead speed of 1 mm/min. The load was applied using a ball indenter (4 mm in diameter). BFS (S; Pa) was calculated using the following equation.

$$S = \frac{F}{d^2} \{ (1+\nu) [0.485 \ln(\frac{r}{d}) + 0.52] + 0.48 \} \quad \text{Equation 5}$$

Where F is the load at failure (N), d is the specimen's thickness (m), r is the radius of circular support (m), and ν is Poisson's ratio (0.3). Additionally, BFM was calculated using the following equation.

$$E = \left(\frac{\Delta H}{\Delta W_c} \right) \times \left(\frac{\beta_c d^2}{q^3} \right) \quad \text{Equation 6}$$

Where E is BFM of the specimen (Pa), $\frac{\Delta H}{\Delta W_c}$ is rate of change of load with regards to central deflection or gradient of force *versus* displacement curve (N/m), β_c is center deflection junction (0.5024)²³, q is ratio of support radius to the radius of disc, and ν is Poisson's ratio (0.3).

Fluoride release

Disc specimens were prepared according to section 2.2 and placed in a tube containing 5 mL of deionized water. The tubes were incubated at the controlled temperature of 37°C for up to 6 weeks. At each time point (1–5 days and 1–6 weeks), the storage solution was collected and replaced with a fresh solution. The collected solution was mixed with TISAB III solution (Orion Ionplus, Thermo Fisher Scientific) with a ratio of 1:10 (volume ratio). Fluoride calibration standards (0.1, 1, 10, 100 Fppm) were prepared using a standard fluoride solution (Orion Ionplus, Thermo Fisher Scientific). Fluoride ion concentration in the solution was then measured using fluoride specific ion electrode (Orion Versastar Pro,

Thermo Fisher Scientific). The cumulative fluoride ion release was calculated using the following equation.

$$F_c = \sum_0^t F_t \quad \text{Equation 7}$$

Where F_c is the cumulative fluoride ion release (ppm), F_t is the amount of fluoride (ppm) at time t .

Statistical analysis

Data reported in this study was mean and 95% confidence interval (95%CI). Data was then analyzed using SPSS for Window version 25 (IBM, Armonk, NY, USA). Levene's test was used to assess the homogeneity of variances of results. One-way ANOVA followed by *post-hoc* Tukey multiple comparison was used if variances between materials were equal. Dunnett T3 *post-hoc* multiple comparison was used when the variances were not equal. Additionally, *post hoc* power analysis was performed using G*Power 3.1 Software (University of Dusseldorf, Germany). The results confirmed that the sample size of each test gave power > 0.99 at $\alpha = 0.05$.

RESULTS

Monomer conversion

The highest monomer conversion was observed with FL ($88 \pm 2\%$) followed by RL $73 \pm 10\%$, CN ($59 \pm 2\%$), and Z350 ($50 \pm 2\%$) respectively (Fig. 1). All results are significantly different from each other ($p < 0.05$).

Mass and volume changes

Mass of RL and FL increased linearly with square root of hour for up to 24 h then reached plateau at 10.22 ± 0.04 and 3.96 ± 0.12 wt%, respectively. Mass of Z350 increased slightly to 0.79 ± 0.10 wt% at 1 week then levelling off. Mass of CN increased to 0.07 ± 0.05 wt% at 1 week then gradually decreased and became negative. The average mass change of CN at 2–4 weeks was -0.28 ± 0.05 wt%. Volume changes of RL and FL increased linearly for up

Fig. 1 Monomer conversion after light cured of all materials tested at room temperature (25°C). Error bars are 95%CI ($n=5$).

Fig. 2 A) Mass and B) volume changes plotted against square root of hour of all materials upon immersion in deionized water for up to 4 weeks. C) Diffusion coefficient of water sorption induced mass change of all materials. Error bars are 95%CI ($n=3$).

Fig. 3 A) Examples force *versus* displacement curves of all materials. B) BFS and C) BFM of all materials after immersion in deionized water for 24 h. Error bars are 95%CI ($n=8$).

Fig. 4 A) fluoride release in deionized water for up to 6 weeks. B) Diffusion coefficient of fluoride for up 1 week. C) Cumulative release of fluoride at 6 weeks. Error bars are 95%CI ($n=3$).

to 1 week then levelling off. The average volume change at 2–4 weeks of RL and FL were 19.4 ± 0.2 and 4.5 ± 0.3 vol%, respectively. The average volume change at late time of Z350 and CN were 2.0 ± 0.1 and 0.8 ± 0.5 vol%, respectively.

The diffusion coefficient of water sorption induced mass change observed with RL ($5.5 \times 10^{-7} \pm 1.2 \times 10^{-11}$ cm²/s) was comparable with that of Fuji II LC ($5.5 \times 10^{-7} \pm 1.1 \times 10^{-11}$ cm²/s) ($p=0.275$). The diffusion coefficient of both RL and FL were however significantly higher than that of CN ($1.1 \times 10^{-7} \pm 1.5 \times 10^{-8}$ cm²/s) and Z350 ($1.5 \times 10^{-8} \pm 2.6 \times 10^{-9}$ cm²/s) ($p<0.05$) (Fig. 2C).

BFS and BFM

All materials except for RL showed a rapid increase of applied force then reached maximum or breaking point within the displacement range of 0.3 mm (Fig. 3A). For RL, the maximum force was maintained for up to displacement of 0.6 mm before reaching the breaking point. The highest BFS was observed with Z350 (248 ± 27 MPa) followed by CN (180 ± 20 MPa), FL (122 ± 6 MPa), and RL (43 ± 9 MPa) (Fig. 3B). The BFM of CN (4.5 ± 0.3 GPa), FL (4.1 ± 0.6 GPa), and Z350 (4.0 ± 0.3 GPa) were comparable ($p>0.05$, Fig. 3C). BFM of these three materials were significantly higher than that of RL (1.5 ± 0.6 GPa, $p<0.05$).

Fluoride release

Fluoride ions was not detected from the storage solution of Z350. At 1 day, RL, FL, and CN exhibited a burst

fluoride release of 25.4 ± 3.1 , 4.3 ± 0.6 , and 4.4 ± 0.8 ppm, respectively (Fig. 4A). Fluoride release of all materials, except for CN, increased linearly with square root of time (h) up to the end of measurement (6 weeks). Fluoride release of CN, however, began to level off at 4 weeks. The highest diffusion coefficient of fluoride was observed with CN ($2.6 \times 10^{-9} \pm 9.8 \times 10^{-11}$ cm²/s) followed by FL ($2.5 \times 10^{-9} \pm 4.9 \times 10^{-10}$ cm²/s) and RL ($1.8 \times 10^{-9} \pm 3.3 \times 10^{-11}$ cm²/s) (Fig. 4B). The cumulative fluoride release at 6 weeks of RL (137 ± 22 ppm) was significantly higher than that of CN (37 ± 4 ppm, $p=0.024$) and FL (30 ± 3 ppm, $p=0.021$) (Fig. 4C).

DISCUSSION

Monomer conversion

The degree of monomer conversion of polymer-based filling materials was primarily governed by chemical structure of monomers²⁴, polymerization environments²⁵, and thickness of the specimens²⁶. High monomer conversion of resin-based restorative materials is required to ensure good physical properties and cytocompatibility of the materials^{27,28}. The minimum degree of monomer conversion required from resin-based materials is not yet established. It is however demonstrated that composites with the conversion lower than 55–65% exhibited low abrasive resistance²⁹. It is also proposed that composites with monomer conversion lower than 55% may not suitable for occlusal restorative layers³⁰.

The monomer conversion of FL, CN, and Z350 obtained from the current study were comparable with that reported in published studies^{22,31}. In general, monomers with lower glass transition temperature (T_g) showed higher degree of monomer conversion than monomers with higher T_g ²⁴. The primary base monomer contained in FL and RL is 2-hydroxyethyl methacrylate (HEMA, $T_g = -60^\circ\text{C}$)^{24,32}. For CN and Z350, their primary base monomer is urethane dimethacrylate (UDMA, $T_g = -35^\circ\text{C}$) and bisphenol A-glycidyl methacrylate (Bis-GMA, $T_g = -7.7^\circ\text{C}$) respectively. Hence, high monomer conversion was observed with FL and RL which was greater than that of CN and Z350. Furthermore, the recommended powder to liquid ratio (PLR) of CN (PLR 4.6:1) was higher than that of RL (PLR 3:1) and FL (PLR 3.2:1). The high PLR may lead to high viscosity of the polymerizing mixture of CN. This may limit the mobility of free radicals which could subsequently reduce the rate of polymerization of the material²⁵.

The current study measured monomer conversion after light curing for 5 min to represent the level of monomer conversion initially after placement. It is however known that degree of monomer conversion can be increased slightly until 24 h³³. It is therefore expected that final monomer conversion of all products can potentially increase to enhance their physical and mechanical properties.

Mass and volume changes

Water are required for acid-base neutralization of GICs setting reaction. Furthermore, the absorbed water also enables release of ions from hydrophilic/reactive fillers. The water sorption induced expansion of polymer-based materials could be governed primarily by the flexibility of polymer matrix and the addition of hydrophilic components^{14,16,21}.

Low diffusion coefficient of water sorption induced expansion observed in CN and Z350 could be due to the high rigidity of UDMA and Bis-GMA polymer network in both CN and Z350 which may limit water sorption resulting in the low mass and volume changes²¹. High mass and volume increase observed with RMGICs (RL and FL) could be due primarily to their hydrophilic and flexible HEMA polymer and polyacrylic salts network facilitating diffusion of water into materials³⁴.

Excessive water sorption may cause the decline in mechanical properties of restorative materials¹⁷. It is however demonstrated that water sorption induced volume expansion could potentially help to compensate polymerization shrinkage and relieve shrinkage stress¹³. Mass and volume increase observed with Z350 and CN were in the range of results obtained from conventional resin composites in published studies (~1 wt% and ~2 vol%)^{26,35}. However, the loss of mass was observed with CN which could be due to the release of soluble species from reactive fillers. The similar result was obtained in the experimental resin composites containing soluble reactive fillers^{21,36}.

BFS and modulus of elasticity

Restorative materials should exhibit high flexural strength to ensure that the materials can function under occlusal loads³⁷. It has been demonstrated that BFS test yielded comparable strength to three-point bending test but the result can be reproducible³⁸. The result from the current study indicated that only Z350 and CN exhibited flexural strength greater than that required from the ISO 4049 (80 MPa)³⁹. The flexural strength of RMGICs (RL and FL) was also greater than 25 MPa required by the ISO 9917-2:2017 Dentistry-Water-based cements (Part 2: Resin-modified cements)⁴⁰. RL exhibited lower strength than FL which could be due to the highest hygroscopic expansion. This may increase solubility and subsequently reduce strength of the materials. The flexural strength and flexural modulus of CN also in the range of values obtained from the hybrid resin composites (100–161 MPa and 6–10 GPa)^{26,41}.

Lowest BFS was observed with RL. The force-displacement curve of RL revealed the less brittle and more plastic behaviour compared with other materials. This could be due to high water sorption induce plasticization of resin matrix¹⁷. This could detrimentally affect mechanical properties of the materials.

FL exhibited comparable modulus to CN and Z350 but the strength of FL was lower than both CN and Z350. The possible explanation could be that the low level of water sorption induced hygroscopic expansion observed in FL led to the low solubility of specimens and the incomplete acid-base reaction. This may increase the amount of the unreacted glass fillers that act as the reinforcing phase in GICs. This partially reacted fillers may therefore attribute to the high stiffness of FL which was in the range of that obtained from CN and Z350. However, CN is resin-based composite containing high T_g monomers that provide high flexural strength such as Bis-GMA or UDMA reinforced with glass fillers^{12,42}. The strength of CN was therefore expected to be in the range of that observed with resin composites which was usually higher than that of RMGICs⁹. Z350 exhibited the highest flexural strength as was expected due primarily to the use of high T_g monomers dispersed with high nanofiller contents in the material. The presence of these closely spaced disperse fillers may increase the strength and fracture energy of resin composites⁴³. Furthermore, CN contains alkaline filler (Ca-F-Si glass fillers) which may react with water and release ions from the material. The dissolution of the fillers may consequently reduce mechanical properties of CN.

Fluoride release

It is expected that the released fluoride ions may help to reduce the risk of tooth demineralization by promoting the formation of low soluble fluorohydroxyapatite⁴⁴. Additionally, fluoride can inhibit bacterial metabolism which may help to reduce bacterial growth⁴⁵. Diffusion-controlled release of active ingredients is usually observed in polymer-based restorative materials⁷. The current study showed that fluoride release in deionized water of all materials except for Z350 increased linearly

with square root of time indicating a diffusion-controlled release. This can be demonstrated by the modified Fickian equation.

$$\Delta F = \Delta F_0 + \Delta F_{\infty} \sqrt{\frac{2D_t}{\pi d^2}} \quad \text{Equation 8}$$

Where ΔF ; the change in cumulative fluoride in the solution, ΔF_0 ; early burst release, ΔF_{∞} ; maximum change in the solution, D ; fluoride diffusion coefficient, t ; time, d ; sample thickness. Fluoride release from GICs is mainly controlled by the composition of materials, powder to liquid ratio, solubility of glass particles, rate of acid neutralization, water sorption, and the initial concentration of fluoride that contained in the materials²²⁾.

The possible explanation of high concentration of fluoride release observed with RL could be high water sorption promoting acid-neutralization reaction and release fluoride ion from fluoro-alumino-silicate glass. This high level of fluoride release from RL could be of benefit to promote remineralization and the formation of low soluble fluorohydroxyapatite in tooth surface. This could potentially help to reduce the risk of tooth demineralization due to bacterial microleakage leading to secondary caries, which is the major cause of composite restoration failure⁴⁶⁾. CN showed comparable fluoride diffusion coefficient and releasing profile to FL. This could be due to the low monomer conversion and low crosslinking density in the polymer network of CN which may facilitate the diffusion of fluoride. Additionally, the mixing of CN required higher PLR than that of RL and FL. This high PLR may increase the risk of air entrapment during mixing. The pores generated due to air incorporation could also facilitate the diffusion of fluoride^{47,48)}.

In summary, RMGICs (RL,FL) showed higher monomer conversion than Z350 and CN due to the use of low molecular weight monomer (HEMA) as the primary base monomer. The hydrophilicity of HEMA polymer may increase water sorption to plasticize resin matrix thereby reducing strength of the materials. This may increase the risk of restoration fracture in load-bearing areas which was the main cause of GICs restoration failure⁴⁹⁾. The flexural strength of CN and FL were lower than that of Z350 but still in the acceptable range of the ISO standard for resin composite. CN and FL can, however, exhibit fluoride release which could potentially help to reduce the risk of secondary caries.

CONCLUSION

Monomer conversion of CN was higher than that of Z350 but lower than that of FL and RL. CN and Z350 showed lower mass and volume changes (good dimensional stability) and higher BFS than FL and RL. However, CN exhibited lower BFS than Z350 but can release fluoride in the range of that observed with FL.

ACKNOWLEDGMENTS

The authors would like to express their heartfelt gratitude to Faculty of Dentistry, Thammasat University for providing the grant to conduct this research (grant ID: 1/2560).

CONFLICTS OF INTEREST

The authors report no conflicts of interest.

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