Resin viscosity determines validity of exposure reciprocity law in resin-based dental composites

S. Palagummi[1*], T. Hong[1], Z. Wang[2], M. Y. M. Chiang[1*]

[1]Biosystems and Biomaterials Division, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
[2]Department of Engineering Mechanics, School of Civil Engineering, Wuhan University, Wuhan, Hubei 430072, China
Keywords: Exposure reciprocity law, radiant exposure, polymerization exotherm

In restorative dentistry, the replacement of quartz tungsten halogen with light-emitting diodes (LEDs) light curing units for photocuring dental composites has been a trend due to their narrow bandwidth of operation, lower heat generation, and better power efficiency. These advancements along with the ever-increasing demand from dentists for reduced chair side times has led to high irradiance (e.g., >2 W/cm^2) LED light curing units [1]. The justification of using high irradiance and short curing time springs from an archaic law, borrowed and derived from Bunsen-Roscoe reciprocity law [2] in photochemistry, termed as exposure reciprocity law (ERL). The ERL states that for a given radiant exposure, defined as the total light energy received per unit area (= irradiance × exposure time), the final physical and mechanical properties of the composite do not change with any combination of irradiance and irradiation time. The applicability of ERL has been tested in literature with regards to several properties, some of them are, degree of conversion (DC) [3-7], flexural strength [8-10], flexural modulus [4,10-12] and hardness [11,13,14]. A great number of these studies have been done on commercial materials and the subsequent discussion was based on known partial knowledge of its constituents [5,6,15,16].

Feng et al. [3,7] have stated for a variety of resin mixtures and composites that ERL was valid only if the resin viscosity was above a certain threshold value. When the resin viscosity was below the threshold, the DC achieved at the high irradiance was significantly lesser than that at a lower irradiance for the same radiant exposure. Accordingly, other studies suggested that the ERL would be held given that a minimum irradiation time is allowed when high irradiances are used. Similar conclusions on the minimum irradiation time was reached with experiments on commercial composites in literature [8,13,17]. However, to the best of the authors' knowledge, no effort has been made to estimate this minimum time requirement, in other words, minimum radiant exposure requirement needed for validity of ERL at higher irradiances for different resin mixtures.

Some articles discussed that high irradiance curing of dental resin-based composites may increase polymerization stress (PS) and decrease their bond strength to the tooth [18-20]. An increase in DC rate, as a result of increasing irradiance, will cause an increase in the PS rate but it is speculative as to whether it will lead to an overall higher PS, considering that the radiant exposure is kept a constant [21-26]. Also, the increase in DC rate increases the exothermic temperature rise due to polymerization, this along with the temperature rise due to absorbance form the high irradiance should be of significant concern as they can potentially lead to pulpal and tissue damage. Only a few studies in literature [11,27], have considered the effect of these high irradiance on the temperature change (TC) in the composites.

The current research addresses some of these issues by systematically studying the polymerization of various experimental model composites using a NIST-developed cantilever-beam based instrument [28,29]. The instrument provides real-time, simultaneous measurements of the DC, TC and PS and thereby reduces ambiguity in measurements generally perceived when making such measurements from different yet similar samples [30]. Model dental composites comprising of traditional dimethacrylate monomers, mixed with the visible-light initiator system, camphorquinone and ethyl 4-dimethylaminobenzoate, and commercial silanized micro-sized glass fillers were used to conduct this study. The results show that ERL is valid above a threshold radiant exposure which is defined by a function of the viscosity of the resin of the composite. At this threshold radiance exposure, the PS varied with irradiance for the composites in no particular trend which can be attributed due to a competition between a multitude of factors, whereas, the peak TC, as expected, increased with irradiance.

Acknowledgements

Financial support was provided through an Interagency Agreement between the National Institute of Dental and Craniofacial Research (NIDCR) and NIST [ADE12017-0000]. The donation of the monomers from Esstech Inc. and the filler particles from Dentsply Sirona are greatly appreciated.

References

[1] K.D. Jandt, R.W. Mills, A brief history of LED photopolymerization, Dental Materials. 29 (2013) 605-617.

[2] R. Bunsen W., H. Roscoe E., Photochemical studies, Annals of Physics. (1923) 108-193.

[3] L. Feng, B.I. Suh, Exposure Reciprocity Law in Photopolymerization of Multi-Functional Acrylates and Methacrylates, Macromolecular Chemistry and Physics. 208 (2007) 295-306.

[4] M. Dewaele, E. Asmussen, A. Peutzfeldt, E.C. Munksgaard, A.R. Benetti, G. Finné, G. Leloup, J. Devaux, Influence of curing protocol on selected properties of light-curing polymers: Degree of conversion, volume contraction, elastic modulus, and glass transition temperature, Dental Materials. 25 (2009) 1576-1584.

[5] D. Selig, T. Haenel, B. Hausnerová, B. Moeginger, D. Labrie, B. Sullivan, R.B.T. Price, Examining exposure reciprocity in a resin based composite using high irradiance levels and real-time degree of conversion values, Dental Materials. 31 (2015) 583-593.

[6] M. Hadis, J.G. Leprince, A.C. Shortall, J. Devaux, G. Leloup, W.M. Palin, High irradiance curing and anomalies of exposure reciprocity law in resin-based materials, J.Dent. 39 (2011) 549-557.

[7] L. Feng, R. Carvalho, B.I. Suh, Insufficient cure under the condition of high irradiance and short irradiation time, Dental Materials. 25 (2009) 283-289.

[8] L. Musanje, B. Darvell W., Polymerization of resin composite restorative materials: exposure reciprocity, Dental Materials. 19 (2003) 531-541.

[9] M. Miyazaki, Y. Oshida, B. Keith Moore, H. Onose, Effect of light exposure on fracture toughness and flexural strength of light-cured composites, Dental Materials. 12 (1996) 328-332.

[10] A. Peutzfeldt, E. Asmussen, Resin Composite Properties and Energy Density of Light Cure, J.Dent.Res. 84 (2005) 659-662.

[11] L.D. Randolph, W.M. Palin, D.C. Watts, M. Genet, J. Devaux, G. Leloup, J.G. Leprince, The effect of ultra-fast photopolymerisation of experimental composites on shrinkage stress, network formation and pulpal temperature rise, Dental Materials. 30 (2014) 1280-1289.

[12] F.C. Calheiros, M. Daronch, F.A. Rueggeberg, R.R. Braga, Degree of conversion and mechanical properties of a BisGMA:TEGDMA composite as a function of the applied radiant exposure, Journal of Biomedical Materials Research Part B: Applied Biomaterials. 84B (2008) 503-509.

[13] A. Peutzfeldt, A. Lussi, S. Flury, Effect of High-Irradiance Light-Curing on Micromechanical Properties of Resin Cements, BioMed Research International. 2016 (2016).

[14] R.L. Erickson, W.W. Barkmeier, R.H. Halvorson, Curing characteristics of a composite – Part 1: Cure depth relationship to conversion, hardness and radiant exposure, Dental Materials. 30 (2014) e125-e133.

[15] M.M. AlShaafi, Effects of delivering the same radiant exposures at 730, 1450, and 2920 mW/cm2 to two resin-based composites, European Journal of Dentistry. 11 (2017) 22-28.

[16] R.H. Halvorson, R.L. Erickson, C.L. Davidson, Energy dependent polymerization of resin-based composite, Dental Materials. 18 (2002) 463-469.

[17] J.G. Leprince, M. Hadis, A.C. Shortall, J.L. Ferracane, J. Devaux, G. Leloup, W.M. Palin, Photoinitiator type and applicability of exposure reciprocity law in filled and unfilled photoactive resins, Dental Materials. 27 (2011) 157-164.

[18] R.B. Price, A.C. Shortall, W.M. Palin, Contemporary Issues in Light Curing, Oper.Dent. 39 (2014) 4-14.

[19] L. Feng, B.I. Suh, A mechanism on why slower polymerization of a dental composite produces lower contraction stress, Journal of Biomedical Materials Research Part B: Applied Biomaterials. 78B (2006) 63-69.

[20] G.L. Unterbrink, R. Muessner, Influence of light intensity on two restorative systems, Journal of Dentistry. 23 (1995) 183-189.

[21] J. Wydra W, N. Cramer B., J. Stansbury W., C. Bowman N., The reciprocity law concerning light dose–relationships applied to BisGMA/TEGDMA photopolymers: Theoretical analysis and experimental characterization, Dent. Mater. 30 (2014) 605-612.

[22] E. Asmussen, A. Peutzfeldt, Polymerization contraction of resin composite vs. energy and power density of light-cure, Eur.J.Oral Sci. 113 (2005) 417-421.

[23] R.R. Braga, J.L. Ferracane, Contraction Stress Related to Degree of Conversion and Reaction Kinetics, J.Dent.Res. 81 (2002) 114-118.

[24] R.L. Sakaguchi, B.D. Wiltbank, C.F. Murchison, Contraction force rate of polymer composites is linearly correlated with irradiance, Dental Materials. 20 (2004) 402-407.

[25] H.-. Bang, B.-. Lim, T.-. Yoon, Y.-. Lee, C.-. Kim, Effect of plasma arc curing on polymerization shrinkage of orthodontic adhesive resins, J.Oral Rehabil. 31 (2004) 803-810.

[26] N. Emami, K.M. Söderholm, L.A. Berglund, Effect of light power density variations on bulk curing properties of dental composites, Journal of Dentistry. 31 (2003) 189-196.
[27] E. Armellin, G. Bovesecchi, P. Coppa, G. Pasquantonio, L. Cerroni, LED Curing Lights and Temperature Changes in Different Tooth Sites, BioMed Research International. 2016 (2016) 10.
[28] M.Y.M. Chiang, A.A.M. Giuseppetti, J. Qian, J.P. Dunkers, J.M. Antonucci, G.E. Schumacher, S. Gibson, Analyses of a Cantilever-Beam Based Instrument for Evaluating the Development of Polymerization Stresses, Dent. Mater. 27 (2011) 899-905.
[29] Z. Wang, F.A. Landis, A.A.M. Giuseppetti, S. Lin-Gibson, M.Y.M. Chiang, Simultaneous measurement of polymerization stress and curing kinetics for photo-polymerized composites with high filler contents, Dent. Mater. 30 (2014) 1316-1324.
[30] J.W. Stansbury, Dimethacrylate network formation and polymer property evolution as determined by the selection of monomers and curing conditions, Dental Materials. 28 (2012) 13-22.