CHARACTERIZATION OF FIBER REINFORCED NANOHYBRID DENTAL COMPOSITES FROM AGRICULTURAL BIOWASTE USING KENAF FIBER

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CHARACTERIZATION OF FIBER REINFORCED NANOHYBRID DENTAL COMPOSITES FROM AGRICULTURAL BIOWASTE USING KENAF FIBER

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LIST OF ABBREVIATIONS

ADA	American Dental Association
ANOVA	Analysis of variance
GIC	Glass ionomer cement
Corp	Corporation
CR	Composite resin
CS	Compressive strength
CTE	Co-efficient of thermal expansion
FS	Flexural strength
FTIR	Fourier transform infra-red
HA	Hydroxyapatite
HA/ZrO2	Hydroxyapatite/zirconia
HEMA	2-hydroxyethyl methacrylate
FRC	Fiber reinforced composites
PAA	Poly acrylic acid
RMGIC	Resin modified glass ionomer cement
rpm	Revolutions per minutes
SD	Standard deviation
SEM	Scanning electron microscope
SEM/EDX	Scanning electron microscope/Energy dispersive x-ray
sol	Solution
SR	Surface roughness
WHO	World Health Organization

PENCIRIAN KOMPOSIT GIGI NANOHYBRID DARI SISA BUANGAN PERTANIAN BERTETULANG GENTIAN MENGGUNAKAN SERAT KENAF.

ABSTRAK

Latar belakang: Komposit bertetulang gentian diperkenalkan seiring dengan perkembangan baru di dalam bidang biobahan, kemajuan teknologi, dan pendekatan rawatan yang lebih efektif. **Tujuan:** Kajian ini bertujuan untuk menentukan mekanisma ikatan antara serat kenaf dan komposit gigi nanohybrid yang berasal dari bahan buangan agrikultur. Kekuatan lenturan dan mampatan komposit bertetulang gentian kenaf juga dinilai. Ciri-ciri retakan permukaan dari spesimen lenturan turut dinilai. **Bahan dan kaedah:** Penyediaan komposit bertetulang serat kenaf dilakukan menggunakan silika dari sekam padi dan selulosa kenaf terawat. Selulosa kenaf diproses dan dirawat menggunakan gel sol tetraethyl orthosilicate (TEOS). Spektroskopi inframerah transformasi Fourier (FTIR) dan Mikroskop electron imbasan (SEM) digunakan untuk meneliti selulosa kenaf terawat. Enam specimen disediakan bagi setiap kumpulan; kumpulan komposit A, B, C, D, E, F, G masing-masing mengandungi selulosa kenaf 0%, 1% tidak terawat, 2% tidak terawat, 1% terawat, 2% terawat dan komersil komposit Neofil serta ever-X. Untuk ujian kekuatan lenturan dan mampatan, specimen disediakan menggunakan acuan keluli tahan karat berukuran 6mm x 4mm dan 25mm x 2mm x 2mm. *Light curing* dikenakan ke atas specimen menggunakan unit *Light cure* (Elipar Deepcure L, 3M, USA) selama 40 saat dan seterusnya diuji menggunakan Mesin Ujian Universal Instron (Shimadzu, Jepun). SEM digunakan untuk meneliti kekuatan lenturan patah. **Hasil:** SEM menunjukkan purata diameter bagi selulosa kenaf terawat adalah 7.4µm manakala purata panjang selulosa kenaf terawat adalah 537µm. Hasil keputusan FTIR menunjukkan pembentukan ikatan kimia antara selulosa kenaf dan silika dari gel sol tetraethyl orthosilicate (TEOS). Untuk ujian kekuatan lenturan dan mampatan, ANOVA satu arah menunjukkan terdapat perbezaan yang signifikan secara statistik ($P < 0.05$) antara semua kumpulan. Komposit nanohibrid yang mengandungi selulosa kenaf yang terawat menghasilkan kekuatan mampatan yang setanding dengan komposit Neofil komersial namun kekuatan lenturan adalah lebih rendah daripada komposit komersial. Analisa SEM mendapati permukaan serat adalah lebih halus dengan pengurangan lompang dan jurang di antara matrik dan serat kenaf, menunjukkan terdapat peningkatan ikatan di antara permukaan. **Kesimpulan:** Rawatan permukaan selulosa kenaf meningkatkan mekanisma ikatan di dalam komposit nanohybrid bertetulang kenaf. Adaptasi di antara permukaan serat kenaf dan matrik komposit telah menyumbang kepada peningkatan kekuatan mampatan komposit bertetulang serat. Namun begitu, tiada peningkatan ketara dari aspek kekuatan lenturan.

Kata kunci: Komposit bertetulang gentian, Kenaf, Kekuatan lenturan, Kekuatan mampatan

CHARACTERIZATION OF FIBER REINFORCED NANOHYBRID DENTAL COMPOSITES FROM AGRICULTURAL BIOWASTE USING KENAF FIBER.

ABSTRACT

Background: Fiber reinforced composite was introduced due to development of new biomaterials, technological advancements, and more effective treatment approaches. **Aims of the study:** This study aimed to determine the bonding mechanism between kenaf fibers and nanohybrid dental composite derived from agricultural biowaste. The kenaf fiber reinforced composite was investigated for flexural and compressive strength. The features of fractured surface of the flexural specimens were also assessed. **Materials and method:** Preparation of kenaf fiber reinforced composite was done using the treated silica from rice husk and kenaf cellulose. The kenaf cellulose was processed and treated using tetraethyl orthosilicate (TEOS) sol gel. Fourier- transform infrared spectroscopy (FTIR) and scanning electron microscopy (SEM) were used to investigate the treated kenaf cellulose. Six specimens were prepared for each group; composite groups A, B, C, D, E, F, G with 0%, 1% untreated, 2% untreated, 1% treated, 2% treated kenaf cellulose, Neofil and ever-X composite respectively. For compressive and flexural strength tests, specimens were formed using stainless steel molds with dimensions of 6mm x 4mm and 25mm x 2mm x 2mm, respectively. After light curing using a light cure unit (Elipar Deep cure L, 3M, USA) for 40 seconds, the specimens were tested using an Instron Universal Testing Machine (Shimadzu, Japan). SEM was used to examine the fractured flexural strength samples. **Results:** SEM showed the average diameter of treated kenaf fibers was 7.4µm whereas the average length of treated kenaf fibers was 537µm. The FTIR results suggested formation of chemical bonds between the kenaf cellulose and silica from TEOS sol gel. For flexural and compressive strength tests, one way ANOVA showed there was statistically significant difference ($P<0.05$) between all groups. The nanohybrid composite incorporated with treated kenaf cellulose resulted in comparable compressive strength with the commercial Neofil composite and no improvement for flexural strength. SEM analysis showed smoother fiber surface with presence of lesser voids and gaps between matrix and kenaf fiber which suggested enhanced interfacial bonding. **Conclusion:** Surface treatment of the kenaf cellulose improved the bonding mechanism in the kenaf reinforced nanohybrid composite. Adaptation between surface treated kenaf fiber and composite matrix contributed to improvement in compressive strength of fiber reinforced composite.

Keywords: Fiber reinforced composite, Kenaf, Flexural Strength, Compressive strength.

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Composite resins are the most common restorative materials that provide good aesthetics and good clinical results for a long time (Dionysopoulos *et al.*, 2016). The dental composite resin is a form of restorative material used to restore the decayed or broken teeth. The growing demand for aesthetic, tooth colored, and mercury free restorations has led to an increased application in resin-based composites dental materials (Chesterman *et al.*, 2017).

Dental composite consists mainly of the matrix of resin, filler particles, and coupling agents. Recent resin matrix work has resulted in the manufacturing of new materials with smaller particle size and increased filler loads. New composites can therefore have smaller particle sizes with lower filler volumes. One of the most significant developments was the of nano filled materials by combining nanometric particles and nanoclusters into a conventional resin matrix (Topcu *et al.*, 2010).

Nanohybrid dental composite is a resin composite with special nanofiller technology adding more nanoparticles in dental composite cements. The composite is intended to combine the strength of hybrid material as a single material with the shine of a microfill. The average particle size of the filler is between 0.2 and 1µm, and the organic matrix composition contains methacrylate, dimethacrylate, bisphenol A-glycidyl methacrylate (bis-GMA), triethylene glycol dimethacrylate(TEGDMA) (Noushad *et al.*, 2016).

Natural fibers have captivated massive attention in recent years as a substitute material for synthetic fibers. FRCs are composite materials characterized by three different components: the matrix (continuous phase), the fibers (dispersed phase), and the interphase region. In comparison to other structural materials, FRC materials offer a high stiffness and weight resistance, as well as acceptable toughness (Scribante *et al.*, 2018). Bast fibers such as hemp, jute, flax, kenaf, and sisal are the most common natural plant used in applications (Akil *et al.*, 2011).

Natural fibers are eco-friendly, low-cost, easy to get, and have a high specific strength and modulus. Natural fibers are harvested and graded accordingly from different parts of the plant (stem, leaf, bark, and fruits) (Thiruchitrambalam *et al.*, 2012). Kenaf is a natural (plant) fiber that is utilized as reinforcement in Polymer Matrix Composites (PMCs). Kenaf (*Hibiscus cannabinus*, L. family Malvacea) has been discovered to be a major source of fibers for composites and other industrial applications (Akil *et al.*, 2011). The quality and readiness to use with unique manufacturing processes that were never previously linked with other natural fibers is the most intriguing development of kenaf fiber reinforced composite (Thiruchitrambalam *et al.*, 2012).

Kenaf fiber is among the most common natural fibers. Kenaf is an annual hot season fiber crop which grows in temperate and tropical areas. It is like cotton, okra, and hibiscus. Kenaf is an annual plant as well as a cheap and renewable source plant due to its rapid growth. Kenaf fiber is derived from the bast stems of plants genus Hibiscus, Malvaceae family, *H. cannibinus* species. Species of *H.cannibinus* needs less water to grow, as it has a 150–180 day cycle having average yield of 1700 kg / ha (Rowell and Stout, 1998). It is a fibrous plant with inner core fiber (75–60 percent) producing pulp of less quality, and an outer bast fiber (25–40 percent) producing pulp of excellent quality in the stem. Malaysian kenaf consists of two separate fibers, bast and core, about 35% and 65% respectively. The plant rises to a height of 2.7–3.6 m and is cultivated for its fiber-producing stalks. Kenaf takes less than 6 months to achieve a size appropriate for practical use. Based on its technical and commercial potential, kenaf could be Malaysia's next industrial crop (Khalil *et al.*, 2010).

The mechanical properties of fiber reinforced composites (FRC) were thoroughly studied, with an emphasis on fracture toughness, compressive strength, load-bearing capacity, flexural strength, fatigue resistance, and fracture resistance (Scribante *et al.*, 2018). The incorporation of rigid fibers in soft matrices can lead to new materials which integrate the advantages with outstanding mechanical properties (Ben Mlik *et al.*, 2018). The fibers used in fiber-reinforced composites can be categorized according to the fiber portion, direction of fiber, whether they are pre-impregnated and the reason for which they are used. The laboratory process is simplified since no casting is required, and the bonding capacity of resin and reinforcing fiber is superior to that of resin and casting metal, whereas surface treatment of fiber is required for bonding (Bae *et al.*, 2001). The efficiency and action of any structural constituents is usually associated to their mechanical properties, such as compressive strength, tensile strength, flexural strength, impact properties and wear resistance (Akil *et al.*, 2011). Essentially, kenaf fiber reinforced composites have different tensile strength and flexural strength depending on the types of fiber, fiber direction (unidirectional or random), form and content (fabric or fiber) and the form of matrix used (Adole *et al.*, 2019)

1.2 Objectives of the study

1.2.1 General objective

To characterize fiber reinforced nanohybrid dental composite from agricultural biowaste using kenaf fiber.

1.2.2 Specific objectives

1. To determine the bonding between kenaf fibers and nanohybrid dental composite from agricultural biowaste by determining the fibers' diameter and length, and surface treatment of the fibers.
2. To evaluate flexural and compressive strength of fiber reinforced composite using kenaf fibers using Universal Testing Machine.
3. To assess the features of fractured surface, quantity of the fibers, integrity, and homogeneity along the surface of the fibers and resin using scanning electron microscopy.

1.3 Research hypotheses

1. Surface treatment of kenaf fibers will improve the bonding between the fibers and nanohybrid composite resin.
2. Incorporation of cellulose kenaf fibers has effect on the flexural and compressive strength of fiber reinforced composite using kenaf fibers as well as its surface characteristics.

1.4 Justification of study

Based on our knowledge, at this stage there is no fiber reinforced dental composite resin derived from natural fibers available in the market. Furthermore, up to this date, no report has been found on the interaction between kenaf fibers and nanohybrid dental composite from agricultural biowaste. This study aimed to determine the bonding mechanism between the kenaf fibers and composite matrix and evaluate the effect of the kenaf fibers incorporation on the mechanical characteristics of the nanohybrid dental composite derived from agricultural biowaste. In addition, scanning electron microscopy was also used to evaluate the cracked surface, fiber abundance, fiber integrity, and uniformity along the surface of the fibers and resin.

In 2014, Noushad *et al.*, (2014) developed a hybrid dental composite from rice husk at the School of Dental Sciences, Universiti Sains Malaysia, Kubang Kerian, Kelantan. We used this type of dental composite in our study because to our knowledge, at this stage there is no fiber reinforced composite resin derived from agricultural waste available in the market. In addition, we would like to enhance the characteristics of our dental composite and develop its application for clinical use. The findings from this study would be beneficial to further investigate the suitability of using this resource to produce a fiber reinforced composite resin using natural fibers and broaden its clinical applications either for direct or indirect dental restorations.

CHAPTER 2

LITERATURE REVIEW

2.1 Dental Composites

Composite resin has been used for dental restoration for nearly 50 years. The use of this material has recently gained popularity because of consumer demand for aesthetic restorations coupled with rising concerns about mercury-containing dental amalgam. More than 95 percent of all anterior teeth direct restorations and 50% of all posterior teeth direct restorations now utilize composite (Stein et al., 2005). Dental composite is a type of restoration that is aesthetically pleasing, durable, and can change the color and contour of the tooth. Many industrial dental composites have liquid dimethacrylate monomers (including BIS-GMA or variations of it) and formulations consisting of silica as inorganic filler particles.

A composite material is made up of several different physical components. "A three-dimensional combination of at least two chemically distinct materials with a separate interface separating the components," according to the definition. The bulk of dental composites usually comprise of organic matrix (continuous or polymer phase), inorganic fillers (filler or dispersed phase) and inter-phase (fillers or coupling agent). Other components such as initiators, accelerators, photo-initiators, photosensitizers, polymerization inhibitors, and UV absorbers were also added into composite resin material. The resin matrix consists mainly of bisphenol-aglycidyldimethacrylate (Bis-GMA). Since Bis-GMA is extremely viscous by itself, it is combined with short-chain monomers such as TEGDMA (triethylenglycol-dimethacrylate) in various combinations (Alrahlah, 2013). The organic matrix is based primarily on methacrylate, epoxy and polyethylene (Rodríguez-Quirós and Casanova-Yepes, 2015). The continuous phase is a cross-linked framework resulting from the monomer system; but most dental composites comprise of a mixture of different dimethacrylate monomers, like Urethane dimethacrylate (UDMA) Bisphenol-A-Glycidyl Methacrylate (BIS-GMA) and Triethylene Glycol-Dimethacrylate (TEGDMA) (Amdjadi *et al.*, 2017). Figure 2.1 shows chemical structures of Bis-GMA, TEGDMA, UDMA. Diffuse inorganic filler molecules are used to reinforce the modulus, improve the weaker polymer phase, and reduce the polymerization shrinkage. Silicon dioxide (silica), quartz, various glass powders, ceramic fillers, and other inorganic fillers are examples of inorganic fillers that can optimize the composite's hardness, wear resistance, and translucency. These particles can also serve as carriers for other materials, such as coupling agents, to be integrated (Anusavice, 2013; Nielsen, 2010).

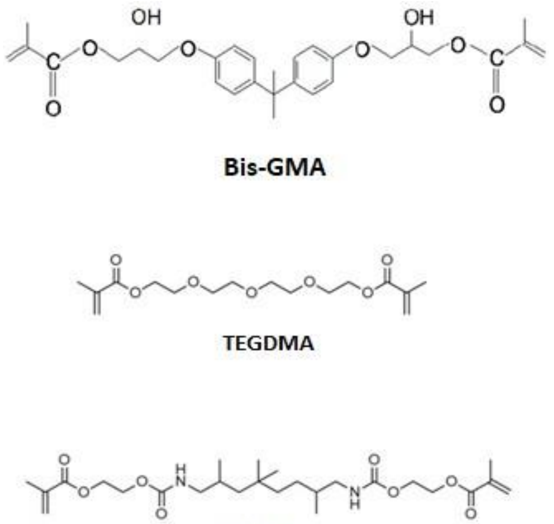


Figure 2.1 Chemical Structures of Bis-GMA, TEGDMA, UDMA

The quality of adhesion at the interface between organic and inorganic materials has a significant influence on the properties of composite materials, on mechanical strength. There are several adhesion promoters, such as zirconate, titanate and silane, which are used to improve the interactions between the filler particles and the polymeric matrix and to promote the interfacial adhesion between these two phases (Amdjadi *et al.*, 2017). The adhesion of coupling agent system typically consists of organic silanes, such as 3-methacryloxypropyltrimethoxysilane (C10H22O4Si) (3-MPS) and 10-methacryloyloxycyl dihydrogen phosphate (10MDP), whose functional chemical groups improve the adhesive strength between the reinforcing filler and the surface, as silane increases the substrate surface energy and improves surface wettability (Attia *et al.*, 2011). Coupling agents other than silane also includes titanated nanobarium titanate (NBT) filled poly (methyl methacrylate) (PMMA) (Elshereksi *et al.*, 2017). Figure 2.2 shows chemical structure of different coupling agents.

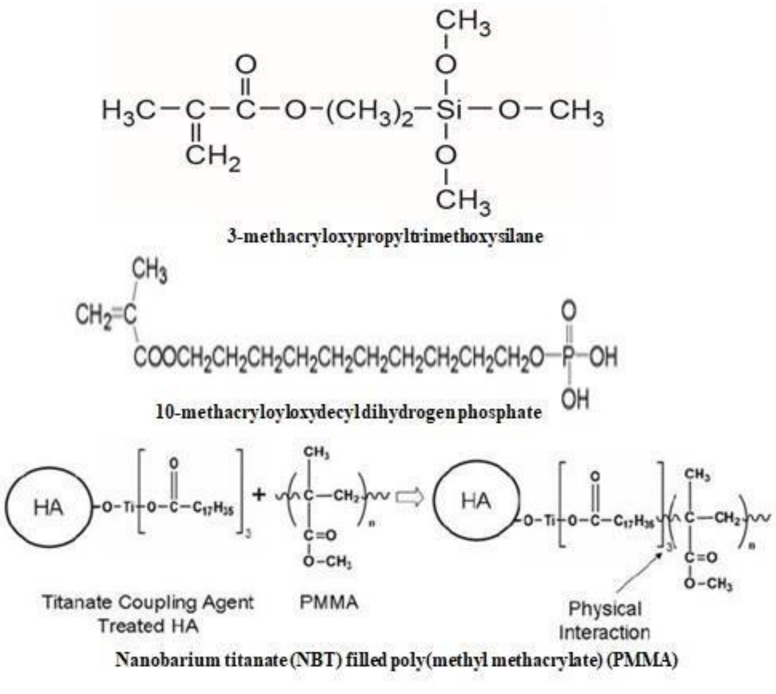


Figure 2.2 Chemical structure of different Coupling Agents.

Developments of resin composite dental restoratives have continued since they were first launched in the 1960s, with much testing being carried out to enhance the environment of the reaction and photo initiator chemistry, which is the main absorber of light in resin-based composites (RBCs). The polymerization of the composite resin begins with the release of free radicals from the methacrylate monomer structure with the inclusion of external energy in the form of thermal, chemical or radiation energy. The chemical activation resin consists of two pastes. One paste contains the activating content of benzoyl peroxide, and the other paste consists of the tertiary amine activator. The most widely used dental photo activator is Camphorquinone (CQ), which has a yellow hue resulting in a yellowish composite restorative oxide (BAPO) and diphenyl oxide (2,4,6-trimethylbenzoyl) phosphine - TPO. These photo initiators are essentially known as Norrish Type-I molecules (Albuquerque *et al.*, 2013). The light absorption that occurs in Camphorquinone varies from 425 to 495 nm (Riva and Rahman, 2019).

In recent decades, the materials of dental composites have improved to enhance biomechanical and chemical properties. The characteristics of dental composites are classified into three types. It is caused by dental resin compounds. The first category includes strength, stiffness, abrasion resistance, and coefficient of thermal expansion. The three elements of the composite are liable for these features: the filler, the matrix, and the coupling agent. In the second category, the softening tendency and color consistency is induced by the form of matrix and photo initiator. The last group is made up of polymerization shrinkage and water sorption. The last category is mainly related to the matrix (Kowalska *et al.*, 2021). The sum of the initiator depends on the nature of photosensitizer. The optimum concentration of the initiator in resin-based composites depends on several aspects, like the solubility of these compounds in the monomer, photo reactivity, color, or biocompatibility. There are two types of photo initiators: 1-type is trimethylbenzoyl-diphenylphosphineoxide (TPO) benzoyl peroxide (BPO (Santini *et al.*, 2012) and 2-type is camphorquinone (CQ), phenanthrenequinone (PQ), benzophenone (BP) and 1-phenyl-1,2 propanodione (PPD) (Santini *et al.*, 2012; Pratap *et al.*, 2019).

Similarly, polymerization inhibitors are compounds that are applied to the monomer to avoid random polymerization. Typically, as a polymerization reaction proceeds, the heat generation of thickening and polymerization occurs. There are chances of degradation and accidents if the reaction is not properly regulated. Therefore, a stabilizer polymerization inhibitor is applied to store the radical polymerizable monomer. Some of the potential inhibitors includes 1,4-naphthoquinone (NQ) and hydroquinone (Takahashi *et al.*, 2019).

The properties and performance of resin composites depend on the three basic components of the material. Some of the properties are primarily related to filler and coupling agent, while other properties are primarily related to the resin matrix (Peutzfeldt, 1997).

In dental composites, fillings are used to ensure stability, increased rigidity, reduced dimensional change on heating and cooling, reduced setting shrinkage, for radiopacity, improved aesthetics, and improved handling. Asmussen and Peutzfeldt (1998) found that the variance of the BisGMA / TEGDMA / UEDMA ratio significantly impacted the composite's mechanical properties. The physical and mechanical properties of the composites generally improve in direct relation to the amount of filler added (Ferracane, 1995). Likewise, in the intraoral series, mechanical and biological properties of composite materials affect other parameters such as thermal transition, corrosive, and mechanical loading.

The mechanical properties of resin-based composites depend essentially on their microstructure and composition. Microstructural features include the distribution of filler particles in the bulk, the morphology of those filler particles and the presence of pre-existing cracks and voids. Such attributes relate directly to the composite composition (Junior *et al.*, 2007).

Flexural strength, diametric tensile strength, compressive strength, flexural modulus, generalized wear resistance, and polymerization shrinkage are some composite properties on which material quality is significantly dependent. Flexural resistance is an important mechanical property for fragile materials. The flexural strength of a material is the highest stress it can withstand when experiencing bending load before failure (Chung *et al.*, 2004). An analysis of 72 restorative materials also showed that the amount of filler has a significant influence on mechanical properties. The relationship between the filler material, flexural strength and elasticity modulus was most evident (Ilie and Hickel, 2009).

2.2 Classification of Dental Composites

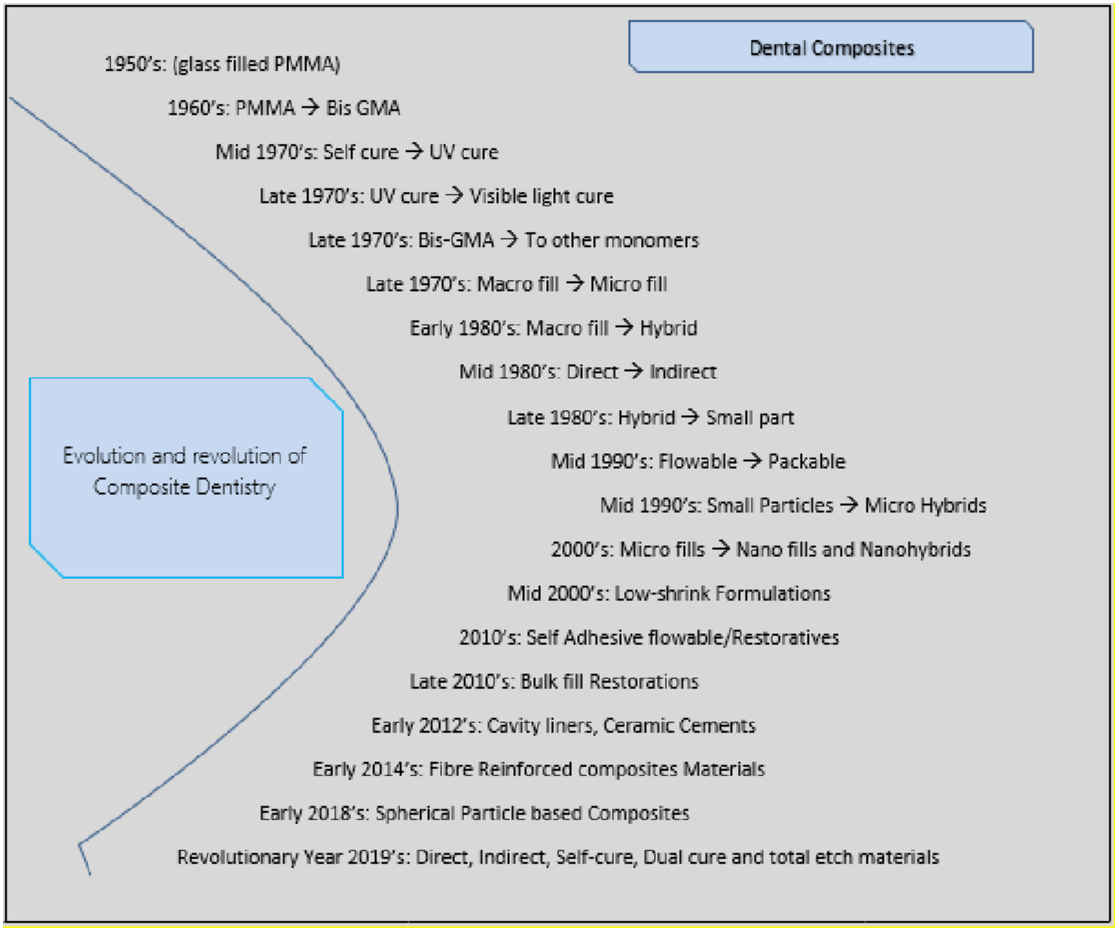


Figure 2.3 Evolution and Revolution of Dental Composites (Ferracane, 2011)

Figure 2.3 shows evolution and revolution of dental composites (Ferracane, 2011). Dental restorative composites used in dentistry are classified differently. These are classified according to the particle size of the filler, the activation process and its implementation. The average filler size used for classification of the composite resin is as follows; macro-filled or conventional composites (10 to 100 µm), midifillers (1 to 10 µm), minifillers (0.1 to 1µm), microfillers (0.01 to 0.1 µm), and more recently nanofillers (0.005 to 0.01 µm) (Mota *et al.*, 2012). Shama Bhat and Nandish (2006) listed dental composite by application such as anterior composites (cavities of class III class V); posterior composites; core builds up composites; pit and fissure composites;

prosthodontic composite resins (crowns of gold or base metal alloy); composites of glaze resin; and bonding agents (Alsharif *et al.*, 2010).

2.3 Nanohybrid Composites

The recent application of nanotechnology research to the field of dentistry has led to the development of plastic materials with more favorable mechanical properties. Composite materials have their origin in two families of materials. In addition to traditional micro-hybrid and micro-filler materials, nano-filled and nano-hybrid composites have recently been developed to provide a material with high initial polish combined with excellent polish and shine (Moraes *et al.*, 2009). In general, nanocomposites are of two kinds: nanohybrid nano fill types. Nanohybrids consist of milled glass fillers in the 40-50 nm size range of nanoparticles. On the other hand, the type of nano-filled composites is formed by a combination of; (a) Nanomers, which are monodisperse nanoscale non-aggregated silicon dioxide filler particles with a size range of 20-75 nm, and (b) nanoclusters, zirconium silica agglomerates, and silicon dioxide nanomeric particles (Sachdeva *et al.*, 2015).

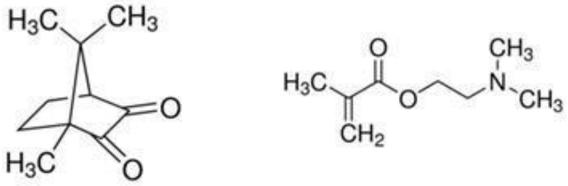
2.4 Composite resin from rice husk

Natural fiber from industrial waste is of great value in the polymer industry due to its many benefits, such as its light weight, lower cost, and environmental conservation. Rice husk (RH) is a sheath that forms around the grains of rice during their formation. As a kind of natural fiber made from agricultural waste, rice husk can be used as a filler for composite materials (Amdjadi *et al.*, 2017).

Current research on dental composites has focused more on filler quality and polymerization. Various fillers have been used in dental composites, with silica being the most common. Silica particles with different proportions, shapes and contents have been used to improve the properties of dental composites (Zakir *et al.*, 2013). At present, the silica used during the manufacturing of dental composites is produced by the sol-gel process, using toxic and costly by products and chemicals such as tetraethyl orthosilicate and high sodium silicate. Silica collected from the rice husk in various ways using several methods (Liu *et al.*, 2011). Until now, spherical nano silica has been produced from rice husks using nitric, carboxylic and orthophosphoric acids as precipitants in combination with various solvents (ethanol and 2-propanol) (Noushad *et al.*, 2012).

Silica has been optimized to have superior morphology for use as a filler in dental nanocomposites; It has already been documented that filler loading improves due to a lower surface-to-volume ratio, which improves mechanical properties (Curtis *et al.*, 2009). Studies concluded that the physico-mechanical properties and the color stability of rice husk dental composites showed comparable results to conventional dental composites (Lin *et al.*, 2022). Similarly, a study from Noushad *et al.*, (2016) concluded that it was possible to prepare nanohybrid silica particles from rice husk, with characteristics ideal for use as fillers in dental composites. The preparation procedure was simple and cost effective while in comparison, commercial composites use two or more fillers like zirconia and aluminum oxide etc in orders to increase properties whereas experimental composite with only silica from rice husk showed reasonable hardness values.

Before using silica particles as fillers, a surface treatment solution must be prepared by adding 6% by weight of silica-based γMPS to a 70/30 ethanol / water solution. Silica particles added to the treatment solution must be stirred with an overhead stirrer for approximately one hour. Camphorquinone and 2-(Dimethylamino) ethyl methacrylate (DMAEMA) are most used photo-initiator system, which later dissolves in the matrix resin (Bis-GMA/TEGDMA, 60/40 wt.%.) under sub-ambient light (Kim *et al.*, 2007). Figure 2.4 shows chemical structure of Camphorquinone and 2-(Dimethylamino)ethyl methacrylate (DMAEMA).



Camphorquinone 2-(Dimethylamino)ethyl methacrylate

Figure 2.4 Chemical structure of Camphorquinone and 2(Dimethylamino) ethylmethacrylate (DMAEMA)

2.5 Agricultural Biowastes and its sources

The agricultural sector plays an important part in global economic growth. In total, 998 million tons of agricultural waste is generated in Malaysia each year and 1.2 million tons of agricultural waste is disposed of in landfills each year (Agamuthu, 2009). It is projected that 15% of the total waste produced in Asia is agro-waste, with agricultural waste production in Malaysia expected to reach 0.210(kg / cap / day) by 2025, that was around 0.122 (kg / cap / day) in 2009 (Tahir, 2012). Agricultural waste is residue from the growth and processing of raw agricultural products as non-product outputs of manufacturing and processing and may include other useful waste materials. Such residues are produced from several agricultural activities and include farming, livestock and aquaculture (Obi *et al.*, 2016). Natural fibers are environmentally friendly, inexpensive, readily available, and have suitable specific strength and modulus. Natural fibers are harvested from different parts of the plant and classified accordingly (stem, leaf, bark, and fruit). Sisal, banana, kenaf, coir are the most commonly used plant fibers (Thiruchitrambalam *et al.*, 2012).

2.6 Kenaf fibers and its sources

Fiber plants have been in human society since the dawn of time. Kenaf (*Hibiscus cannabinus,* L. Malvacea family) has been shown to be an effective source of fiber for composites and other industrial applications. Kenaf is known as a source of cellulose with economic and environmental benefits; In 3 months (after sowing) it can reach a height of more than 3 meter and a base diameter of 3-5 cm in a wide variety of climatic conditions (Akil *et al.*, 2011).

The kenaf plant is made up of many valuable components (for example, stems, leaves, and seeds) and each has different functional parts (for example, fibers and strands, proteins, oils, and allelopathic chemicals). Many factors can affect the yield and composition of these plant components, including variety, planting date, light sensitivity, length of the growing season, plant populations, and plant maturity (Webber III and Bledsoe, 2002). In Malaysia, the Kenaf National Research and Development Program was established to develop kenaf as a potential new industrial crop for Malaysia, recognizing the many possibilities of commercially viable kenaf-derived products. As part of the Ninth Malaysia Plan (2006-2010), the government has allocated the Kenaf-based industry with RM 12 million for research and development (Jamaludin, 2008).

Kenaf has a straight, branchless stem. The kenaf stem consists of an inner woody core and an outer fibrous crust that surrounds the core. The outer fibrous bark fiber is also known as bast fiber. Kenaf bast fiber combines superior flexural strength with excellent tensile strength, making it the material of choice for a wide range of extruded, molded, and nonwoven articles (Ishak, 2007).

Habibi *et al.* (2008) clarified that cellulose is a major component of natural fibers and that the basic unit of the cellulose macromolecule is anhydrodglucose containing three hydroxyl groups (OH). The hydroxyl groups have been shown to form hydrogen bonds within the macromolecule (intramolecular) and between other cellulose (intermolecular) macromolecules, which is why the fiber is hydrophilic in nature. Chemical structure of cellulose in kenaf fiber is as shown in figure 2.5.

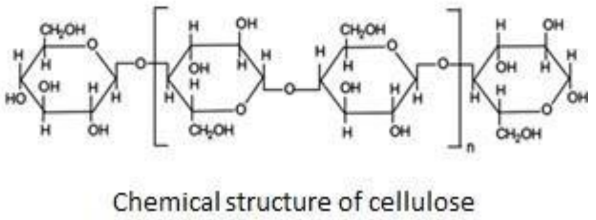


Figure 2.5 Chemical Structure of Cellulose in Kenaf fiber

Grafting enhances matrix fiber wetting by hydrophilizing the fiber surface and facilitates interfacial bonding by diffusing the grafted molecules chain segments into the matrix. Coupling agents and radical mediated adhesion improve interfacial bonding by forming covalent bond between fiber and the matrix (Aji *et al.*, 2009). Kenaf fiber is one of the assets used as reinforcement for composite materials in commercial processes due to its superior properties. The cellulose of kenaf fiber has both economic and environmental benefits, in example it can grow up to 3 m height with a base diameter of 3- 5 cm in a massive variety of climatic conditions within three months (Sgriccia *et al.*, 2008). Due to its growth period 150-180 days, kenaf fiber needs far less water to grow (Rowell and Stout, 2007). The kenaf fiber grows efficiently as it is capable of adapting to different types of soils with limited chemical modification (Ramesh *et al.*, 2018). Kenaf was found to be extremely environmentally friendly for two main reasons: (a) Kenaf accumulates carbon dioxide at a significantly high rate and (b) kenaf absorbs nitrogen and phosphorus from the soil (Mahjoub *et al.*, 2014).

2.7 Fiber reinforced Composites

Fiber Reinforced Composites (FRC) are increasingly used in dentistry to replace metal restorations: periodontal splints, partially fixed dentures, root posts, orthodontic appliances, and some other indirect restorations. In general, the FRC's offers better biomechanical efficiency for the composite structure due to its superior tensile and flexural properties. The biomechanical properties of dental materials are of great importance in dentistry, and the development of fiber-reinforced composite structures is of ongoing scientific interest (Zhang and Matinlinna, 2012). High use in aerospace, aeronautics, marine and automotive engineering has been observed for the mechanical properties of composite materials such as tensile modulus and flexural modulus, enhanced surface finish, durability of molded parts during manufacturing biodegradability and minimal health hazards. In contrast with other structural materials FRCs materials have high rigidity and strength per weight along with sufficient toughness (Saidulu and Husain, 2017). Experimental results for this study demonstrate the increase in number of layers fiber reinforcement which improves the strength.

Typical properties of FRC are high strength-to-weight and modulus weight ratios, high fatigue strength and tolerance to fatigue damage with anisotropic properties (Zhang and Matinlinna, 2012). Fiber reinforcement was introduced to increase both the flexural strength and modulus of these materials. However, due to the composite fiber interface, the reinforcing component can act as a stress concentrator (Nayar *et al.*, 2015). FRC is a blend of polymeric matrix materials and reinforcing fibers. Composite fibers are the reinforcing phases in the system when load is applied to the composite. The load is transferred to the fibers and the material becomes firm and hard. The reinforcing fibers can be unidirectional continuous (roving’s), bidirectional continuous (woven fabric), randomly oriented continuous (mat) or randomly oriented discontinuous. FRC can be isotropic, orthotropic or anisotropic, meaning that the material properties and the direction of the fibers depend on mechanical, optical, curing shrinkage and thermal properties of the FRC depend on the fiber quantity and orientation (Vallittu, 2018).

2.8 Properties of fiber reinforced Composites

The efficiency of the material is always shown in its mechanical properties such as tensile properties, flexural properties, friction properties, impact properties and wear behavior. In recent years, several studies have been conducted on kenaf fiber reinforced composites to explain their mechanical performance. Kenaf fiber compression, bending, and tensile strength depend on the fiber category, its orientation (random or unidirectional), the fiber type, the treatment method, the fiber length, and the fiber content (Abbas et al., 2022). These properties are important for determining the quality of the material, especially in difficult and critical conditions, which are directly related to durability._Flexural module represents matrix and fiber's ability to transfer elastic deformation and restrain the strains to prevent interface fracture (Zhafer *et al.*, 2016).

According to ISO guidelines, flexural strength and flexural modulus can be measured by a three-point flexural test (Ilie and Hickel, 2009). A universal testing machine (Zwick 1435, Ulm, Germany) and a 50 N force measuring device (type: KAPS; Angewandte SystemTechnik, Dresden, Germany) were used to measure the test bar at a distance of 20 mm and a constant speed of the crosshead of 1 mm / minute. Sample measurements were made with a precise ± 10 µm digital electronic caliper (Piwowarczyk and Lauer, 2003).

Compression is the composite material's ability to withstand the load which tends to resize it. The work included compressive test, which was carried out at room temperature according to the ASTM D695 test method. The sample was tested with the Instron 4201 universal testing machine with a crosshead speed of 3mm / min (Fairuz *et al.*, 2016).

2.9 Surface treatment of Kenaf Fiber

Biofibers are naturally cellulose and consist of cellulose, hemicellulose, lignin, and pectin. Cellulose and lignin are the main components of natural fibers (Hao *et al.*, 2018). Improving natural fibers in different conditions, i.e., through different chemical treatments of fibers and hybridization with synthetic fibers, considerable changes in the performance of polymeric compounds can be achieved (Dittenber and GangaRao, 2012). It is important to impart hydrophobicity to the fibers by surface treatment to produce composites with improved mechanical strength (Ali *et al.*, 2018). Surface treatments are also used to improve the efficiency of natural fiber reinforced composites by integrating the difference in compatibility between hydrophilic fibers and hydrophobic matrices (Mittal *et al.*, 2016). A variety of experiments have been performed to improve the properties of natural fiber reinforced composites (NFRCs) using different chemical treatments. A study by Bakar *et al.* (2010) showed that alkali-treated fiber-reinforced composite materials showed an improvement in mechanical properties, in particular flexural strength and flexural modulus. The results of this study indicated that flexural strength and flexural modulus increased approximately 24% and 83%, respectively, with a fiber load of 25% by weight. In the case of composites with a fiber load of 5%, a slightly lower value was observed for flexural strength and flexural modulus. Due to lower fiber content, a weak interface was created due to poor fiber wetting in the matrix (Bakar *et al.*, 2010).

Atika *et al.* (2014) recorded a three-hour treatment of kenaf fibers with 6% sodium hydroxide solution (NaOH) through the mercerization process. Mercerization reduces the diameter of the fiber and removes the lignin, hemicellulose, waxes and oils that cover the surface of the fiber. Maximum flexural, tensile and impact strengths were achieved from an unsaturated polyester reinforced (UPE) hybrid composite with treated kenaf fibers (Atiqah *et al.*, 2014). Similarly, previous studies on the morphological, physical and mechanical properties of natural fibers and subsequent woven composites found that kenaf fibers have improved tensile properties than betel palm fibers due to their higher cellulose content (Lai and Mariatti, 2008), (Krishna and Kanny, 2016). From morphological findings, they observed that the alkaline treatment of the fibers successfully cleans and improves the surface of the fibers. The alkali treatment also results in fiber fibrillation, which increases the efficient surface area required for wetting the matrix resin and thus improves the mechanical characteristics of the resin (Ben Mlik *et al.*, 2018).

2.10 Bonding mechanism Between kenaf and polymer matrix

There are several important limitations to the effective use of natural fibers in dental composites. These include high susceptibility to moisture absorption and an intrinsic imbalance of natural fibers with certain nonpolar polymer matrices, all of which can lead to composite materials with unfavorable mechanical characteristics (Brodowsky and Mäder, 2012). The lack of good interfacial adhesion between the two components (kenaf fibers and polymeric matrix) and water absorption are the main