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Impact Of Photoinitiators Types And Concentrations On Color Stability And Degree Of Conversion Of Light Cure Resin Based Composites: A Scoping Review

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ABSTRACT

The study aimed to enumerate synthetic and natural photoinitiators (PIs), and to find out the impact of PIs types and concentrations on color stability and degree of conversion (DC) of light cure resin-based composites. A thorough literature search from the year 1991 to 2022 was done on PubMed, Cochrane Library, Science Direct, Scopus, Wiley Online Library using different descriptors. All the duplicates were removed and a total of *102* full text articles were assessed. A total of *68* full text articles were selected for final assessment and write up. The studies for color stability and DC were arranged in a tabular form and it was noted that the type and concentration of PIs and also when used in combination do affect the color stability and DC of resin composites. Type and concentration of PIs affect the DC and color stability of resin composites. Camphoroquinone is the PI that has been utilized the most up until this point among the several options that have been tried.

Highlights

1. Understand the range of photoinitiators available to incorporate into chemical structure of composite resin.
2. The type and concentration of photoinitiators play an important role in degree of conversion and color stability of dental composites.
3. Below optimal concentration of photoinitiators, the degree of conversion and hence polymerization are compromised.
4. Above the optimal concentration of photoinitiators, colour stability and biocompatibility concerns may emerge.
5. The combination of camphoroquinone and its co-initiator remains the most frequently utilized photoinitiators.

Keywords: *Photo initiators (PIs), Color stability, Degree of conversion (DC), Resin-based composites.*

1. INTRODUCTION

In 1975, light curing of dental composites was introduced following revolutionary improvements by Michael

Buonocore, who used orthophosphoric to enhance adhesion mechanisms, and Dr. Bowen, who incorporated BisGMA resin in dental composites[1]. Currently marketed dental composites consist of an organic resin matrix, inorganic fillers, coupling agents, and photoinitiators (PIs)[2]. To meet the increasing demands of aesthetic dentistry, compositions are being modified to enhance aesthetics and properties, which are greatly influenced by the composition. The strength, stiffness, and coefficient of thermal expansion are influenced by the resin matrix, fillers, and coupling agent. Flowability and color stability are highly dependent on the type of matrix and PIs, while the resin matrix mainly influences water sorption and polymerization shrinkage[1,2]. Polymerization is defined as the process by which monomers get converted into polymers using varying activation sources to initiate the release of free radicals from PIs. PIs can absorb light, and as a result, either directly or indirectly, generate free radicals that initiate polymerization and lengthen the chain of polymer[1, 3- 5].

Sometimes additional chemicals are required to complete polymerization process, called co-initiators[6]. PIs have certain wavelengths for excitation/absorption. There should be a correspondence between spectral emission from light cure unit and absorption by PI to ensure complete polymerization[7]. The type and concentration of the PIs are fundamental parameters that determine the polymerization characteristics of a resin composite[8]. An optimum correlation between PI and co-initiator type and concentration will maximize photon absorbance efficiency. To obtain optimum polymerization the PI concentration should be optimum enough to ensure high monomer conversion because excessive PI or unreacted monomer both can lead to cytotoxicity[9].

The degree of conversion (DC) is defined as the percentage of Carbon-carbon Double bonds (-C = C-) converted to Single bonds (-C-C-) to form a polymeric resin[10]. DC is a crucial factor used to measure the effectiveness of polymerization. It directly influences the properties of light-curing restorative resins and is affected by multiple factors, including the PI light absorption range, the region of the emitted light spectrum, and the wavelength and intensity of irradiation lamps[11-13]. Incomplete double bond conversion leads to toxicity by leaching out unreacted monomers[12]. Color stability is one of the criteria for a restoration to be considered clinically successful and aesthetically pleasing. It was reported that mismatch in color and translucency leads to esthetic failure of a restoration. For a restoration to be esthetically successful it should maintain its color for a longer period of clinical life[14, 15]. Color stability is influenced by composition and finishing polishing of restoration and dietary habits of patients[16, 17].

The current study aims to review and analyze the published research on synthetic and natural PIs. The second stream of study focuses on the effect of different types and concentrations of PIs on color stability and DC of resin-based composites. We mapped our literature to have maximum understanding of previously existing knowledge and finding the limitations in literature and suggesting the future perspective.

2. MATERIALS AND METHODS

2.1 Data sources: The search strategy for this scoping review on dental photoinitiators (PIs) was specifically tailored to various databases including Scopus, PubMed, Cochrane Library, Wiley Online Library, Web of Science. The literature search spanned from 1991 to 2022.

2.2 Search Strategy: The following descriptors were used in the title and abstract fields as well as Medical Subject Headings (MeSH) was utilized to identify possible qualifying articles: dental PI AND photoinitiators AND concentration of PIs AND type-1 PIs AND type-2 PIs AND CQ AND benzophenone AND 1-phenyl-1,2 Propanedione AND natural PI AND synthetic PI.

2.3 Data filtering and screening: A comprehensive search was conducted. The data filtration involved selection of relevant studies based from their description and titles from each data base. Then the results from all searches were pooled and duplicate studies were removed from the pool. Titles were examined by author and abstracts. Abstracts were scrutinized to select relevant studies. The review aimed to map existing literature regarding different types of PIs and their effects on color stability and degree of conversion (DC) of composites within the fields of dentistry, dental materials, and biomaterials. A total of 68 full-text articles were selected for final assessment and inclusion in the review write-up as shown in figure 1. This rigorous process ensured that the literature included was both relevant and of high academic quality, aligning with the objectives set forth at the beginning of the review. Only original research articles, review papers, and conference papers published in English were considered. Articles which were in English language were selected. Exclusion criteria was case reports and books and those articles which were focused on physico mechanical properties other than degree of conversion and color stability. Full text articles for final review preparation were obtained and read thoroughly to ensure their relevance before confirming their eligibility for the review. Information was collected by the members and relevant data and information was obtained. Findings from the studies were used to prepare review article and to present the relevant articles in tabular form and in final conclusion.

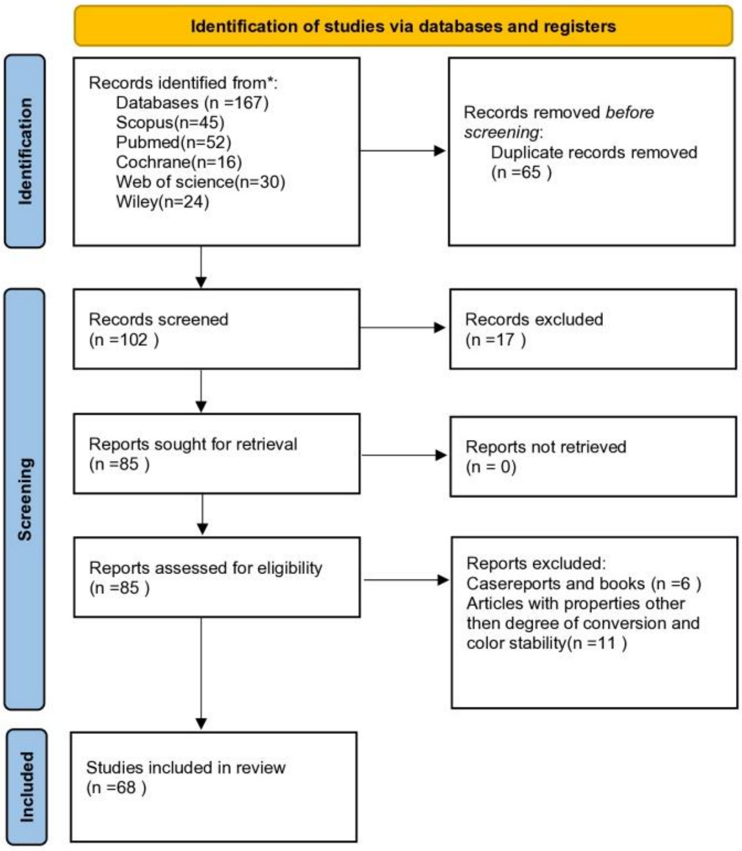


Figure 1: Prisma flow chart

3. RESULTS AND DISCUSSION

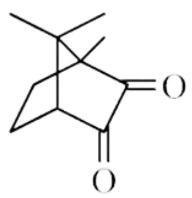
3.1 Types of PIs

The type and concentration of the PI system are fundamental parameters that determine the polymerization characteristics of a resin composite[8]. They can be classified based on their origin and polymerization technique. According to source of origin, they are classified as natural or synthetic, as stated in table 1. Naturally derived PIs have numerous applications, including experimental dentin bonding agents, primers, guided tissue regeneration membranes, and 3D printing materials. They are abundant, biocompatible, and cost-effective compared to synthetic PIs[1, 5]. According to polymerization process the PIs are divided into two types, type I and II as shown in table 1. Type I PIs do not require co-initiator molecule to generate free radicals whereas type II PIs need co-initiator molecule to initiate polymerization process[18]. Absorbance range, maximum absorbance, color and light spectrum for PIs are mentioned in table 1. The structures of PIs are shown in figure 2 (A-M).

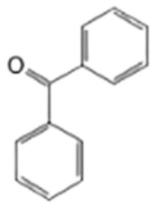
Table 1. Types and Specification of Photo initiators[1,5]

Names and their abbreviation	Type	Natural/ synthetic	Absorbance range	Absorbance maximum	Color	Light spectrum
Camphoroquinone (CQ)	II	Synthetic	380-500nm	468nm	Yellow	Blue light
Benzophenone (BP)	II	Synthetic	Two broad bands weak: 320-370nm Stronger: 240-300nm	294nm	White	UV light region

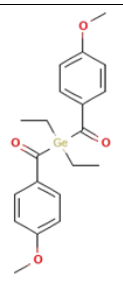
Ivocerin (IVO)	II	Synthetic	390-445nm	418	-	Both blue and violet
9, 10-Phenanthrenequinone (PQ)	II	Synthetic	400-490nm	420nm	Orange	Both UV and visible light region
Phenyl propanedione (PPD)	II	Synthetic	350-490nm	410nm	Pale yellow	Violet light
Bisacylphosphine oxide (BAPO)	I	Synthetic	365-416nm	400nm	Solid	Violet light emitting diode
Trimethylbenzoyldiphenylphosphine Oxide (TPO)	I	Synthetic	380-425nm	400nm	Colorless	UV light
Benzoyl germanium substances (BTMGe)	-	Synthetic	Longest wavelength range	411nm	Yellow	Visible light
Riboflavin	I or II	Natural/ plant origin	200-470nm	223 nm, 267 nm, and 373 nm in the UV region, and 444 nm in the visible light region	Orange yellow	Both UV and visible light
Coumarin	II	Natural	270-510nm	405 and 421nm	Colorless or white	Near UV and visible light
Curcumin	II	Natural	350-750nm	417nm	Orange yellow	UV visible light
Vanillin derivatives	I	Natural	-	385nm	white	UV region
Chlorophyll	II	Natural	430-670nm	Blue region 453 Red region 642	green	Blue red



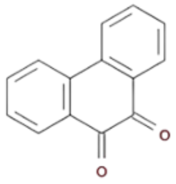
A. CQ

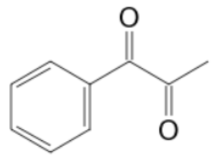


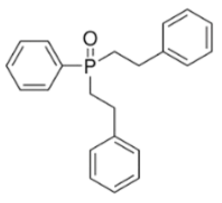
B. BP



C. IVO







BAPO (Fig. 1.6)

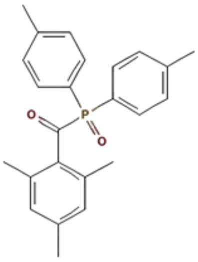
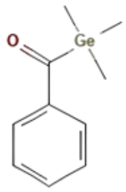
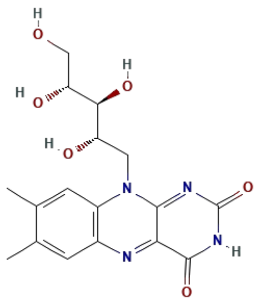
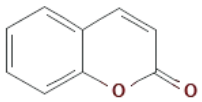
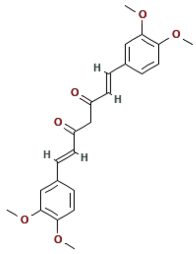
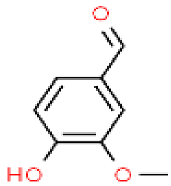
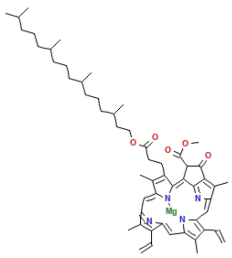
D. PQ	E. PPD	F. BAPO
 G. TPO	 H. BTMGe	 I. Riboflavin
 J. Coumarin	 K. Curcumin	 L. Vanillin
 M. Chlorophyll		

Figure 2 (A-M) Chemical structure of 13 photoinitiators [19]

Camphorquinone (CQ) is most commonly used type II PI in light cured dental composites [1]. Dimethyl Aminoethyl Methacrylate (DMAEMA), is usually added as co-initiator with CQ to accelerate the initiation process during polymerization[4,20]. Due to compatibility with most dental units, there is increased polymerization with CQ which results in improved mechanical properties[21]. The disadvantages of CQ/amine include hydrophobicity and toxicity, undesired yellowing, and low effectiveness in constructing chemical networks in acidic monomers at early stages[17,22]. CQ may lead to toxicity if higher concentrations are used [3]. 1-Phenyl-1, 2- Propanedione (PPD) is a type II PI that improves polymerization kinetics by maintaining a stable DC and reducing the yellowing effect of CQ [22]. It was discovered that utilizing PPD alone or in conjunction with CQ did not improve final attributes when compared to CQ alone, which may explain its limited commercial use[24].

Benzophenone (BP) is one of the widely marketed PIs because of its curing properties, suitable solubilities, oxygen inbition effects and cost effectiveness[25-27]. As radicals generated from BP are usually ineffective in initiating free radical polymerization due to the delocalization of unpaired electrons and the steric hindrance it requires co-initiators to induce polymerization[1,28]. Ivocerin is a type-II PI, which when exposed to light cleavages a chemical bond within itself and reacts with monomer to form polymerization network[29]. In comparison to CQ, Ivocerin possesses faster, deeper polymerization and a higher reactivity to curing light[30,31].

Thyroid Peroxidase Antibodies (TPO) is a type-1 PI that does not require coinitiators to initiate polymerization reaction[3,32]. TPO is more efficient at generating free radicals than CQ and yet promotes color stability on aging due to elimination of coinitiators [33,34]. However, TPO has significant downsides, including higher polymerization stresses than CQ, a shorter depth of cure, and being hydrophobic, making it less appropriate for water-containing adhesive systems[35]. Bisacylphosphine oxide (BAPO) is a type-1 photosensitizer that does not need a co-initiator to begin the polymerization process. BAPO is solid with a symmetric chemical structure, and it is poorly soluble in a variety of monomers and oligomers. The reactivity of BAPO is only limited by poor solubility in UDMA[36.] Howeever this instability does not occur when other monomers are used[37].A group of researchers claimed efficient polymerization and good degree of conversion of BAPO owing to release of four reactive radicals[13].Benzoyl germanium substances-BTMGe are type I PIs. The photolysis of these PIs generates two radicals[1].

Riboflavin, or vitamin B2, is derived from plant origins, such as asparagus, broccoli, and spinach, or animal origins such as kidneys and liver. Riboflavin is the most commonly used naturally derived PIs in dental and biomaterials applications[5,38,39].Riboflavin has the ability to produce superoxide radicals that can consequently initiate a polymerization reaction[5].

Disadvantage associated with riboflavin is discoloration of final polymer structure[40]. Coumarins and keto-coumarins are naturally derived compounds of plant origin[41]. Coumarins are type II PIs that can initiate the free radical polymerization of methacrylate, cationic polymerization of epoxy resins and photopolymerization of photosensitive 3D printing resins etc. High concentration of coumarin can be hepatotoxic thus care should be taken to use the least efficient amount to photoinitiate polymerization reaction[42].

Curcumin is derived from the rhizomes of Curcuma longa. Curcumin has been proven to reduce dental bacterial biofilm formation[5]. It is a type II PI that is used with coinitiators and accelerators in three- component PI system for free radical polymerization of methacrylates, leading to formation of highly cross-linked polymers with high thermal stability and mechanical strength[43] Vanillin-derived PIs do not require co initiators which increase their biocompatibility. They possess antimicrobial properties and are used in 3D printed scaffolds for tissue engineering[5,44]. Chlorophyll and its derivatives are found in plants, eukaryotes, and cyanobacteria, and are essential for photosynthesis process. Chlorophyll derivatives have proven antiviral activity against SARS-CoV-2 virus[5,45] They can initiate the free radical photopolymerization of methyl methacrylate, to produce cross-linked polymethyl methacrylate[5,46]

3.2 Effect of PI type and concentration on Color Stability

Table 2 shows a number of studies carried out on effect of type and concentration of PIs on color stability. It is reported in many studies that using the combinations of PIs improves physical characteristics, DC, color stability, and bond strength[15,23,48,49,50]. Being type I PI TPO does not require coinitiators which ensures color stability when used as PI[3,32,34]. It was reported in a study that efficacy of Benzoyl germanium substances—BTMGe when incorporated in luting cements was higher in terms of DC and color stability[1]. Coumarins being white or colorless PIs have an advantage of maintaining color stability over a period of time[50,51]

3.3 Effect of PI type and concentration on degree of conversion (DC)

Table 3 shows a number of studies which reported effects of type and concentration of PIs on DC of resin-based composites. Generally higher concentration leads to better conversion[12,52,53]. At higher PI concentrations, the probability of excitation is increased as more PI molecules are present and the light energy absorbed increases proportionally when compared to lower concentrations, regardless of PI type. However, in the case of colored or 'pigmented' PIs such as CQ, the use of high concentrations of CQ and amine may lead to undesirable yellowing effects and also biocompatibility issues[54,55]. Many studies are depicted in table 3 to show the effect of type and concentration of PIs on DC. Using too low concentrations causes insufficient conversion, resulting in undercured polymer structure which leads to lower mechanical and physical properties[52,56]Using CQ up to an optimum concentration improved the DC and mechanical properties. Above that there is no significant impact of concentration on mechanical properties[57,58].

It was reported that DC of PPD is comparable to CQ if halogen light is used for photoactivation. Using combination of the two shows increased DC and better physical properties[48,49]. DC of CQ and BAPO containing composites was comparable as reported in another study[25]

However, improved DC, rate of polymerization and improved mechanical properties were reported if activated by high power LED light curing unit[59,60] It was also reported that higher efficiency of TPO-based and BAPO-based resins might lead to higher C=C conversion than materials formulated with CQ[61,62]. It was observed by researchers' group that light activation of PPD is more profound when it is carried out by light sources which are in wavelength limit closer to ultraviolet and visible light leading to greater values of DC[59,63,64].

4. CONCLUSION

The type and concentration of PI plays an important role in DC and color stability of dental composites. A higher concentration (optimal level) of PI produces a higher DC. Below optimal concentration, the DC and hence polymerization are compromised, and above the optimal concentration, biocompatibility concerns may emerge. It is also established that using PIs in combination increases DC more than using them alone. Many researchers have found that the type and concentration of PIs affect the color stability of dental composites. To this day, the combination of CQ and its co-initiator remains the most frequently utilized PI, even though numerous alternatives are now being considered and tested.

Author, year	objectives	methods	outcomes
Arikawa et al, 2009.[33]	To investigate effects of various PIs on the polymerization and color of the light-activated resins.	PPD, TPO, BAPO and CQ were used in this study. Color value was determined before and after polymerization using spectral transmittance meter.	TPO containing resins less color change as compared to resins with other PIs
Shin DH and Rawls HR, 2009[64]	To evaluate efficacy of OPPI as component in PI system.	Camphorquinone (CQ) and OPPI were combined in various proportions with DMAEMA. Each CIELAB scale was determined with a colorimeter.	Greater color stability obtained when OPPI included.
Silami et al, 2013[65]	To evaluate the color stability (ΔE) of composites containing different PIs submitted to accelerated artificial aging.	CQ, PPD used alone or in combination were tested. After they were submitted to AAA for 300 h, the final color readouts were made.	PI, with tendency towards yellowing, did not interfere in the color stability of composites submitted to AAA.
De Oliveira et al,2015[48]	To evaluate the effect of PIs and reducing agents on cure efficiency and color stability of composites using different LED wavelengths.	Color stability of TPO, BAPO and CQ/DMAEMA was tested after aging.	CQ-systems presented higher color stability than BAPO and TPO despite their higher cure efficiency.
Albuquerque et al, 2015.[23]	To determine: DC, depth of cure, and color stability of composites formulated CQ and PQ PIs.	Color stability of formulations containing PIs was observed. The optical properties were determined with a spectrophotometer.	Groups formulated with PQ produced greater yellowing and less color stability than the traditional combination CQ and amine.
Manojlovic et al, 2016.[66]	To study the effect of a low-shrinkage methacrylate monomer and monoacylphosphine oxide photoinitiator on color, translucency, and color stability of model resin-based composites (RBCs).	Four micro-hybrid RBCs containing CQ or TPO as PIs were prepared and optical properties were studied using Thermo Scientific Evolution and SpectroShade ™ Microspectrophotometers. Color stability was evaluated after immersion in black tea and distilled water.	TPO containing RBCs showed better color stability than CQ-containing RBCs irrespective of the base monomer.
Maciel et al, 2018[3]	To evaluate the effect of PI concentration on polymerization characteristics and physical-mechanical properties of experimental flowable composite.	Model composites based were prepared containing different concentrations of CQ on resin matrix (0.25%, 0.50%, 1%, 1.50%, and 2% by weight. Color was assessed by reflectance spectrophotometer, employing the CIE-Lab system.	A strong linear correlation was found among CQ concentration and color parameters, positive for yellowing and negative for luminosity.

Table 2. Effect of PI type and concentration on color stability of resin composites

Author, year	Objectives	Methods	Outcomes
Yoshida and Greener 1994[57]	To evaluate effect of PI on DC of unfilled light-cured resin.	The DC of combination of different concentrations of CQ and DMAEMA was measured using FTIR.	Maximum DCs upto certain level and at CQ concentrations of 2.0 mol.% and above
Park et al 1999[67]	To Development of a new PI system for dental light-cure composite resins	Effect of PI type (CQ or PPD) and ratio (PPD/CQ) on DC was investigated using FTIR	Greater DC values were observed for combination rather using a PI alone
Musanje et al 2009[52]	To determine efficacy of OPPI when use as component of PI system	Combinations of CQ and OPPI were used along with DMAEMA in varying concentrations and DC was measured with FTIR.	Using OPPI in combination with CQ reduces CQ and amine concentration while maintaining or improving
Shin and Rawls 2009[64]	To determine the concentrations of CQ and EDMAB that resulted in maximum conversion	Combination of CQ and EDMAB was used in varying ratios to observe the effects on DC.DC was measured using micro FTIR.	DC was optimized at a CQ:EDMAB ratio of 2.40:0.83mol% beyond this concentration a decline in properties was observed
Arikawa et al, 2009[33]	To determine effect of various visible light PIs on the polymerization and color of light- activated resins	Efficiency of four visible light PIs CQ, PPD, TPO and BAPO were tested using different light cure units. FTIR was used to determine DC	PPD-, TPO-, and BAPO- containing resins polymerized with V-LED light unit had approximately the same or higher degree of monomer conversion, in comparison with that of CQ- containing resin polymerized with a dental QTH LCU
Schneider et al, 2012[24]	To evaluate the kinetics of polymerization, and the depth of cure for filled dimethacrylate resins formulated with different photoinitiator systems	CQ+EDMAB (control); TPO and TPO+EDMAB were used in this study. Photoinitiator absorption and QTH-light emission were evaluated	The photoinitiator TPO produced higher DC and faster reactions (higher Rpmax) than CQ, regardless of the presence of the amine EDMAB
Alonso et al, 2014[56]	PI concentration and modulated photoactivation: influence on polymerization characteristics of experimental composite	DC of composites using different concentrations of CQ was measured using FTIR spectroscopy	PI concentration was the determinant factor on polymerization characteristics of the composites. Higher concentration of PIs increased the DC.
Meereis et al, 2014[13]	To evaluate the performance of BAPO as an alternative photoinitiator	Real time FTIR was used to measure DC of PI systems formed with combinations of CQ, EDAB, BAPO and DPIHFP	CQ alone: lower monomer conversion. BAPO alone: increase in conversion was higher. The ternary system. BAPO+EDAB+DPIHFP showed the highest polymerization and conversion rate.
Najafi et al 2018[68]	To evaluate physical mechanical properties of novel dental resin composites incorporating BAPO and CQ	DC of micro hybrid composites with CQ/BAPO as PIs measured using FTIR	BAPO exhibited better results as compared to both CQ and CQ/BAPO PI systems.
Maciel et al 2018[3]	To evaluate the effect of CQ concentration in physical-mechanical properties of experimental flowable composites to find the concentration that results in maximum conversion	DC was measured by using varying concentrations of CQ in experimental composites using FTIR.	Significant increase in DC was observed up to 1% CQ concentration while at higher concentrations (1.5% and 2%) no significant difference was observed.

Wang and Wang 2021[58]	To determine optical concentration of CQ for light cured dental resin	DC of composites with varying conc of CQ was measured using in-situ near-infrared (NIR) spectroscopy	DC increased with increasing CQ concentration. However, above a certain concentration (~0.5 wt%), no change was observed

Table 3. Effect of PI type and concentration on DC

Author contributions

All authors contributed in designing the study, collecting the data, interpreting the results, writing up the draft of the manuscript, reviewing and finalizing the manuscript.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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