

**EVALUATION OF COLOUR STABILITY,
GLOSS, MICROHARDNESS AND SURFACE
ROUGHNESS BETWEEN MICROHYBRID AND
NANO-BASED LIGHT CURED COMPOSITES IN
DIFFERENT pH SOLUTIONS**

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UNIVERSITI SAINS MALAYSIA

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by

NAZIAH BINTI MD JASIN

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for the degree of
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LIST OF SYMBOLS

μm	Micronmeter
ΔE	Colour difference
α	Significant level

LIST OF ABBREVIATIONS

ANOVA	Analysis of variance
Bis-GMA	Bisphenol A-glycidyl methacrylate
bis-EMA	Bisphenol a diglycidyl methacrylate ethoxylated
C	Chroma (saturation)
C=C bonds	Carbon double bonds
C-C bonds	Carbon single bonds
CIE	Commission International de l'Éclairage
DOC	Degree of conversion
gm	Gram
GU	Gloss unit
H	Hue
kgf	Kilogram-force
L	Lightness
mm	Milimeter
nm	Nanometer
PEGDMA	Polyethylene dimethacrylate
QC	Camphoroquinone
R	Free radical
R _a	Roughness average
SD/σ	Standard deviation
Si	Silicone
SiO ₂	Silicon dioxide
SEM	Semi electron microscope
SPF	Small particle-filled
TEDGMA	Triethylene glycol dimethacrylate
VHN	Vickers hardness number

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**PENILAIAN TERHADAP KESTABILAN WARNA, KILAUAN, KEKUATAN
MIKRO DAN KEKASARAN PERMUKAAN ANTARA KOMPOSIT
PEMPOLIMERAN CAHAYA MIKROHIBRID DAN BERASASKAN NANO
DALAM LARUTAN pH YANG BERBEZA**

ABSTRAK

Teknologi nano dalam pergigian bukanlah sesuatu yang baharu. Aplikasi teknologi nano memberikan banyak kelebihan dan faedah terutamanya dalam penghasilan bahan restoratif yang berkualiti tinggi. Penyelidikan dan penambahbaikan berterusan dijalankan bagi memastikan resin komposit baru yang dihasilkan mempunyai ciri-ciri unggul yang tahan terhadap cabaran mekanikal untuk kekal dan bertahan dalam persekitaran mulut sepanjang hayat. Kajian ini bertujuan untuk menilai pengaruh pelbagai jenis minuman yang digunakan secara meluas di Malaysia terhadap kestabilan warna, kilauan permukaan, kekerasan mikro, dan kekasaran permukaan resin komposit kontemporari. Sebanyak 450 sampel disediakan terdiri daripada tiga jenis komposit yang diuji; Filtek™ 250; Filtek™ 250 XT; Filtek™ 350 XT (n=150). Semua sampel direndam dalam lima larutan rendaman yang terdiri daripada air suling, air liur tiruan, minuman ringan, minuman tenaga dan kopi hitam segera selama 100 hari. Dapatan kajian ini dianalisis menggunakan ujian ANOVA sehala dan post-hoc pada aras signifikan $p < 0.05$. Hasil kajian mendapati kesemua komposit yang direndam dalam pelbagai nilai pH mengalami perubahan warna dan kilauan yang ketara selepas penilaian 24 jam, D10, D20, D30, D60 dan D100 menggunakan *colorimeter* dan *gloss meter*. Penilaian tahap kekerasan mikro menggunakan Ujian Kekerasan Vickers telah dilakukan selepas tempoh rendaman 24 jam, D30 dan D100 mendapati komposit isian nano mempunyai kekuatan yang lebih tinggi terhadap tegasan mekanikal berbanding

komposit mikrohibrid. Penggunaan *Scanning Electron Microscope* (SEM) untuk menilai degradasi morfologi resin komposit menunjukkan sampel yang direndam dalam air suling kurang terjejas berbanding sampel yang direndam dalam air liur tiruan, minuman ringan, minuman tenaga dan kopi hitam segera pada D100. Kesimpulannya, perubahan mekanikal dan fizikal yang dialami oleh resin komposit tidak mempunyai hubungan langsung dengan keadaan dedahan berasid sebaliknya bergantung kepada komposisi bahan isian, saiz, berat, agihan serta proses pempolimeran di samping tempoh pendedahan yang panjang.

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ABSTRACT

Nanotechnology in dentistry is not something new. The application of nanotechnology provides many advantages and benefits especially in the production of high-quality restorative materials. Research and continuous improvement are conducted to ensure that the newly developed composite resins have superior characteristics that are resistant to mechanical challenges to remain and sustain in the oral environment for a lifetime. This study aims to evaluate the influence of various types of beverages that are widely used in Malaysia on colour stability, surface gloss, microhardness, and surface roughness of contemporary composite resins. A total of 450 samples were fabricated from the three types of composites tested; Filtek™ 250; Filtek™ 250 XT; Filtek™ 350 XT (n=150). All samples were immersed in five immersion solutions consisting of distilled water, artificial saliva, soft drink, energy drink and instant black coffee for 100 days. Findings of this study was analysed using one-way ANOVA and post-hoc test at significant level of $p < 0.05$. The results of the study showed that all the composites immersed in various pH levels experienced a significant change in colour and gloss after the assessment periods of 24 hours, D10, D20, D30, D60 and D100 using a colorimeter and gloss meter. Evaluations of the microhardness level using Vickers Hardness Test was performed at 24 hours, D30 and D100 found that nanofilled composites present a higher strength against mechanical stress compared to microhybrid composites. Evaluations of morphological degradation of the composite resins using Scanning Electron Microscope (SEM) showed that

samples immersed in distilled water were less affected than those immersed in artificial saliva, soft drink, energy drink and instant black coffee on D100. In conclusion, the mechanical and physical changes experienced by the composite resins have no direct relationship with the acidic conditions exposure but instead depend on the filler composition, size, weight, distribution and polymerisation process in addition to the long exposure period.

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Development in material science, robotics and biomechanics has dynamically and dramatically evolved in terms of human anatomy components replacement. Historically, many dental approaches were introduced and used to replace the loss of tooth surface structure caused either by diseases or other injuries. The replacement or reconstruction treatment of tooth surface loss has contributed to and continues to be a great part of general dental practice. In dentistry, restorative dental materials are important as it is the foundation for the replacement and reconstruction of the lost structure mainly dental hard tissue. Comparable with other anatomical prostheses development, dental materials have rapidly progressed due to their long-term success with higher expectation to outperform natural materials replacement.

Interaction between the complexity of the oral cavity including bacteria, high masticatory forces, ever-changing pH situation and thermal environment create a unique, specific requirement and manipulation of dental materials. The diversity of the oral cavity is considered to be the harshest environment for the restorative materials when directly placed into the tooth cavities. Knowledge of dental material sciences is very important, particularly in choosing the right materials that suit specific dental applications or in designing the best solution for teeth restorations.

Restorative dental materials are representative of a broad class of materials and are generally categorised as metals, polymers, ceramics and composites. Each class of materials is distinctly different from others in terms of density, stiffness, translucency, processing method, application, and cost. Generally, the dental materials used in

restorative procedures can be classified as composite resins, cements, glass ionomers, ceramics, amalgam alloys, gypsum materials, dental waxes, denture resin base and impression materials.

The demands for material characteristics and performance are varied, ranging from high flexibility, high stiffness, bone integration (osteointegration), excellent adaptation to the existing tooth structure as well as geometric dimensional and structural reproducibility. Most restorative materials are characterised by their physical, chemical and mechanical parameters obtained from laboratory-tested data. However, aesthetic, stiffness and osteointegration are factors that normally dictate the choice of material classes. In addition, the coordination between restoration and material composition is essential and critical to ensure the final outcome meets the desired standard and effectively function.

Metal and alloy are the two common materials used in most aspects of dental practice including direct and indirect dental restorations, dental implant components and dental instruments. During the twentieth century, metal and alloy restoration namely dental amalgam has been the predominant preferred restorative material in restoring the posterior teeth. Dental amalgam was chosen due to its advantages of providing relatively low cost, durability and being less sensitive to clinical procedures compared to other materials for the same purpose. Despite the strength, dental amalgam was also found to have some shortcomings in terms of the absence of adhesion to the dental structure, which requires extensive cutting of healthy sound tooth structure (Cenci & Powers, 2004).

Since composite resin was invented and introduced in the early 1960s, dramatic changes in selecting restorative materials have been shifted. The first developed

composite resins have improved clinical performance by providing higher mechanical properties, lower thermal coefficient of expansion, lower-dimensional change on the setting, and higher resistance to wear (Ronald L. & Powers, 2012). Therefore, composite resins have become a viable alternative dental restorative material for both anterior and posterior restorations. A dynamic and continued improvement over the years has resulted in modern composite materials with excellent durability, wear resistance, and aesthetics that mimic natural teeth. Despite patients' priority on aesthetic satisfaction, a combination of a total-etch acid technique provides excellent adhesive properties between the material and tooth structure. Thus, restricting the excessive cutting of sound healthy tissue during cavity preparation upon removal of the carious lesion, which is not possible when dealing with dental amalgam restoration (Demarco et al., 2007).

The success of dental composites in restorative dentistry compared to other restorative materials is due to their increasing aesthetic properties and high acceptance by both clinicians and patients. Increasing aesthetic requirements combined with adequate durability for long-term success are considered as essential factors (Zhang et al., 2021) and therefore, composite is considered the material of choice for the restoration of both anterior and posterior load-bearing surfaces (Lynch et al., 2014). Along with the aim of mimicking the natural properties of the tooth, the bond strength, favourable strength, ease of application and relatively low cost compared to ceramics contribute to the improvement of mechanical properties, allowing them to persist in various oral environments (Dionysopoulos et al., 2015; Domingos et al., 2011; Hengtrakool et al., 2011; Malek et al., 2015).

Changes in the composition of the matrix, such as the type and size of fillers, have contributed to the significant constant development over the years (Chen, 2010). Previous *in vitro* and *in vivo* studies have also reported that direct aesthetic restorative materials are colour-unstable, probably due to chemical reactions as well as exposure to saliva and stains from foods or beverages in the oral cavity (Reddy et al., 2013; Borah, Diganta et al., 2017; Ceci et al., 2017; Mara da Silva et al., 2019). Prolonged exposure to intrinsic and extrinsic factors may alter the composition of composite resin, mainly due to an inadequate polymerisation process, water absorption and discolouration due to dietary and hygiene habits (Topcu et al., 2009; A. B. Borges et al., 2014). Food and beverage compositions have been reported not only to cause discolouration but also to affect the surface of the composite resin. Thus, promoting the degradation of the organic dispersion phase and lead to a change in surface hardness (Hengtrakool et al., 2011; Torres et al., 2015).

The surface degradation response is mainly influenced by the type of filler, matrix, chemical bonding, pH, copolymer composition and water absorption. Among these factors, pH is an unfavourable factor for hydrophilic resins and is known to affect the rate of degradation by catalysis (Moon et al., 2015). Low pH solutions have been reported to affect wear resistance while highly alkaline solutions have been shown to accelerate hydrolysis, resulting in damage to the surface microstructure (Cilli et al., 2012). On the other hand, continuous and frequent contact with acidic conditions increases the degradation potential through changes in surface roughness, translucency and surface gloss (Nooh et al., 2020). Due to the diversity and complexity of the intraoral environment in combination with exposure factors, *in vitro* studies are very important to gain insight into the basic mechanisms of biodegradation of current composite resins (Briso et al., 2011; Catelan et al., 2011).

After many centuries of the dental practice, dental practitioners continue to be confronted with the problem of replacing the composite restorations due to discolouration, secondary caries or fracture (Demarco et al., 2017). In order to improve the longevity of composite restorations, potential failure factors related to the patient, operator, material and tooth (Kubo et al., 2011) shall be continuously investigated.

1.2 Problem statement

Two types of composites, microhybrid and nano-based light cured composites, are commonly used for restorative dental procedures due to their superior mechanical properties and aesthetic appearance. The performance, durability, aesthetic qualities and longevity of dental restorations are significantly influenced by the properties of the composite materials used. However, the dynamic oral environment conditions characterized by the fluctuations in pH levels, can impact the physical attributes of these materials, potentially affecting their durability and visual appearance over time (Elbishari et al., 2020).

Exposure to neither acidic nor alkaline pH solutions which representing various conditions found in oral conditions reported to affect the degree of colours stability of composites resins (Topcu et al., 2009). In addition to disrupting colour stability, different pH environment may also affect the gloss retention and surface shine, thus affecting the aesthetic appeal as it will interfere the colour harmony with the surrounding teeth or restorations (Lee, 2015).

Intermittent and continuous exposure to various pH conditions has proven to increase the ability to abrade and soften the matrix of composites resins (Jyothi et al., 2012). Erosion effects have also been reported to interfere with the hardness rate which reflecting the mechanical durability towards wear resistance (Xavier et al., 2016). The

variation of pH levels was also found to have great impact on surface roughness. The alteration on composite surface is not limited to the occurrence of exterior colour change but been reported to cause a prominent role in plaque accumulation and the formation of oral biofilm (Park et al., 2012).

As reported by Salleh et al. (2020), about 36.9% of Malaysian adolescents had consumed carbonated soft drinks at least once daily throughout the 30 days of survey. Due to an increasing trend of carbonated soft drink intake and wider use of aesthetic restorative materials particularly composite resins in dentistry, experimental *in vitro* study was conducted to assess the affected attributes after a prolonged exposure to different pH solutions that mimic the challenges faced in the oral environment.

Limited evidence of cause-effect relations and separate investigation of all variables in Malaysia has prompted to this present study. Therefore, this experimental *in vivo* study was conducted to further investigate by integrating all four factors including their cause-effect associations.

1.3 Research questions

1. How does pH affect the colour of the composite resin?
2. How does pH affect the surface gloss of the composite resin?
3. What is the impact on the microhardness of the composite resin after a prolonged immersion in various beverages?
4. Do composite resins undergo morphological changes in term of surface roughness after a prolonged immersion in various beverages?

1.4 Research hypothesis

1.4.1 Null hypothesis

1. The pH of beverages has no significant difference to the colour stability of the composite resins tested.
2. The pH of beverages has no significant difference to the gloss of the composite resins tested.
3. The pH of beverages has no significant difference to the microhardness of the composite resins tested.
4. The pH of beverages has no significant difference to the surface roughness of the composite resins tested.

1.5 Research objectives

1.5.1 General objective

This study aims to evaluate the influence of different pH of highly consumed beverages in Malaysia on the colour stability, gloss, microhardness, and surface roughness of contemporary composite resins.

1.5.2 Specific objectives

The specific objectives of this study are:

1. To determine the pH value of commonly consumed beverages using the digital pH meter.
2. To determine the influence of pH in selected beverages on the colour change of the composite resins using a colorimeter.

3. To determine the influence of pH in selected beverages on the gloss of the composite resins using a gloss meter.
4. To evaluate the microhardness of different types of composite resins after immersion in selected beverages using the Vickers hardness test.
5. To investigate the surface roughness of the composite resins after exposure to selected beverages using Scanning Electron Microscope (SEM).
6. To determine the association between each composite resin tested with chemical challenges encountered.

1.6 Significance of research

Recent studies that reveal the association between common canned beverages consumed by Malaysian adolescents and the available modern composite resins are limited. Therefore, this research was conducted to focus on the influence of acidity levels contained in the majority of beverages available in the market on the aesthetic and mechanical properties of the most commonly used composite resins in general dental practice. Since the available data regarding the influence of acid beverages on composite resins in Malaysia is limited, perhaps these experimental *in vitro* findings may provide valuable preliminary data and pave way for further clinical studies. However, *in vitro* findings may not always perfectly replicate the complex interactions and conditions encountered in the human oral cavity. Therefore, experimental *in vitro* study or clinical trials are crucial to validate the findings in a real-world context that can provide more relevant data on how these materials might react in real-life settings.

CHAPTER 2

LITERATURE REVIEW

Over five decades ago, tooth-coloured restorations mainly the composite resins have become the most popular dental material of choice to directly restore both anterior and posterior teeth, and it is currently considered as a universal material. Ever since the 1960s, composite resins have rapidly developed to increase their physical-mechanical properties and aesthetic characteristics. Recently, a published study reported that composite restoration had a long-lasting durability in clinical service with good clinical performance and respective minimal annual failure rate throughout six years of clinical evaluation (Ruya et al., 2021).

2.1 Historical perspective of restorative materials

Back in the 18th century, operative dentistry was embraced and observed by chairside dentistry including the barbers/artisans. In the beginning of the 19th century, the role was shifted and no longer conducted by the barbers/artisans but was professionally practiced by dentists or surgeons. Earlier before, Charles Allen (1678) produced the first written material on dentistry entitled “Operator of the Teeth”. The written material on dentistry was then precisely translated and practiced by Pierre Fauchard (Father of Dentistry) who later published his first “Traite des dents” in 1728. Following that, the textbook of Operative Dentistry was introduced in 1889 with the inclusion of oral surgery and fixed prosthodontics as part of operative dentistry (Fillerbrown, 1889). The field of operative dentistry has been expanded to include oral surgery, fixed prosthetics and other related branches as today’s existence (Schulein, 2005).

By the 20th century, various studies on restorative materials had been conducted to refine and improve the quality and process by applying engineering science with physical and mechanical tests to its molecular structures. Apart from that, the shortcomings of the restorative materials were also identified and improved by introducing them to the current technology. These persistent efforts are intended to produce higher-quality restorative materials with specific properties for specific purposes (Singh et al., 2017).

Aesthetic dentistry is not a special discipline of dentistry. It is one of the existing branches of operative dentistry by emphasising the functional and biological aspects. Aesthetic dentistry was developed to represent the goal of dental treatment including the specialised areas from restorative and preventive to periodontics, orthodontics, prosthodontics, oral surgery and maxillofacial surgery (Blatz et al., 2019).

The quest for appearance improvement, especially facial and teeth has been started since ancient times (Hussain & Khan, 2014). This has been a breakthrough for the development of another separate medical discipline known as aesthetic dentistry. In the 20th century, the roles of aesthetic dentistry have been expanded to include preventive measures, restoration replacement materials as well as partial and complete dentures fabrication. In other words, aesthetic dentistry was developed to facilitate a specialised dental treatment related to functional, aesthetic and facial deficiencies (Blatz et al., 2019).

The importance of aesthetic dentistry was supported by various strong scientific evidence related to the appearance of an individual's face and teeth. The imperfection of facial appearance or teeth may leave a profound impact on others'

judgement and perceptions (Blatz et al., 2019). Although perception depends on a psychological basis, aesthetically pleasing appearance contributes to attractiveness and well-being life (Van Der Geld et al., 2007; Blatz et al., 2019).

Initially, the composite resin was developed to restore the anterior teeth. However, the continuous and dynamic developments in restorative materials have produced more versatile composite resins that benefit both parties; clinicians and patients (Figure 2.1). Despite resembling and matching colour of the natural tooth, the current composite resins combined with excellent adhesive protocols allow clinicians to conduct more conservative, safer, feasible dental procedures and successfully beautiful results (Murariu et al., 2020).

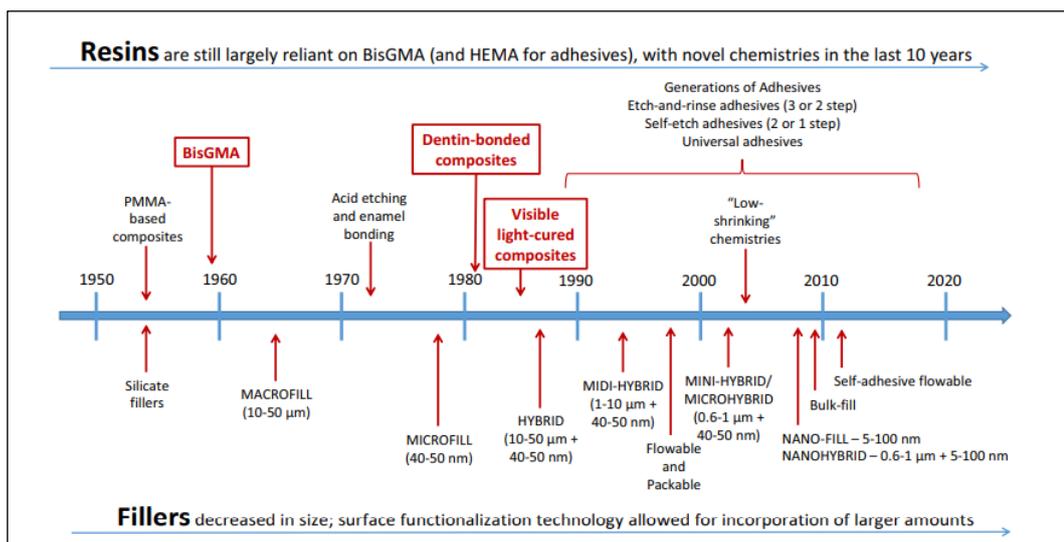


Figure 2.1 Chronology and timeline of the development and improvements of the composites and adhesives (Adapted from Figure 1.3.2D.1 in Ferracane, J.L. & Pfeifer, C.S. (2020). *The Organic Matrix of Restorative Composites and Adhesives*. In *Biomaterials Science* (pp. 139-152). Academic Press

2.2 Composite resins formulation

2.2.1 Conventional system – the monomer matrix

Composite resin is a unique and multiphase restorative material that exhibits organic and inorganic polymers which complement each other to enhance superior

properties. Since it was introduced more than 50 years ago, composite resin has been through many significant enhancements. Oscar Hager (1951), a Swiss chemist invented the first composite resin composed of dimethacrylate molecule that allowed a cross-polymerised interaction between matrices. Advancement in chemistry was later developed by Rafael Bowen in 1962 by introducing a large molecular monomer composed of an organic monomer called bisphenol A-glycidyl methacrylate known as Bis-GMA or Bowen's resin. Due to the larger molecular structure, Bis-GMA plays a major role to reduce the stress of polymerisation shrinkage and increase the fracture resistance (Ilie & Hickel, 2011; R. Wang et al., 2018). However, it has a limited amount of aggregated filler particles and high in viscosity, which causes difficulty in mixing the fillers and results a non-ideal filler loading. Several experiments have been carried out to overcome the shortcomings of Bis-GMA and introducing the triethylene glycol dimethacrylate (TEGDMA) as a viscosity diluent to reduce the viscosity. The combination of two matrix monomers; Bis-GMA and TEGDMA (Figure 2.2) has become the basis of the current composite resins and is widely used.

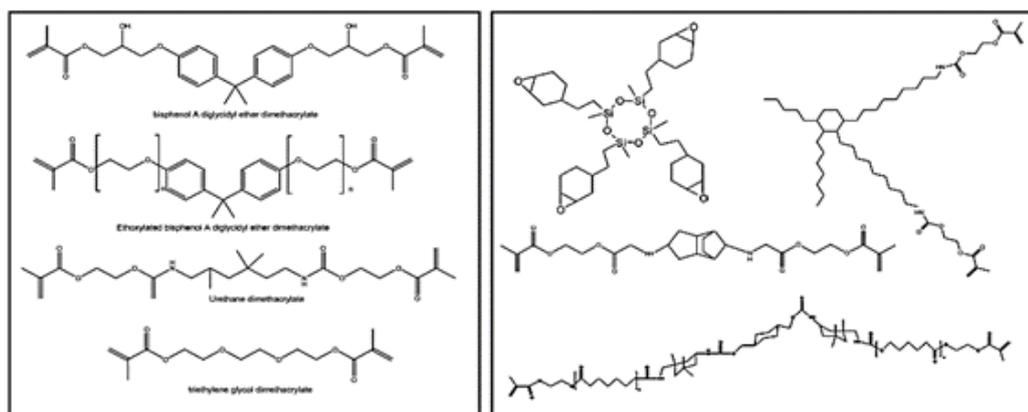


Figure 2.2 Molecular structure of Bis-GMA and TEGDMA used in modern restorative materials (Adapted from Figure 1.3.2D.2 in Ferracane, J.L. & Pfeifer, C.S. (2020). *The Organic Matrix of Restorative Composites and Adhesives*. In *Biomaterials Science* (pp. 139-152). Academic Press

The integration of the two monomers creates two reactive double bonds that form a cross-link covalent bond between the polymers upon polymerisation. The cross-linking bonds has proven to improve the composite resin's physical and mechanical properties (Kalotra et al., 2020; Tarle et al., 2012).

2.2.2 Inorganic fillers

Apart from the resin monomer, various inorganic fillers such as silicon dioxide (SiO₂), aluminium silicate, glass, quartz and lithium aluminium silicate have been widely employed as the main property of measurement parameters. The ability of inorganic fillers to correspond with the resin monomer plays a major role in providing a good stability, improving the mechanical properties, biocompatible and easy to manipulate (Habib et al., 2016).

Despite enhancing the physical and mechanical properties, incorporation between resin matrix and inorganic fillers reinforced the durability in terms of the strength, elasticity and toughness. The increase of filler (generally 50%-85% by weight) has proven to reduce the degree of polymerisation shrinkage stress, thermal expansion and water sorption. In addition, inorganic fillers control the viscosity of the resin which allows proper handling as well as providing a radiopacity effect (Habib et al., 2016; Blatz et al., 2019).

2.2.3 Polymerisation of methacrylate resin

Generally, composite resins were produced from a linear molecular monomer that ends with a methacrylate group. The formation of polymerisation chains is important for the monomers converting process into polymer molecular structures. The polymerisation process will convert the molecular structure in three phases; initiation, propagation and termination (Figure 2.3). Upon photoinitiated polymerisation of

methacrylate based composite resin, free radical (R) was generated to convert the carbon double bonds (C=C bonds) into a single bond (C-C bonds) in between the molecular structure. Interaction between radical and alkene groups of methacrylates will denote an electron which reaches the opposite end of other methacrylate molecule to produce a whole chain of free radical. Interaction between the two radical chains will then polymerise/cure the uncured resin up to the maximum level of conversion. Increment of polymerised resin is needed to complete the polymerised process and was measured as a Degree of Conversion (DOC) (Pratap et al., 2019).

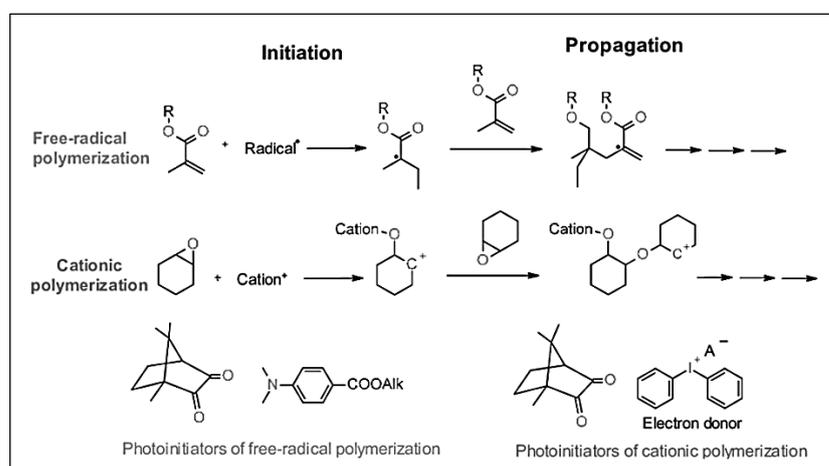


Figure 2.3 Mechanism of free radical polymerisation with corresponding photoinitiation systems. Adapted from Figure 2 in Topa, M., & Ortyl, J. (2020). Moving towards a finer way of light-cured resin-based restorative dental materials: Recent advances in photoinitiating systems based on iodonium salts. *Materials*, 13(18), 4093

2.2.4 Silane coupling agent

Adhesion between the inorganic filler and the organic resin matrix is a determinant factor for a successful clinical performance. To accomplish a good bond in methacrylate resin, the use of coupling agent compound particularly organic silicone (Si) compound known as silane coupling agent is needed to form a covalent bond with silica (inorganic fillers) and methacrylate matrix (organic resin) (Figure 2.4). These covalent bonds produce a significant effect due to the ability in performing a reversible

reaction to the chemical bond hydrolysis between both filler and resin matrix (Pratap et al., 2019; Ronald L. & Powers, 2012).

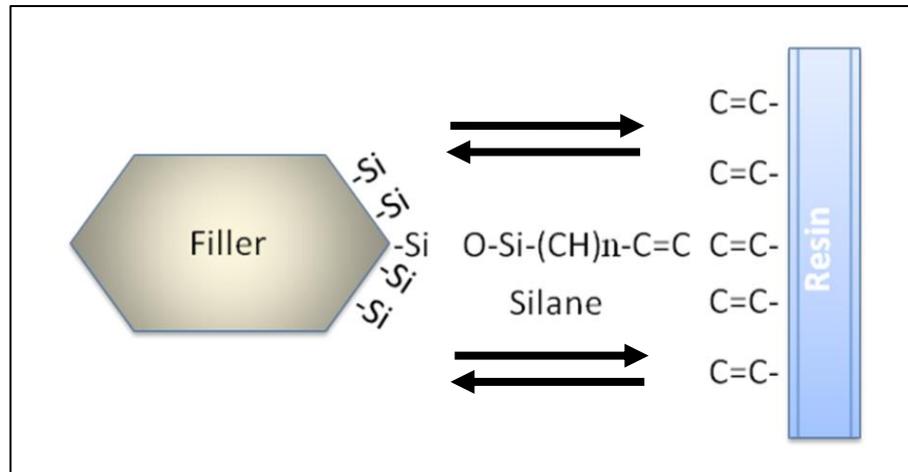


Figure 2.4 Schematic diagram of reversible covalent bonds between the filler and resin matrix.

2.3 Current classification based on filler distribution

Ever since composite resins were introduced by Dr. Rafael Bowen in 1962, clinicians, researchers and manufacturers have been trying to find a way to classify the composite resin. Established classification is important to identify and justify the use of material for therapeutic purposes (Ravi et al., 2013). Lutz and Phillips (1983) had introduced the first composite resin classification system based on the average size of filler particles and their chemical composition as well as the clinical usage (Table 2.1). Among the various classifications introduced, the most current classification system used is relatively related to the filler particles size and they are divided into six (6) main groups; macrofilled; microfilled; hybrid; microhybrid; nanofilled; and nanohybrid (Cangul & Adiguzel, 2017; Sadeghi et al., 2010) as shown in Table 2.2.

Table 2.1 Classification of composite resins and their indications for clinical use (Rawls & Whang, 2019)

Class of composite	Particle Size	Clinical use
Traditional (large particle)	1- to 50- μm glass or silica	High-stress areas
Hybrid (large particle)	1- to 20- μm glass & 40-nm silica	High-stress areas requiring improved polishability (Classes I, II, III, IV)
Hybrid (midfilled)	0.1- to 10- μm glass & 40-nm silica	High-stress areas requiring improved polishability (Classes III, IV)
Hybrid (minifilled/SPF*)	0.1- to 2- μm glass & 40-nm silica	Moderate-stress areas requiring optimal polishability (Classes III, IV)
Nanohybrid	0.1- to 2- μm glass or resin microparticles & ≤ 100 -nm nanoparticles	Moderate-stress areas requiring optimal polishability (Classes III, IV)
Packable hybrid	Midfilled/minifilled hybrid, but with lower filler fraction	Situations where improved condensability is needed (Classes I, II)
Flowable hybrid	Midfilled hybrid with finer particle size distribution	Situations where improved flow is needed and/or where access is difficult (Class II)
Homogeneous microfilled	40-nm silica	Low-stress and subgingival areas that require a high luster and polish
Heterogeneous microfilled	40-nm silica & Prepolymerised resin particles containing 40-nm silica	Low-stress and subgingival areas where reduced shrinkage is essential
Nanofilled composites	<100-nm silica or zirconia Homogeneous independent nanoparticles or nanoclusters	Anterior and noncontact posterior areas

*SPF: Small Particle-Filled

Table 2.2 Current classification of composites according to filler size and particle percentage

Type of Composite	Filler Size	Particle Percentage
Macrofilled	10-100 μm	70-80%
Microfilled	0.01-0.1 μm	75-85%
Hybrid	0.04-1 μm	35-60%
Microhybrid	0.4-0.8 μm	56-66%
Nanofilled	0.005-0.01 μm	75-80%
Nanohybrid	0.05-0.3 μm	30-55%

2.3.1 Macrofilled composites

The macrofilled composites were named based on the large size of its filler particles, ranging from 10-100 μm . Macrofilled composites were chemically activated material and considered the hardest composite restorative materials due to their crystalline quartz fillers. However, macrofilled composites may potentially abrade the opposing tooth structure and were hard to polish. In addition, easy detachment of the softer polymers leads to exposure of the hard-crystalline quartz resulting in a higher surface roughness and prone to discolouration. Initially, macrofilled composites were developed to restore the anterior teeth, but due to their poor mechanical properties, it is no longer used (Ilie & Hickel, 2011). In general, macrofill composites have good wear resistance and compressive strength, making them ideal for areas with significant occlusal forces. Larger particles additionally facilitate easy handling and manipulation. However, obtaining a highly polished and glossy surface has become challenging because of these large particles. This could have an impact on how it appears and restrict its application in anterior restorations. Larger filler particle sizes may produce a rougher surface texture than in other composites, though it may also contribute to strong mechanical qualities.

2.3.2 Microfilled composite

Microfilled composites were developed to overcome the drawbacks of the macrofilled composites for a better polishability aesthetic material. Since the average size of their filler particles are tiny (ranging from 0.01 to 0.1 μm), microfilled composites produced a smoother surface, greater shading stability and smaller marginal discolouration. Besides, microfilled composites are more translucent and suitable to reproduce the enamel layer for the anterior teeth. Microfilled composites also possessed high resistance to wear and lower flexural strength compared to other

resins (Poggio et al., 2018). Generally, the stiffness and strength of microfilled composites were slightly lower than macrofilled composites due to their lower filler content, thus limit their usage to non-stress-bearing areas (Ferracane, 2011). Microfilled composites are the great option for visible region of the mouth due to their excellent cosmetic features, polishability, and ease of manipulation. Microfilled composites are particularly popular for anterior restorations when aesthetic considerations are crucial, as they can create a high gloss and smooth surface with a more translucent and natural appearance. However, their low mechanical strength prevents them from being utilized in high-stress areas solely due to their significantly smaller particle size.

2.3.3 Hybrid composites

About 60% of the hybrid composites are composed of barium glass (inorganic phase) with the average size of filler particles between 0.04 to 1.0 μm . The so-called hybrid characters are obtained from the incorporation of both microfilled (0.01-0.1 μm) and macrofilled (10-100 μm) composites particles with about 70-80% increment in the fillers quantity (Murariu et al., 2020). In addition, the combination between the polymer groups (organic phase) and glass enhances the physical and mechanical properties. Besides improvement in the handling protocols and reduction of the stickiness (Al-Qarni et al., 2021), hybrid composites offer a wide range of colour selection, opaqueness and more translucent, resembles the dental structure, low polymerisation shrinkage, low water absorption, polishable, wear resistance and similar thermal expansion coefficient to natural tooth that make them suitable to restore the posterior teeth (Y. Wang et al., 2021). The hybrid composites are more versatile than macrofill and microfill composites because they represent an advancement in composite technology. Hybrid composites have a balanced qualities

that make them flexible, easy manipulation, adaptable to cavity preparation, and suitable with both thick and thin layers. Because of their balanced qualities, they are therefore frequently utilised in modern dentistry for restorative procedures that require both strength and aesthetic appeal.

2.3.4 Microhybrid composites

Ongoing study related to microparticles has succeeded in producing another subdivision of hybrid resin known as microhybrid composite resins. Due to its smaller particle size which is less than 1 μ m, microhybrid composites are more transparent, polishable to resemble the enamel of anterior teeth and have a higher load ratio and matrix to enhance the mechanical properties upon restoring the posterior teeth (Ferooz et al., 2020). The ability and success of microhybrid resin to restore anterior and posterior teeth, has ennobled it as a "universal" composite material for more than a decade (Murariu et al., 2020). Microhybrid composites offer a reasonable compromise between strength and aesthetic alternatives for a range of anterior and posterior dental restorations due to diverse combination of filler particle sizes. Most dental professionals recently consider microhybrid composite as a dependable option because of its balanced performance and versatility.

2.3.5 Nanofilled composites

The existence of nanotechnology has encouraged continuous research to improve the characteristics of existing composite resins. The development of nanofilled composite resins is mainly based on the desire to produce a nano-sized particles resins that is capable to transform the material structure. This structural transformation is expected to improve its mechanical, chemical, excellent optical features and low polymerisation shrinkage, thus develop a functional resin composite that can optimally be used in both anterior and posterior regions (El Shahawi & El

Batanony, 2020; Krämer et al., 2014). Due to nanometer-sized fillers (0.005-0.01 μm), nanofilled composites are found to be more translucent and have an excellent surface-to-volume ratio (Naguib et al., 2022). Excellent visual characteristics and strong mechanical properties provided by this special combination allow it to be chosen as the ideal option for both functional and esthetic restorations. It is because they can achieve higher degrees of polishing than other conventional composite resins.

2.3.6 Nanohybrid composites

Although both nanofilled and nanohybrid composites are considered as nanotechnology materials, both resins represent a significant difference in terms of filler formulations. Nanofilled composites are solely composed of nano-sized filler that cluster its matrix into a large secondary particle, while nanohybrid composites combine both nanometer and micrometer-sized filler obtain from microhybrid composites (Wang R. et al., 2018; Lin et al., 2020). Recently, nanohybrid composites are the most popular aesthetic restorative material compared to others due to the improvement in filler distributions to achieve superior characteristics (Saen et al., 2016; R. Wang et al., 2018; Lin et al., 2020). By incorporating nanotechnology, the superior characteristic of composite resins has been enhanced such as being highly polishable, glossier and resistance to high-stress-bearing (Mitra et al., 2003). Nanohybrid composites combine the mechanical strength, aesthetic benefits, and all the good aspects of microfill and nanofilled composites to provide a comprehensive solution for dental restorations. Furthermore, the formulation of nanohybrid composites minimises marginal microleakage and diminishes polymerisation shrinkage, enhancing the durability and integrity of the restorations.

2.4 Mechanical and physical properties

Regardless of any dental restorative materials, sufficient mechanical integrity and physical aspects are essential to perfectly function in the oral environment. Instead of being able to last to an extended period of time, it is also expected to last for a lifetime (Demarco et al., 2023). Furthermore, studies on the mechanical and physical properties of composite resins are closely related and relevant to clinical performance. Therefore, enhancement over composite resins mechanical and physical properties would likely enhance its lifespan and longevity. Due to its superior mechanical and physical properties, composite resins have been known as a one of the versatile restorative materials and are widely used.

2.4.1 Microhardness

The ability to resist penetration and indentation of hard objects by composite resin is the main aspect in determining the mechanical properties. Generally, the microhardness of the composite resin is closely related to the long-term stability either in the oral or clinical environment over the abrasive wear resistance and durability that reflects the overall performance of the restorative material (Alzraikat et al., 2018). There are various factors that attribute to the microhardness of the composite resins. The volume of filler particles such as glass or silica that incorporated into the resin matrix often reported to positively influence and contribute to the overall microhardness. In fact, the size of the filler particles is also reported to enhance the microhardness. Apart from the composition of the filler, the degree of curing or polymerisation of composite resins is typically crucial to obtain an optimal hardness (Farias Pontes et al., 2013; Kaminedi et al., 2014; Alzraikat et al., 2018; Alshabib et al., 2019). Microhardness is the important characteristic of composite resins that shows how resistant the material towards deformation or surface indentation. Additionally,

microhardness has a crucial role in determining wear resistance and durability, particularly in load-bearing surfaces where chewing forces are high. Thus, the right combination of fillers in terms of both quantity and types has a big impact on microhardness.

2.4.2 Colour stability

In order to function as an aesthetic restorative material, composite resins need to maintain their colours to blend with the natural teeth even after immediate polymerisation but also throughout a long-term functional period. The unacceptable clinical colour changes and failure to retain the initial colour were reported as primary causes especially for anterior restoration replacement (Alkhadim et al., 2020). Exposure to staining dietary substances and beverages such as tea and coffee has reported to cause the extrinsic surface staining by affecting the perceived colour of the restorations (Bahbishi et al., 2020). One of the most important qualities of composite resins in dentistry is colour stability. In order to guarantee that dental restorations blend in seamlessly with natural teeth and continue to be aesthetically acceptable over time, colour stability must be maintained.

2.4.3 Surface texture

Other physical conditions that affect the composite resin performance as an aesthetic restoration is the surface texture. The effect of composite surface texture deterioration is closely related to their inorganic fillers. Hence, the ability to maintain the smoothness of the restoration surface is very important as it can reduce the accumulation of extrinsic staining. Furthermore, smoother restoration surface also provides a comfortable feeling to the patient's tongue when in contact with the restoration (Al-Ghamdi et al., 2018; Bahbishi et al., 2020). The success and durability

of dental restorations are significantly influenced by the surface texture of composite resins. Additionally, a smoother surface texture minimises the chance of surface microcracks and chipping, which can reduce wear and degradation and lengthen the restoration's lifespan.

2.5 Mechanical and physical testing methods

The mechanical and physical testings of composite resins in dentistry is vital. The purpose of conducting a relevant testing method is important to determine the material properties, quality and application that complies with the requirements in accordance with the standard specifications (Saba et al., 2018).

2.5.1 Scanning Electron Microscopy (SEM)

In the early 1950's, the Scanning Electron Microscopy (SEM) has been developed to allow variety of specimens' evaluations and analyses by the researchers. SEM is a versatile and powerful analytical method to a modern science. The use of SEM as a test method has many advantages over traditional microscopes. Unlike traditional microscopes that use light to produce an image, SEM is extensively equipped with a focused beam of electrons lens. This highly focused lens is able to scan and analyse a wide range of specimen morphology, topography and composition at a high magnification degree (approximately 1 nm) even on a small spot and generate a high-resolution detailed image (Ahmed W & Jackson M.J, 2014; Akhtar et al., 2018). The special detector of the SEM is not only capable in detecting a scattered high-energy electron emitted on the surface but also able to distinguish the difference of the specimens (Figure 2.5). The optimisation of some of the SEM variables such as voltage acceleration, objective aperture size, multiple working distances, focus and alignment results in the best quality images (Akhtar et al., 2018). As such, SEM is considered a

relatively relevant, inexpensive, and essentially non-destructive surface analysis technique.

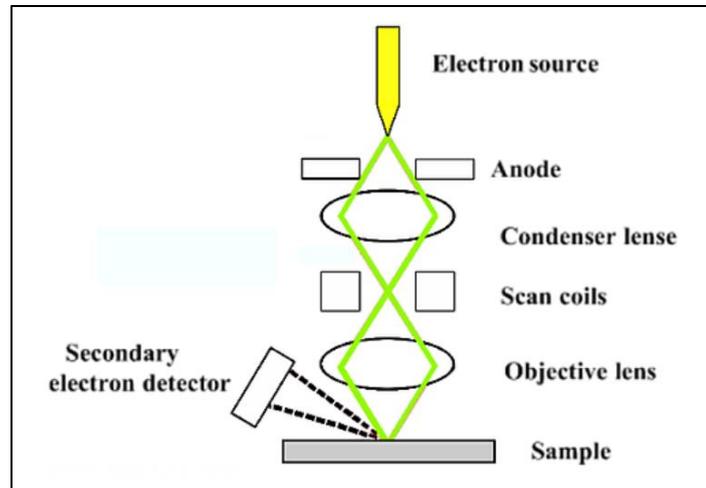


Figure 2.5 Schematic diagram of SEM working principle. Adapted from <https://www.tribonet.org/wiki/scanning-electron-microscope/>

2.5.2 Vickers hardness test

Vickers hardness test or generally referred as microhardness test is used to measure the indentation load resistance. The Vickers test has been employed as one of the rudimentary and authentic hardness tests for almost any types of materials including composite resins. Since the Vickers test is based on an optical measurement system, indentation hardness value is obtained by measuring the depth of indentation area. With the help of diamond indenter and certain specific light loads ranging from 10 gm to 1 kgf, a square base pyramid diamond indentation shaped is created (Figure 2.6). The smaller the shape, the harder the material. The Vickers test is widely used because it provides a universal indentation shape with less calibration time. Due to its micro-sized indentation, wastage of test samples can be reduced and used for other purposes (Kampouris et al., 2022). Basically, the hardness of a material can be translated through a measurement of the structural integrity and deformation resistance after continuous stress exposure.