ARTICLE IN PRESS
+ MODEL
Journal of Dental Sciences xxx (xxxx) xxx

Available online at www.sciencedirect.com
ScienceDirect
journal homepage: www.e-jds.com

Journal of Dental Sciences

Original Article

Polymerization shrinkage and shrinkage stress of bulk-fill and non-bulk-fill resin-based composites

Peng Yu [a,b], Yong-Xiang Xu [b,c], Yu-Shu Liu [b,d]*

[a] Department of Cariology and Endodontology, Peking University School and Hospital of Stomatology, Beijing, China
[b] National Clinical Research Center for Oral Diseases & National Engineering Laboratory for Digital and Material Technology of Stomatology & Beijing Key Laboratory of Digital Stomatology, Beijing, China
[c] Department of Dental Materials, Peking University School and Hospital of Stomatology, Beijing, China
[d] Second Dental Clinical Division, Peking University School and Hospital of Stomatology, Beijing, China

Received 6 November 2021; Final revision received 3 December 2021
Available online ■ ■ ■

KEYWORDS
Bulk-fill;
Resin-based composite;
Polymerization shrinkage;
Shrinkage stress

Abstract *Background/purpose:* Bulk-fill resin-based composites (RBCs) are a new class of restorative materials, and polymerization shrinkage (PS) is concerned due to their single increment up to 4 mm. The aim of this study was to evaluate the PS and shrinkage stress (SS) of bulk-fill RBCs in vitro.
Materials and methods: Three bulk-fill RBCs and three conventional non-bulk-fill RBCs were selected. The PS was determined with Acuvol volumetric shrinkage analyzer by calculating the specimen volume variation before and after light irradiation. The SS was investigated using universal testing machine method with a polymethyl methacrylate rod as a bonding substrate. The force generated during the polymerization process was detected by a load cell linked to a computer. SS was calculated by dividing the maximum stress force by the area of the rod.
Results: The mean PS of various RBCs ranged from 1.72% to 2.13%. All PS results of bulk-fill RBCs were comparable to their conventional counterparts. Sonicfill 2 (SF2) and Harmonize (HM) showed the lowest PS ($p < 0.05$; Tukey HSD test). Medians of SS results ranged from 0.55 MPa to 0.67 MPa. All SSs of bulk-fill RBCs were comparable to their conventional counterparts. SF2 showed significantly lower SS than Tetric N-Ceram (TN) and Tetric N-Ceram Bulk Fill (TNB) ($p < 0.0083$; post hoc comparisons with Bonferroni adjustments). A moderate, positive correlation was observed between PS and SS with Pearson's correlation ($r = 0.446$, $p = 0.013$).

* Corresponding author. Second Dental Clinical Division, Peking University School and Hospital of Stomatology, No.22, Zhongguancun South Avenue, Haidian District, Beijing, 100081, PR China. Fax: +86 010 82195385.
E-mail address: roywillow@163.com (Y.-S. Liu).

https://doi.org/10.1016/j.jds.2021.12.004

Please cite this article as: P. Yu, Y.-X. Xu and Y.-S. Liu, Polymerization shrinkage and shrinkage stress of bulk-fill and non-bulk-fill resin-based composites, Journal of Dental Sciences, https://doi.org/10.1016/j.jds.2021.12.004

Conclusion: Both PS and SS are material dependent. A moderate, positive correlation between PS and SS is found with new bulk-fill RBCs and their conventional counterparts.

Introduction

Resin-based composites (RBCs) have gained popularity as restorative materials due to their aesthetic properties, high strength and technical insensitivity. When the resin-based composites are light irradiated and light cured, the van der Waals spaces between methacrylate monomers convert to shorter covalent bonds. The volume of resin-based composites shrinks, and stress arises at the bonding interface.[1] Many clinical problems, such as reduced bonding strength, cuspal deflection, enamel cracking, marginal leakage, postoperative dentine hypersensitivity and secondary caries, are believed to be related to shrinkage stress (SS).[2–5]

To reduce the influence of polymerization shrinkage (PS) of RBCs during clinical application, the incremental filling technique is proposed by reducing the C-factor of each layer.[6] Conflicted results on the incremental filling technique have been reported.[7–9] In addition, the incremental filling technique has other drawbacks, including technical sensitivity and time consumption.[10]

Recently, a new class of RBCs called bulk-fill resin composites has been launched on the market. Claiming a single increment of up to 4 mm, bulk-fill RBCs aim to make clinical procedures simpler and more efficient, as well as fewer technical errors such as voids and impurities between RBC layers.[11] When applied as a large volume, concerns about the PS of bulk-fill RBCs arise. To address this problem, new monomer technologies are employed in bulk-fill RBCs. Urethane dimethacrylate (UDMA) is aromatized to decrease the volume shrinkage.[12] Another modification with UDMA is increasing the molecular weight. A larger molecule means a less reactive aliphatic C=C group and thus less shrinkage. Meanwhile, due to the conformational flexibility of UDMA, the gel point is delayed, and more stress can be released.[13] Moreover, a new addition–fragmentation chain transfer monomer (AFM) is introduced into methacrylate-based composites. It rearranges the polymer network and delays the gel point to release SS.[14] Concerning the changes in the chemical reaction kinetics of new bulk-fill RBCs, investigations on the PS and SS of bulk-fill RBCs are needed before their wide clinical use.

The purpose of this study was to evaluate the PS and SS of bulk-fill RBCs after light irradiation. The null hypotheses were as follows: there were no significant differences in a) PS and b) SS between contemporary bulk-fill and non-bulk-fill RBCs.

Materials and methods

Six RBCs were selected, including three bulk-fill (Sonicfill 2 [SF2], Tetric N-Ceram Bulk Fill [TNB], Filtek Bulk Fill Posterior Restorative [FTB]) and three conventional non-bulk-fill (Harmonize [HM], Tetric N-Ceram [TN], Filtek Z350 XT [FZX]) RBCs. Detailed information on the products is listed in Table 1.

Polymerization shrinkage

Polymerization shrinkage (PS) ($n = 5$) was investigated with a video imaging device (Acuvol Volumetric Shrinkage Analyzer; Bisco Inc, Schaumburg, IL, USA). Each RBC specimen was shaped into a hemisphere and placed on the pedestal in front of the CCD camera. Volume before curing was recorded as V_1. Then, a curing unit (BluePhase; Ivoclar Vivadent, Shaan, Liechtenstein) with the tip close to the surface of the specimen was activated to irradiate the specimens for 20 s. The postcuring volume after 5 min was recorded as V_2. The PS was subsequently calculated as follows:

$$PS = (V_1 - V_2) / V_1$$

Shrinkage stress

Universal testing machine method was used to investigate the shrinkage stress (SS) ($n = 5$). A custom-made accessory is shown in Fig. 1. The upper part of the accessory was

Table 1 Information of materials investigated in this study.

Products	Abbreviation	Filler load (wt%/vol%)	Manufacture	Shade	Lot number
Sonicfill 2	SF2	81.3/65.5	Kerr	A1	6842322
Harmonize	HM	81/64.5	Kerr	A3	6901692
Tetric N-Ceram Bulk Fill	TNB	80-81/55-57	Ivoclar Vivadent	IVA	V34862
Tetric N-Ceram	TN	75-77/53-55	Ivoclar Vivadent	A2	Y10638
Filtek Bulk Fill Posterior Restorative	FTB	76.5/58.4	3M ESPE	A2	N897361
Filtek Z350 XT	FZX	78.5/63.3	3M ESPE	A2	NA89335

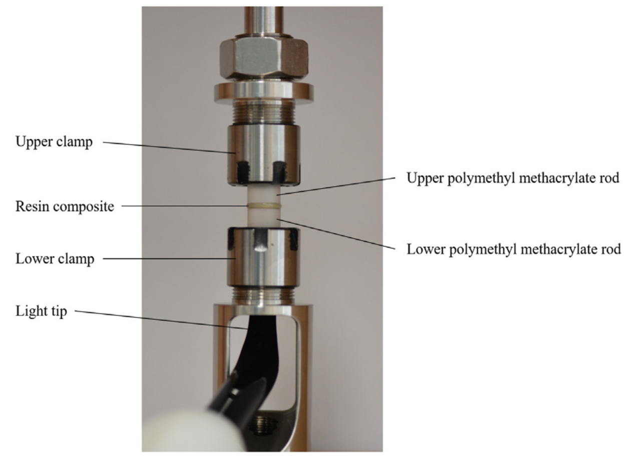

Figure 1 Diagram of the measuring accessory for shrinkage force.

connected to a load cell (500 N Static Load Cell, Instron, Norwood, MA, USA), to which a polymethyl methacrylate rod (5 mm in diameter and 28 mm in length) was screwed. The lower part of the accessory was fixed on a universal testing machine (Instron 5969, Norwood, MA, USA), and another highly transparent polymethyl methacrylate rod (5 mm in diameter and 13 mm in length) was clamped. The bottom surface of the longer rod and the top surface of the shorter rod were roughened with 600-grit silicon carbide paper and treated with a bonding resin (Clearfil SE Bond bond, Kuraray Noritake, Tokyo, Japan). Light irradiation was applied for 10 s for each surface using a curing unit (BluePhase). The distance between two treated surfaces was 1 mm. This space allowed the insertion of RBC materials (19.6 mm^3) with a resulting C factor of 2.5. The curing unit (BluePhase) with an 8 mm diameter tip was positioned closely to the bottom surface of the shorter rod and activated for 20 s. The output of the curing unit itself was 950 mW/cm^3, while that through the 13 mm rod was 570 mW/cm^3, as calibrated by an LED light tester (FB-M2000A, Fibop Medical Instrument, Foshan, China). Specimen height was kept constant with the use of a non-contacting video extensometer (AVE, Instron, Norwood, MA, USA). The forces generated during the polymerization process were detected by means of the load cell linked to an attached computer. Data were recorded 5 min after light irradiation, and the maximum polymerization stress was calculated by dividing the maximum stress force by the cross-sectional area of the rod.

Statistical analysis

Statistical analysis was carried out using SPSS 22.0 (IBM SPSS Inc., Chicago, IL, USA). Before testing the significant differences between each group, the Shapiro–Wilk test and Levene test were performed to test the normality and equality of variance, respectively. PS data were subjected to one-way analysis of variance and Tukey HSD test at a significance level of 0.05. A Kruskal–Wallis nonparametric analysis of variance was performed followed by Mann–Whitney post hoc comparisons with Bonferroni adjustments to test the differences in medians between SS data of different material groups, and the significance level was adjusted to $p < 0.0083$. The relationship between PS and SS was analyzed with Pearson's correlation at a significance level of 0.05.

Results

The PS results are presented in Table 2 and Fig. 2. Mean PS of various RBCs ranged from 1.72% to 2.13%. The ranking of PS from highest to lowest was as follows: FTB = TNB = TN = FZX > SF2 = HM; ($p < 0.05$; Tukey HSD test). All PS results of bulk-fill RBCs were comparable to their conventional counterparts. The PS of SF2 and HM was found to be significantly lower than that of the other bulk-fill and non-bulk-fill groups.

SS data is shown in Table 2 and Fig. 3. Medians of SS results ranged from 0.55 MPa to 0.67 MPa. All SS results of bulk-fill RBCs were comparable to their conventional counterparts. SF2 showed the lowest SS and was significantly lower than TN and TNB.

A moderate, positive correlation was observed between PS and SS with Pearson's correlation ($r = 0.446$, $p = 0.013$).

Discussion

The study investigated the PS and SS of bulk-fill and non-bulk-fill resin-based composites. Based on the results, the null hypotheses were rejected.

A variety of techniques, including water/mercury dilatometry, the bonded-disc method, cuspal deflection, specific gravity analysis, electrical strain gauges, micro-CT and optical measurements, have been employed to determine the PS of RBCs.[15,16] Among those, the Acuvol video-imaging technique was an easy way to give precise and reproducible results.[17] The mean PS for the bulk-fill RBCs evaluated ranged from 1.85% to 2.13%, while that of non-bulk-fill RBCs ranged from 1.72% to 2.08%. Significantly lower PS was observed in SF2 and HM than in TNB, TN, FTB and FZX. The difference can be attributed to larger filler proportions of SF2 (81.3 wt%/65.5 vol%) and HM (81 wt%/64.5 vol%). A larger filler proportion indicates

Table 2 Means and standard deviation (SD) of PS and medians and interquartile range (IQR) of SS for various RBCs.

Materials	PS/%	SS/MPa
SF2	1.85 ± 0.06^{A}	0.55 ± 0.055^{A}
HM	1.72 ± 0.09^{A}	0.59 ± 0.100^{AB}
TNB	2.11 ± 0.07^{B}	0.65 ± 0.020^{B}
TN	2.08 ± 0.05^{B}	0.67 ± 0.015^{B}
FTB	2.13 ± 0.04^{B}	0.63 ± 0.035^{AB}
FZX	2.03 ± 0.16^{B}	0.58 ± 0.030^{AB}

*Different uppercase letters in each row indicate significant difference between materials for PS ($p < 0.05$; Tukey HSD test) and for SS ($p < 0.0083$; Mann–Whitney post hoc comparisons with Bonferroni adjustments). SF2 (Sonicfill 2), HM (Harmonize), TNB (Tetric N-Ceram Bulk Fill), TN (Tetric N-Ceram), FTB (Filtek Bulk Fill Posterior Restorative), FZX (Filtek Z350 XT).

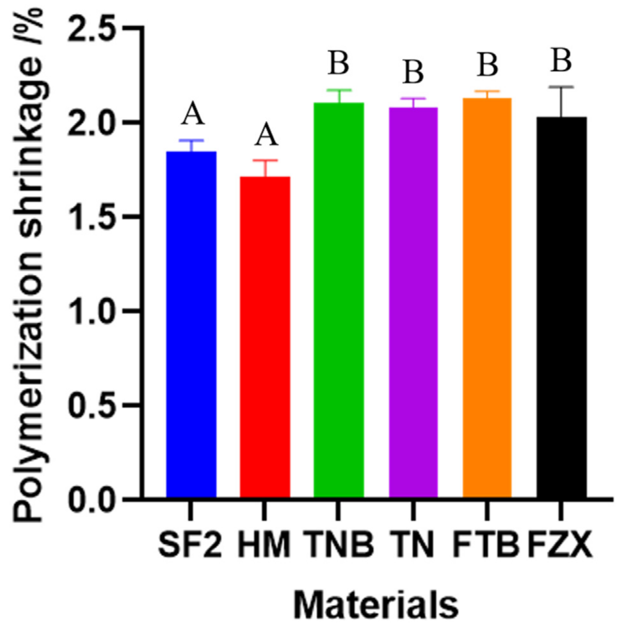

Figure 2 Polymerization shrinkage of RBC materials. Different uppercase letters over the bar indicate significant difference between materials ($p < 0.05$; Tukey HSD test).

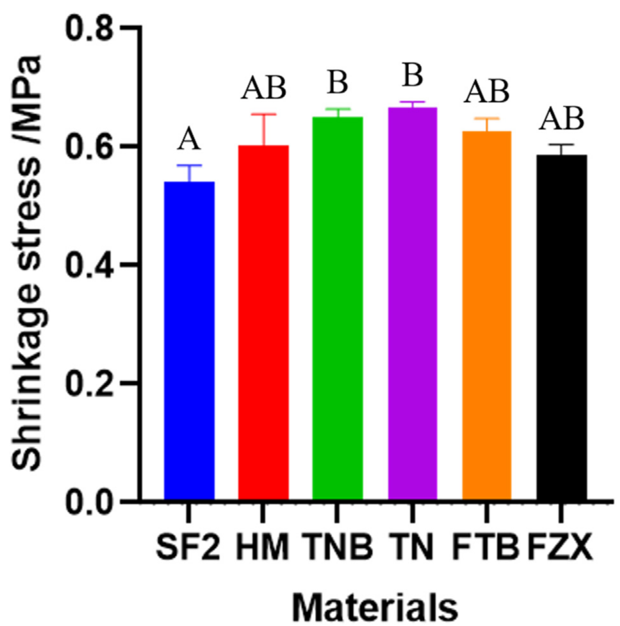

Figure 3 Shrinkage stress of RBC materials. Different uppercase letters over the bar indicate significant difference between materials ($p < 0.0083$; Mann–Whitney post hoc).

less conversion of active organic C=C into C–C at a given volume, resulting in less shrinkage.[18] Although an increased filler proportion was observed in TNB compared with TN, no significant difference was found concerning PS. The most likely reason was the improvement in the degree of conversion of TNB, which was found to have a moderate, positive and significant correlation in a previous study.[17] This was achieved by the addition of the creative photoinitiator Ivocerin, a dibenzoyl germanium derivative.[19] On the other hand, a decrease in the filler proportion of FTB did not result in a significant increase in PS compared with FZX. Shrinkage of FTB may be partially compensated by new monomers such as aromatic urethane dimethacrylate (AUDMA), which can constrain shrinkage during polymerization reactions.[12]

Different from PS, SS is not a material property. Multiple factors, including the PS and elastic modulus of RBC materials, degree of monomer conversion, configuration of the cavity, chemical reaction kinetics and clinical handling technique, influence SS.[20] It is difficult to identify the effect on SS of an individual factor, since most of them are interrelated. However, large quantities of laboratory research have shown evidence of RBC materials generating SS, and there is consensus that SS is clinically relevant.[21] Direct investigation of SS of RBCs can guide clinical selection and further improvement of materials.

To measure the SS, universal testing machine method was utilized in this study. It is the most widely used method to directly quantize SS.[21] Medians of SS of all RBCs evaluated ranged from 0.55 MPa to 0.67 MPa. SS of SF2 was significantly lower than both TN and TNB. A previous study showed that high inorganic contents were associated with low SS due to reduced PS with a similar instrumental design.[22] The filler proportion of SF2 was highest in volume, while that of TN and TNB was the lowest among all RBCs tested. Meanwhile, a low filler proportion was also observed in FTB, but the SS value of FTB was comparable to those of the other groups. New monomers in FTB called addition–fragmentation-chaintransfer monomers (AFMs) alter the polymerization kinetics by decelerating the reaction rate.[23] AFM can rearrange polymer networks by breaking and reforming covalent bonds. The gel point was therefore delayed, and SS was partially released. However, a limited amount of AFM is added into FTB as a modulator, causing more than 5 wt% AFM to significantly reduce the degree of monomer conversion and decrease the postcure physico-mechanical properties.[14]

A less intensive correlation between PS and SS was found in our study (Pearson's correlation $r = 0.446$, $p = 0.013$) than in previous studies based on traditional RBC materials.[24–27] A possible explanation is that SS is affected by multiple factors, the major of which is that polymerization kinetics have changed with innovations in new monomers, new photoinitiators and new modulators in recent years.[28,29] Further studies investigating PS and SS with newly marketing bulk-fill RBCs and the related factors, including elastic modulus, degree of conversion, and chemical reaction kinetics, are needed.

Under the limitations of the present study, it could be concluded that both PS and SS are material dependent. A moderate, positive correlation between PS and SS is found with new bulk-fill RBCs and their conventional counterparts.

Declaration of competing interest

The authors deny any conflicts of interest related to the present study.

Acknowledgments

This study was supported by the Chinese Stomatological Association Youth Founding (No. CSA-R2018-03) and Clinical Medicine Plus X - Young Scholars Project, Peking University, the Fundamental Research Funds for the Central Universities.

References

1. Meereis CTW, Münchow EA, de Oliveira da Rosa WL, da Silva AF, Piva E. Polymerization shrinkage stress of resin-based dental materials: a systematic review and meta-analyses of composition strategies. *J Mech Behav Biomed Mater* 2018;82:268—81.
2. Cerda-Rizo ER, de Paula Rodrigues M, Vilela A, et al. Bonding interaction and shrinkage stress of low-viscosity bulk fill resin composites with high-viscosity bulk fill or conventional resin composites. *Operat Dent* 2019;44:625—36.
3. Nakano EL, de Souza A, Boaro L, Catalani LH, Braga RR, Gonçalves F. Polymerization stress and gap formation of self-adhesive, bulk-fill and flowable composite resins. *Operat Dent* 2020;45:E308—16.
4. Kim RJ, Kim YJ, Choi NS, Lee IB. Polymerization shrinkage, modulus, and shrinkage stress related to tooth-restoration interfacial debonding in bulk-fill composites. *J Dent* 2015;43: 430—9.
5. Han SH, Sadr A, Shimada Y, Tagami J, Park SH. Internal adaptation of composite restorations with or without an intermediate layer: effect of polymerization shrinkage parameters of the layer material. *J Dent* 2019;80:41—8.
6. Münchow EA, Meereis CTW, de Oliveira da Rosa WL, da Silva AF, Piva E. Polymerization shrinkage stress of resin-based dental materials: a systematic review and meta-analyses of technique protocol and photo-activation strategies. *J Mech Behav Biomed Mater* 2018;82:77—86.
7. Soares CJ, Bicalho AA, Tantbirojn D, Versluis A. Polymerization shrinkage stresses in a premolar restored with different composite resins and different incremental techniques. *J Adhesive Dent* 2013;15:341—50.
8. Loguercio AD, Reis A, Ballester RY. Polymerization shrinkage: effects of constraint and filling technique in composite restorations. *Dent Mater* 2004;20:236—43.
9. Correia AMO, Tribst JPM, Matos FS, Platt JA, Caneppele TMF, Borges ALS. Polymerization shrinkage stresses in different restorative techniques for non-carious cervical lesions. *J Dent* 2018;76:68—74.
10. Furness A, Tadros MY, Looney SW, Rueggeberg FA. Effect of bulk/incremental fill on internal gap formation of bulk-fill composites. *J Dent* 2014;42:439—49.
11. Lima RBW, Troconis CCM, Moreno MBP, Murillo-Gómez F, De Goes MF. Depth of cure of bulk fill resin composites: a systematic review. *J Esthetic Restor Dent* 2018;30:492—501.
12. Moszner N, Fischer UK, Angermann J, Rheinberger V. A partially aromatic urethane dimethacrylate as a new substitute for Bis-GMA in restorative composites. *Dent Mater* 2008;24:694—9.
13. Ilie N, Hickel R. Investigations on a methacrylate-based flowable composite based on the SDR™ technology. *Dent Mater* 2011;27:348—55.
14. Shah PK, Stansbury JW, Bowman CN. Application of an addition-fragmentation-chain transfer monomer in di(meth)acrylate network formation to reduce polymerization shrinkage stress. *Polym Chem* 2017;8:4339—51.
15. Ersen KA, Gürbüz Ö, Özcan M. Evaluation of polymerization shrinkage of bulk-fill resin composites using microcomputed tomography. *Clin Oral Invest* 2020;24:1687—93.
16. Marx P, Wiesbrock F. Expanding monomers as anti-shrinkage additives. *Polymers* 2021;13:806.
17. Yu P, Yap A, Wang XY. Degree of conversion and polymerization shrinkage of bulk-fill resin-based composites. *Operat Dent* 2017;42:82—9.
18. Luo S, Liu F, Yu B, He J. Preparation of low shrinkage stress Bis-GMA free dental resin composites with a synthesized urethane dimethacrylate monomer. *J Biomater Sci Polym Ed* 2019;30: 137—49.
19. Moszner N, Fischer UK, Ganster B, Liska R, Rheinberger V. Benzoyl germanium derivatives as novel visible light photoinitiators for dental materials. *Dent Mater* 2008;24:901—7.
20. Muc A, Romanowicz P, Chwał M. Description of the resin curing process-formulation and optimization. *Polymers* 2019; 11:127.
21. Ferracane JL, Hilton TJ, Stansbury JW, et al. Academy of Dental Materials guidance-Resin composites: Part II-Technique sensitivity (handling, polymerization, dimensional changes). *Dent Mater* 2017;33:1171—91.
22. Gonçalves F, Pfeifer CC, Stansbury JW, Newman SM, Braga RR. Influence of matrix composition on polymerization stress development of experimental composites. *Dent Mater* 2010; 26:697—703.
23. Correia A, Andrade MR, Tribst J, Borges A, Caneppele T. Influence of bulk-fill restoration on polymerization shrinkage stress and marginal gap formation in class V restorations. *Operat Dent* 2020;45:E207—16.
24. Kleverlaan CJ, Feilzer AJ. Polymerization shrinkage and contraction stress of dental resin composites. *Dent Mater* 2005;21:1150—7.
25. Boaro LC, Gonçalves F, Guimarães TC, Ferracane JL, Versluis A, Braga RR. Polymerization stress, shrinkage and elastic modulus of current low-shrinkage restorative composites. *Dent Mater* 2010;26:1144—50.
26. Al Sunbul H, Silikas N, Watts DC. Polymerization shrinkage kinetics and shrinkage-stress in dental resin-composites. *Dent Mater* 2016;32:998—1006.
27. Tauböck TT, Jäger F, Attin T. Polymerization shrinkage and shrinkage force kinetics of high- and low-viscosity dimethacrylate- and ormocer-based bulk-fill resin composites. *Odontology* 2019;107:103—10.
28. Fugolin APP, Pfeifer CS. New resins for dental composites. *J Dent Res* 2017;96:1085—91.
29. Wiesner T, Haas M. Do germanium-based photoinitiators have the potential to replace the well-established acylphosphine oxides? *Dalton Trans* 2021;50:12392—8.