

**EVALUATION OF PHYSICAL AND  
MECHANICAL PROPERTIES OF ZIRCONIA  
AND ALUMINA REINFORCED EXPERIMENTAL  
NANOHYBRID DENTAL LUTING COMPOSITE  
CEMENT USING SILICA FROM RICE HUSK**

**MD. SAIFUL ISLAM**

**UNIVERSITI SAINS MALAYSIA**

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by

**MD. SAIFUL ISLAM**

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for the degree of  
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## LIST OF ABBREVIATIONS

379 GPa	alumina with its higher elastic modulus
$\gamma$ -MPS	3- methacryloxypropyltrimethoxysilane
3Y-TZP)	yttrium cation-doped tetragonal zirconia polycrystals
ADA	American dental association
ANSI	American national standards institutes
AFM	atomic force microscopy
AL <sub>2</sub> O <sub>3</sub>	aluminium oxide is the solid oxide form of aluminium
AL <sub>2</sub> O <sub>3</sub>	nano-sized alumina
Bis-GMA	Bisphenol A Glycidyl methacrylate
BTDMA	benzophenone tetracarboxylic dianhydride
CEMA	N-cyanoethyl-methylaniline
CQ	camphoroquinone
DMA	dynamic mechanical analysis
DMAEMA	dimethylamino ethylmethacrylate
DMPT	N-dimethyl-p-toludine
EDX	energy dispersive x-ray
Filtek Z250TM /3M ESPE	conventional micro-hybrid composite
FTIR	Fourier transform infrared
ISO	International Organization for Standardization
Mg-PSZ	magnesium cation-doped partially stabilized zirconia
MTA	mineral trioxide aggregate
MTS	methyltrimethoxysilane
NHDLCC	nanohybrid dental luting composite cement
RH	Rice husk

RHA	Rice husk ash
RMGIC	resin modified glass ionomer cement
SEM	scanning electron microscope
SiO <sub>2</sub>	silicon dioxide
Si-OH	silanol
SSS	sodium silicate solution
STM	scanning tunneling microscopy
TEGDMA	Triethylene glycol dimethacrylate
TEM	Transmission electron microscope
TEOS	tetrathoxysilane
UDMA	urethane dimethacrylate
γ-MPS	3- Trimethoxysilyl propyl methacrylate
ZrO <sub>2</sub>	zirconia
ZTA	zirconia toughened alumina

**PENILAIAN SIFAT FIZIKAL DAN MEKANIKAL SIMEN LEKAT  
KOMPOSIT NANOHIRID PERGIGIAN EKSPERIMENTAL YANG  
DIPERKUAT OLEH ZIRKONIA DAN ALUMINA MENGGUNAKAN  
SILIKA DARI SEKAM PADI**

**ABSTRAK**

**Tujuan:** Kajian ini bertujuan untuk mengesktrak silika daripada sekam padi, kemudian mencirikan pengisi silika yang disintesis, zirkonia dan alumina komersial untuk menghasilkan simen lekat nanohibrid pergigian (NHDLCC) eksperimental. NHDLCC eksperimental yang diperkukuh dengan zirkonia dan alumina telah dinilai melalui ujian kekuatan mampatan, kekuatan lenturan, kelikatan, penyerapan air dan kelarutan. **Bahan dan kaedah:** Nano silika daripada sekam padi tanpa silana dicirikan menggunakan mikroskop elektron imbasan (SEM) dilengkapi dengan sinaran-X penyebar tenaga (EDX). Pengisi silika, zirkonia dan alumina menjalani rawatan permukaan menggunakan agen gandingan silana. Kemudian, kesemua pengisi yang mempunyai silana dan tanpa silana dicirikan menggunakan spektroskopi inframerah transformasi Fourier (FTIR). Tiga kumpulan eksperimen telah dibuat berdasarkan jenis pengisi yang diperkukuh yang berlainan: kumpulan 1 (3 berat % zirkonia); kumpulan 2 (3 berat % alumina); kumpulan 3 (3 berat % zirkonia and 2 berat % alumina) dan NHDLCC eksperimental tanpa pengukuhan zirkonia dan alumina digunakan sebagai kawalan negatif dan simen lekat komposit nanohibrid pergigian komersial, manakala simen lekat Rely-X™ U200 (3M ESPE; USA) digunakan sebagai kawalan positif. Kemudian, kesemua kumpulan eksperimental dicirikan menggunakan SEM yang dilengkapi dengan EDX dan menjalani penilaian sifat fizikal dan mekanikal yang merangkumi kekuatan mampatan, kekuatan lenturan, kelikatan,

penyerapan air dan kelarutan. ANOVA sehalu digunakan untuk perbandingan kumpulan berganda diikuti dengan post hoc (Bonferroni). Tahap signifikan secara statistik ditetapkan pada  $p=0.05$ . **Keputusan:** SEM and EDX mengesahkan pengisi berbentuk sfera yang diagihkan secara homogen dan kehadiran zarah pengisi dalam setiap kumpulan NHDLCC eksperimental. Kekuatan mampatan kumpulan pengisi 3 berat % didapati secara signifikan jauh lebih tinggi berbanding dengan kawalan negatif ( $p < 0.05$ ). Sementara itu, untuk kekuatan lenturan, penyerapan dan kelarutan air, kumpulan pengisi 3 berat % zirkonia and 2 berat % alumina adalah secara signifikan lebih tinggi berbanding dengan kawalan negatif ( $p < 0.05$ ). Untuk pengujian kelikatan, kumpulan pengisi 3 berat % alumina menunjukkan peningkatan kelikatan yang signifikan secara statistik berbanding kawalan pada frekuensi ayunan ( $\omega$ ) 1 rad/s ( $p < 0.05$ ). Namun, tiada perbezaan signifikan secara statistik pada NHDLCC dengan frekuensi ayunan ( $\omega$ ) 10 rad/s ( $p > 0.05$ ). **Kesimpulan:** Silika berjaya diekstrak daripada sekam padi dan ketiga-tiga pengisi dicirikan sebelum dan selepas rawatan permukaan dengan agen gandingan silana. Pengukuhan zarah pengisi zirkonia dan alumina dengan peratusan yang berbeza dalam NHDLCC eksperimental menambahbaik kekuatan mampatan, kekuatan lenturan, sifat kelikatan dan penurunan penyerapan dan kelarutan air.

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**ABSTRACT**

**Aims:** This study aimed to extract silica from the rice husk, then characterised the synthesised silica and commercial zirconia and alumina fillers to fabricate experimental nanohybrid dental luting composite cement (NHDLCC). The zirconia and alumina reinforced experimental NHDLCC were subjected for the evaluation of compressive strength, flexural strength, viscosity, water sorption and solubility testings. **Materials and methods:** The unsilanated nano silica from rice husk was characterized using a scanning electron microscope (SEM) equipped with energy dispersive X-ray (EDX). Silica, zirconia and alumina fillers were subjected to a surface treatment using silane coupling agents. All the silanated and unsilanated fillers were then characterized using Fourier transform infrared spectroscopy (FTIR). Three groups were fabricated based on different types of filler reinforcement: Group 1 (3 wt.% zirconia); Group 2 (3 wt.% alumina); Group 3 (3 wt.% zirconia and 2 wt.% alumina) whereas experimental NHDLCC without zirconia and alumina reinforcement was used as a negative control and a commercial nanohybrid dental luting composite, Rely-X™ U200 luting cement (3M ESPE; USA) was used as a positive control. All the experimental groups were then characterized using (SEM) equipped with (EDX) and subjected to its mechanical and physical properties evaluation that included compressive strength, flexural strength, viscosity, water sorption and solubility. One-way ANOVA was used for multiple group comparison followed by post-hoc

(Bonferroni) A statistically significant level was set at  $p = 0.05$ . **Results:** SEM and EDX confirmed the homogeneously distributed spherical-shaped filler and the presence of filler particles in each group of experimental NHDLCC. The compressive strength of 3 wt.% zirconia reinforced filler group was found significantly higher when compared to the negative control ( $p < 0.05$ ). Meanwhile, for the flexural strength, the water sorption and solubility, 3 wt.% zirconia and 2 wt.% alumina reinforced filler group showed significantly higher compared to the negative control ( $p < 0.05$ ). For the viscosity testing, 3 wt.% alumina reinforced filler group showed statistically significant increase of viscosity compared to control at the oscillation frequency ( $\omega$ ) of 1 rad/s ( $p < 0.05$ ). However, the difference was found not statistically significant at the oscillation frequency ( $\omega$ ) of 10 rad/s ( $p > 0.05$ ) of NHDLCC. **Conclusion:** The silica was successfully extracted from the rice husk and all three fillers were characterised before and after surface treatment with the silane coupling agent. The reinforcement of different percentages of zirconia and alumina filler particles in the experimental NHDLCC improved compressive strength, flexural strength, viscosity properties, and decreased water sorption and solubility.



# CHAPTER 1

## INTRODUCTION

### 1.1 Nanotechnology

The term nano is from the Greek word Nanos which means ‘dwarf’. It is a prefix used to describe one-billionth or  $10^{-9}$  of a meter, which is roughly the size of two or three atoms. In general, nanotechnology can be specified to those areas of science and engineering that are related in the manufacture, investigation, and utilization of materials, structure and devices. Based on the improvement in empowering creations in the field of atomic force microscopy (AFM) and scanning tunneling microscopy (STM), in the earlier of 1980s, the nanotechnology have been established (Malinauskas *et al.*, 2016). Generally, it deals with the perception and control of matter with a size roughly between 1 and 100 nanometer (Singh *et al.*, 2017). Although there are numerous instances of structures that exist with nanometer measurements such as elemental particles of the human body and segments of food, and albeit various advancements in nanotechnology, it has only been in the last quarter of a century that scientists have been conceived to effectively and deliberately alter particles and structures inside this size range (Zhang, 2011).

Researchers at present are debating on the future repercussions of nanotechnology. Nanotechnology might provide the opportunity to the invention of materials and gadgets with immense scope of uses in many fields such as nanomedicine, nanoelectronics and biomaterials energy creation. However, nanotechnology raises many interchangeable issues as in other new technology, such as its potential toxicity and natural effect of nanomaterials, as well as their potential consequences for a worldwide financial burden (Iavicoli *et al.*, 2017).

Nanotechnology has been widely used in dentistry such as in preventive and diagnostic nano dentistry, nanomaterials, biotechnology, nanorobotics, nanomedicine, etc. and that will possibly ensure the preservation of nearly perfect oral health (Abiodun-Solanke *et al.*, 2014). Dental caries and the periodontal diseases are one of the most common dental problems affecting people. Different strategies to restrict and fight them have been contrived and applied since long time ago but still there need to be some consistent requirement. Nanotechnology, with its ever-expanding extension, give researchers new opportunities to go for further research (Bhavikatti *et al.*, 2014). Although researchers have endeavored to develop an effective and palatable drug delivery system where the nanoparticles are impregnated with triclosan and the use of triclosan particles into the test region lessened the aggravation (Ozak and Ozkan, 2013). The presence of nanosized calcium carbonate in toothpaste allows the remineralization of early enamel lesions (Dianti *et al.*, 2018). Nanotechnology has already achieved a great interest for the treatment purposes of different oral diseases and now its use has been reached out from treating dentin hypersensitivity, root canal disinfection to tissue engineering and drug delivery applications. Using the nanodevices, it will guide the diagnosis of an early existence of diseases, tumor cells and distinguish and evaluate the toxic molecules as well. Nanotechnology in future dentistry which will bring some constitutional changes in health care by developing an ideal method for the diagnosis and prevention, delivering drug and gene therapy, and customizing therapeutic selection according to the patient's profile (Ousey *et al.*, 2018). Nanotechnology has remodeled the restorative dentistry by adding nano filler in dental composite, because of the increasing demand for esthetic restorations and in spite of criticism and predicted failures (Abou Neel *et al.*, 2015). Nano silica as a nanofiller has been widely used in dental composite because of its size, surface area,

biocompatibility and low-density properties (Priyadarsini *et al.*, 2018). Different sizes and outline of packing of nano silica influence the materials retention and extensively utilized to enhance the physical and mechanical properties of dental cements (Bajraktarova-Valjakova *et al.*, 2018). Furthermore, in addition to treating the dentin hypersensitivity and tooth polishing purposes, nano silica has also been used for a long period of time (Vano *et al.*, 2014).

## **1.2 Silica Nanoparticles**

Renovation in nanotechnology has led to the fabrication of silica nanoparticles, SiO<sub>2</sub>, which has been popularly used in scientific research as well as in the field of engineering (Sun *et al.*, 2005). Silica or silicon dioxide (SiO<sub>2</sub>), is a colorless, white chemical compound commonly found in nature such as quartz, glass, sand, and in plants like rice, wheat, sugarcane bagasse, bamboo leaf and oil palm ash (Fernández *et al.*, 2015; Osman and Sapawe, 2019).

In the past few years, the application and demand of silica nanoparticles have been developing dramatically in the scientific field, as well as in the industrial areas, for example, catalysis, stabilizers, colours, and synthetic mechanical polishing (Wang *et al.*, 2010). From the mechanical application to the sustenance and drink industry, silica is utilized everywhere (Sigel *et al.*, 2013). Silica has been utilized in the hardware filed, as a substances for coordinates circuit chips, microchips, semiconductors, transistor, cell phone, and other electronic gadgets (Irimia-Vladu, 2014). Due to its highly porous architecture, silica can absorb active ingredients onto its surface and used as drug delivery agents in the medical and pharmaceuticals sectors (Pijarn *et al.*, 2010). In developing different bioactive devices for the tissue regenerating purposes, silica-based materials are also used (Baino *et al.*, 2016).

Most commonly available silica forms are quartz, tridymite and cristobalite. These are also known as “free silica” that differentiate them from the silica, mineral containing silica bound to one or more metallic cations (Dove and Rlmstidt, 2018). Generally, silica is found either in crystalline or amorphous form in the earth crust (Hernandez *et al.*, 2016). When the earth crust is disseminated and silica-containing rock and sand are utilized for processing, the employer faces several potential respiratory dangers. It usually happens during the rock drilling process when there isn't enough dust suppression, which can lead to higher silica dust concentrations in the air. Several pathological effects such as silicosis, pulmonary tuberculosis, multiple nodular lesion in the lung parenchyma, industrial bronchitis and extra pulmonary diseases are the consequences of this silica dust inhalation (Baur *et al.*, 2019). Although some protective measure has been taken to reduce the risk of this dust inhalation or other associated issues such as using proper respirator during the drilling or cutting of the rock, banding sandblasting techniques are used in some countries, adding water in the cutting surfaces during the drilling, mining and cutting (Ramírez-Gómez, 2016).

Although silica production from the biomass waste like rice husk is the cheapest source, where 50-90% silica can be found. A highly pure silica can be obtained from the biomass waste in a minimal costing (Patel *et al.*, 2017).

Silica and silica containing materials are frequently utilized in dentistry for different purposes. It is most commonly used as a filler in different filling materials such as glass-ionomer cements, compomers, composites, and adhesive systems (Ikemura *et al.*, 2003). Generally, the filler react with acid during the setting mechanism to increase the mechanical properties by improving the physical resistance, thermal expansion coefficient and radiopacity in acrylic filling materials (Marghalani, 2016). The filler materials also help to decrease the polymerization shrinkage which

subsequently improves the handling properties as well as aesthetics issues. In different ceramics or alloys surfaces, silicates are also utilized for the tribochemical silication. In composite resin, the organic matrix and the inorganic phase are connected through silane coupling agent where the silicates create the silane layer

silicates form a silane layer that act as coupling agent to connect the organic matrix and the inorganic phase (Jiapei *et al.*, 2018), which is known as silanization. This silanization process helps to increase the physical and mechanical properties of dental composite restoration. Silicates are also used in dental ceramics as a major ingredient such as veneers, inlays, onlays, full ceramics crowns, etc. In toothpaste, it is also used as a synthetic silica gel.

In mineral trioxide aggregate (MTA), silicon oxide also used as an oxide compound. MTA is commonly used to manage the root perforation, apexification, capping the vital pulp, etc (Schwarze, 2004).

### **1.3 Rice Husk**

Rice is the seed of plant species, which is known as *Oryza sativa* for Asian rice & *Oryza glaberrima* for African rice. Rice seed contains between 20-22% rice husk of its initial weight (Shen, 2017). Rice husk (RH) is an agricultural waste product, which can easily be available in all rice-producing countries, where nearly 90% of the entire rice culture is grown and consumed in Asia (Carmona *et al.*, 2013).

Rice husk is an irregular, boat-shaped, hard protecting coverings of rice seeds that contain cellulose (38%), lignin (22%) and ash (20%) as chief ingredients (Udenta, 2017). The major sources of rice husk ash (RHA) are silica (87-98 wt.% by in mass), with some small percentage of metallic elements (Arenas *et al.*, 2017).

Being biogenic in origin, the silica available in rice husk intrinsically amorphous with large surface area but in case of combustion more than 650 °C, it will convert the silica phase to crystalline. It is recognized as an attractive and cheap source of highly pure silicon (Kaviyarasu *et al.*, 2016). The unburnt rice husk contains up to 20% amorphous silica, depending upon the assortment, atmosphere and geographic area of the development of the rice (Beidaghy Dizaji *et al.*, 2019). Silica from rice husk has very minor impurities in comparison with other silica sources such as sand, bentonite, and diatomaceous earth. Highly pure silica extensively is used as a raw material in industry, ceramics, catalysis, chromatography, power, electronics, coatings, stabilizers, emulsifiers, and organic science. It is also used to synthesize different types of chemicals including sodium silicate, zeolite catalysts, aerogel, highly-pure silicon, silicon nitride, and silicon carbide (Patil *et al.*, 2014).

Due to the unique form and high surface area of silica, it is extensively used as a source for artificial adsorption substances carriers, clinical additives, etc. and shows favourable circumstances when accomplished at nanometre size (Jang *et al.*, 2009; Liu *et al.*, 2005; Shin *et al.*, 2008; Wongjunda and Saueprasearsit, 2010).

As a main ingredient of dental filler for different commercial cement, silica is the materials of choices for researchers (Curtis *et al.*, 2009). Silica is extracted from the rice husk and used as a filler to enhance the strength properties of restorative materials such as dental composite, glass ionomer cement (Van Tuan *et al.*, 2011).

In spite of the abundance of procedures to synthesize and decontaminate silica, most researchers to date have tended to the creation of silica straightforwardly from rice husk, not from its ash. Extracting silica directly from RH requires a high volume of RH which is not affordable and effective (Fernandes *et al.*, 2017). From an

economical perspective, RH should initially be utilized as a fuel for boilers, where it generates RHA as waste. Generally, there are two distinct ways to extract silica from this waste, which varies from one another in the purity of their results. RHA produces 83-95% pure silica when used directly without any acid treatment, while with acid treatment, it offers more than 95% pure silica. But, in the case of alkali or other chemical extraction methods, more than 99% of highly pure silica can be synthesized as well (Hossain *et al.*, 2018).

#### **1.4 Dental Luting Cement**

Dental Luting Cement is a type of dental cement that is used to secure the cast in fixed restoration, hold the orthodontic bands and appliances in place, and perform as a restorative material alone or in combination with other materials (Chandrasekhar and Sharma, 2016). Therefore, the most important point of the luting cement is to restrict the microscopic organisms and oral liquids from infiltration into the prepared surface and protect the thermal conduction just as maintenance of the restoration by filling the gap between the tooth surface and the restoration (Cionca *et al.*, 2017). There are different types of luting agents used in dentistry, such as zinc phosphate cements, zinc polycarboxylate cements, glass ionomer cements, hybrid ionomer cements, resin modified glass ionomer cements, polyacid modified resin cements, and resin cement (Almuhaiza, 2016). In 1878, Peirce (developed zinc phosphate cement which is known as the most established luting cement and has the longest reputation as a luting agent to secure cast restoration for over 130 years. That is why zinc phosphate cement has been considered as standard cement in comparison with the newer cement (Anusavice *et al.*, 2003). Then in early of 20th century (1903), the first direct colored restoration, silicates cement was developed. The durability of silicate cements depends

fundamentally on the consideration taken in dealing with the material as well as the oral cleanliness of the patient. In 1968, D.C. Smith built up the first cement framework with potential for adhesion to tooth structure utilizing separate powder and a fluid segment, known as polyacrylate cement (Petrich *et al.*, 2004). In 1969, Wilson and Kent manufactured glass ionomer cement to capitalize the benefit from the positive properties of both silicate and polycarboxylate cement by utilizing silicate glass powder and an aqueous solution of polyacrylic acid. Later in 1986, to enhance the resistance property of glass ionomer cement as well as reduce the water sensitivity, resin modified glass ionomer cement (RMGIC) was developed (van Dijken *et al.*, 2019). Different types of resin-based cement have now moved toward becoming accessible in view of the advancement of the direct filling resins with improved properties like acid-etch technique.

### **1.5 Nanohybrid Dental Composite:**

Dental amalgam has been considered safe and widely used for restorative materials for more than 150 years. However, the issue of mercury toxicity and unpleasant aesthetic of the amalgam restoration leads to the search for a better replacement. The relationship between amalgam mercury with oral lichen planus and contact dermatitis placed amalgam restoration in the unfavorable situation as a dental restorative material (Thornhill *et al.*, 2003). Although the amalgam has the aesthetic and retention issues, it is still considered as the material of choices for the restoration of posterior teeth (Lynch *et al.*, 2014). The establishment of composite restorative material was a significant achievement, particularly in the esthetics dentistry. The extensive use of composite restorative material in dentistry slowly removing the once-popular dental amalgam from the dentistry market.



The introduction of dental composite resins into the field of conservative dentistry started in the 1940s, overcoming the disadvantages of the acrylic resins and silicate cement. Silica cement was among the earliest aesthetic dental material ever produced (García *et al.*, 2006). In 1955, Buonocore used orthophosphoric acid to create a mechanical adhesion between his new developed acrylate resin and the tooth structure. However, acrylate resin presented with poor wear resistance, high polymerization shrinkage and high thermal expansion which limits their usage. In 1962, Dr L. Bowen designed a new dimethacrylate resin where the Bisphenol A Glycidyl methacrylate (Bis-GMA) and an organosilane coupling agent were used to make a bonding between filler particles and the resin matrix (Menon *et al.*, 2016), which was a significant progression in dental composite resin. Lately, a unique kind of dental composites has been created and named as nanohybrid, where the manufactures have changed the filler size by adding more nanoparticles with the microhybrid filler to get the benefits from both microhybrid and nano particles in a single material. Generally, nanohybrid composite exhibit higher wear resistance property, better optical properties and lower amount of polymerization shrinkage in contrast with other available composite restoration (Lyapina *et al.*, 2016).

## **1.6 Problem Statement and Justification of the Study**

At present, dental composites are not being made in Malaysia and it relies upon imported business items to satisfy the needs of its population making it very costly. The advancement and ideal commercialization of a local dental nanocomposite will profit the nearby population by chopping down the cost of these rebuilding efforts. That will give more employments if commercialization is done everywhere scale and

furthermore overhaul the situation of Malaysia as a maker and most likely an exporter and not only a client and ultimately it will increase the Malaysian economy.

In the Universiti Sains Malaysia (USM), our group of researchers has manufactured the experimental nanohybrid dental luting composite cement from the rice husk and evaluated the mechanical, physical and antibacterial properties. Other similar types of studies are also ongoing by other researchers in USM, to expand this project using different types and percentages of different fillers and resin matrix. Previously in 2015, Dr. Md Noushad extracted silica from the rice husk and used as a filler to fabricate the dental nanocomposites (Noushad *et al.*, 2016). They used two different fillers and resin ratios, which were 40/60 and 50/50 and evaluated the mechanical testing and surface roughness testing of the experimental dental composite. They showed better mechanical strength results which were all above the ISO standards but just lower than the commercial products. Moreover, number of studies have been conducted on silica for fabricating dental composites and now extend to luting cement. Based on this idea, we think that the dental composite resin research can be extended into fabrication of luting cement.

The usage of nano silica with nano fill composite decreases shrinkage and improves the nature of the composite. Nowadays, most of the nano silica molecule is readied utilizing a sol-gel technique which is costly and limits their application (Ciriminna *et al.*, 2013). Rice husk an agricultural waste, which is the result of rice processing contains natural materials and silica as the fundamental constituent. That silica from the rice husk will greatly reduce the costing to produce nanohybrid dental luting cement.

In dental practice, a significant concern is the failure and replacement of dental restorations. Generally, the main function of the dental luting cements is to establish or increase retention of the prosthetic appliance to abutments and to maintain its integrity. To prevail in both, a perfect material ought to satisfy explicit biological, physio-mechanical, and handling characteristics. As of now, accessible materials usually show great biologic execution, albeit some unfavorable impacts which have been recognized such as postoperative sensitivity, and sometimes, patients complain allergies to dental materials, although the rate is generally low (Syed *et al.*, 2015).

The commercialization of a privately made dental nanohybrid will extraordinarily diminish the expense of a dental luting cement. It will also make available to the right masses who can't deal with the high expense of the dental restorations. Thus, it can make a high impact in Malaysian economy being as a manufacturer and most likely exporter of dental nanohybrid luting cements as opposed to only a customer or exporter.

## **1.7 Objectives and Aims**

### **1.7.1 General Objectives**

To develop an experimental nanohybrid dental luting composite cement incorporated with silica from the rice husk together with commercially available zirconia and alumina cement and evaluate the physical and mechanical properties.

### **1.7.2 Specific Objectives**

1. To develop an experimental nanohybrid dental luting composite cement from rice husk incorporated with zirconia and alumina.
2. To evaluate the mechanical properties of the experimental NHDLCC, which are compressive strength and flexural strength testing.
3. To evaluate the mechanical properties of the experimental NHDLCC, which are compressive strength and flexural strength testing.
4. To evaluate the physical properties of the experimental NHDLCC, which were water sorption and water solubility and viscosity testing.

### **1.8 Research Questions**

1. Are the reinforcement of zirconia and alumina increase the compressive strength and flexural strength of experimental nanohybrid dental luting composite cement?
2. Are the reinforcement of zirconia and alumina reduce the water sorption or solubility and increase the viscosity of experimental nanohybrid dental luting composite cement?

### **1.9 Research Hypothesis**

1. Reinforcement of zirconia and alumina increases the compressive strength and flexural strength of experimental nanohybrid dental luting composite cement.

2. Reinforcement of zirconia and alumina reduces the water sorption or solubility and increases the viscosity of experimental nanohybrid dental luting composite cement?

## CHAPTER 2

### LITERATURE REVIEW

#### 2.1 Dental Cement

Dental cement is characterized as an object that solidifies to act as a base, liner, filling material, or adhesive to attach devices & prostheses to tooth structure or one another. The principal objectives of dental cement are to fill the space between the restoration and prepared tooth structure to increase resistance and prevent its dislodgement during mastication (Prates Soares *et al.*, 2020). Lately, numerous luting agents and dental cement have been presented with the case of clinically preferable execution over existing materials because of improved attributes. So, choosing a convenient dental cement is a crucial judgment in a series of steps that needs accurate execution and will establish the long-term success of fixed restorations (Pameijer, 2012).

The development of dental cement has started from a humble beginning, where currently dentists are dealing with plenty of choices. Figure 2.1 introduces an overview of dental cement evolution from the starting days up to the 20<sup>th</sup> century (Yu *et al.*, 2014).

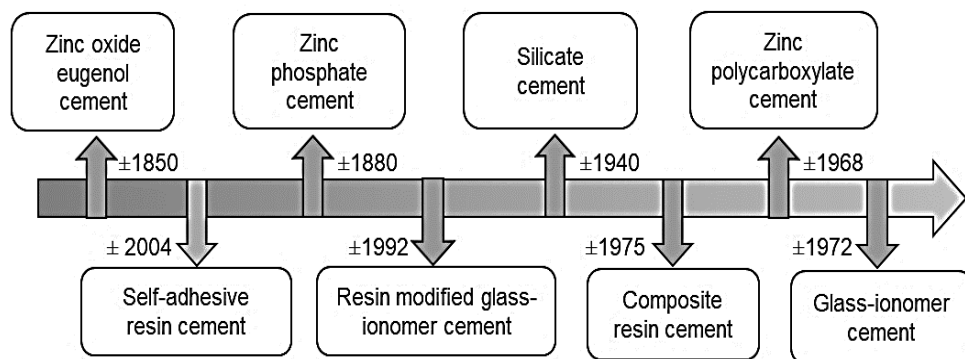


Figure 2.1: Chronological progression of dental cement

Dental cement should accomplish definitive biological, physio-mechanical, and handling characteristics that provide adequate retention and mechanically lock the restoration to the prepared tooth or implant abutments to maintain its integrity. The ideal factors for successful cementation are as follows: (Attar *et al.*, 2003; de la Macorra and Pradíes, 2002; Pegoraro *et al.*, 2007).

- Cheaper with prolonged shelf life and easy to manipulate.
- Good biocompatibility and non-cariogenic and ability to prevent secondary caries formation.
- Adequate physical properties: appropriate film thickness, low solubility, enough working time, low viscosity, radiopaque.
- Good mechanical properties: Adequate shear/ tensile/ compressive/ bonding strength.
- Chemically inert and insoluble in saliva and liquid.
- Good thermal insulator and aesthetically pleasant.

## **2.2 Nanohybride Dental Cement**

There are different types of dental composites used for restoration where the hybrid and nanofiller types are the most conventional types. In case of hybrid composite, it exhibits better mechanical strength due to the presence of different size of filler particles (average 15–20 $\mu$ m and 0.01–0.05 $\mu$ m) with an intermediate esthetics property. To overcome these aesthetic issues, microfill composites were introduced in the market with the formulation of colloidal silica and their average particle size was 0.02 $\mu$ m (at a range of 0.01–0.05 $\mu$ m). Unfortunately, their mechanical strength is not enough especially in the high occlusal area (Van Noort and Barbour, 2014). In recent

years, a special type of dental composites has been developed where the fabricates have changed the filler definition of micro hybrids by adding more nanoparticles to obtain the better mechanical strength of hybrids and higher aesthetics properties of microfill and have named as nanohybrid (Moszner and Klapdohr, 2004; Moszner and Salz, 2001). Decreasing polymerization shrinkage (Chen *et al.*, 2006), higher wear resistance (Terry, 2004; Yap *et al.*, 2004) and increased optical properties are the other positive characteristics found for the nanohybrid composite. However, when contrasted and customary with conventional micro hybrids, nanofiller composite resins will keep up better surface smoothness and polish and are in this manner extraordinary for the single-item client.

### **2.3 Dental Composite resin material**

Dental composite resin is the universally used tooth-colored restorative materials, that is used to restore decayed or destructed teeth. The introduction and evaluation of dental composites is one of the best modern innovations in operative dentistry and become more demanding and well accepted due to the cosmetic improvement of the smile by correcting the shade of the tooth and reshaping the deformed tooth (Stein *et al.*, 2005). Due to its micromechanical properties, dental composite is suitable for patients according to their cavity, that provides good durability and fractures resistance than most of the available restorative materials like amalgam restoration. The entry of dental composites has conquered the usages of dental amalgam. Apart from good aesthetics and mercury lethality fear, one of the major advantages for dental composites over dental amalgam is that it requires a minimal cavity preparation (Pantaleón *et al.*, 2018). Nowadays, almost 50% of the



posterior direct restoration depends on composite restoration and for anterior teeth direct restoration its near about 95% (Sabandal and Schäfer, 2016).

In 1962s, dental composite made access as direct tooth-colored restorative materials when Dr. Ray Bowen developed a monomer named Bisphenol A Glycidyl methacrylate (Bis-GMA) and successful synthesis of composite by adding inorganic fillers (Bayne *et al.*, 2019). These early composite were chemically cured, required the two paste system (base and catalyst paste to be mixed together), causing the drawbacks with the proportion, mixing procedure and color adherence (Kinomoto *et al.*, 1999). To overcome this situation, in the 1970s the electromagnetic radiation was being used to polymerize the composite restoration. Initially, an ultraviolet light source (365nm) was utilized to give the required light energy, which was later replaced by visible light (427-491nm) because of its shallow polymerization and iatrogenic reactions (García *et al.*, 2006). Along with sufficient physio-mechanical and esthetic properties, the dental composite has few disadvantages like polymerization shrinkage, relatively high coefficient of thermal expansion, low wear resistance, and also require longer working time and so forth (Sevkusic *et al.*, 2014). Furthermore, the durability of composite resin restoration is approximately 7 years, which is higher than most other remaining restorative materials except for amalgam, which can last for over 10 years (Demarco *et al.*, 2012). As the composite materials possess satisfactory properties than most of the other conventional materials therefore, the research must be continued further to find out the shortcoming of composite materials and correct them.

The basic structural component of modern dental composites consists of an organic phase or resin matrix, an inorganic filler, a coupling agent, and an initiator & accelerator. Different composite systems employ different formulations of these

ingredients to achieve desired characteristics according to the caries site, caries size, mastication load, and their location.

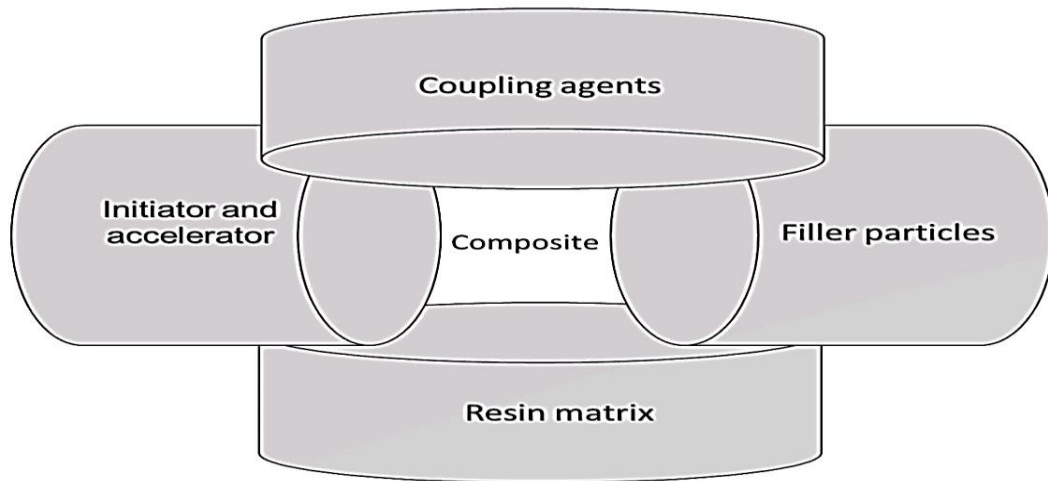


Figure 2.2: Composition of dental composite

## 2.4 Resin Matrix

In the 1960s, Dr. Bowen developed the first epoxy resin and methacrylate derivatives that helped bind the filler particles together (Ruyter and Øysæd, 1987). Generally, the polymerization reaction takes place between the carbon-carbon double bonds of two methacrylate groups via a free radical (Yu *et al.*, 2015). The organic resin matrix is formed on a combination of dimethacrylates monomer like Bis-GMA, UDMA, Bis-EMA, TEGMA, TEGDMA, and DUDMA (Rinastiti, 2016). Some author reported that the types of monomer used in composite is directly related in polymerization shrinkage, viscosity, reactivity, mechanical properties and water intake of the composites (Visvanathan *et al.*, 2007).

Bisphenol A diglycidyl methacrylate (Bis-GMA) is widely used monomer for the fabrication of dental composites (Srivastava *et al.*, 2017). Bis-GMA is a high molecular weight monomer with two aromatic rings in pendant hydroxyl groups

(—OH—), which are responsible for high viscosity and some inescapable water sorption (Botelho, 2003; Palmer *et al.*, 2004; Takahashi *et al.*, 2006). It also specifies the addition of fillers and thus a low degree of conversion (Amirouche-Korichi *et al.*, 2009). Due to its minor volatility and diffusivity into tissues, the choice of using Bis-GMA than other monomer are less shrinkage, high strength, and hardness, and lessen toxicity.

Triethylene glycol dimethacrylate (TEGDMA) is an aliphatic and hydrophilic monomer widely used within most current dental composites (Frencken *et al.*, 2007). Due to their low viscosity and copolymerization shrinkage, typically in a 1:1 ratio of TEGDMA is used as a diluent monomer for both UDMA based composites and Bis-GMA. Although the incorporation of TEGDMA influences the degree of conversion and water sorption of composite resin (Kielbassa *et al.*, 2003).

Different studies are still ongoing to find out a proper fabrication of monomer to develop the properties of the dental composites. Atai *et al.*, 2004, studied the impact of the replacement of a new dimethylacrylate monomer based on 3, 3',4,4'-benzophenone tetracarboxylic dianhydride (BTDMA) over Bis-GMA assumed to exhibit similar or better mechanical properties, because of its rigid and bulky structure (Atai *et al.*, 2004).

Lie *et al.*, 2004, reported the prepared visible light-activated composites which were processed on 1,3-bis[(p-acroxymetyl) phenethyl] tetramethyl disiloxane (BAPD). It is a high molecular weight siloxane monomer with a low viscosity. In comparison with Bis-GMA, during the development of the composite resin with BAPD, it did not require the addition of diluent monomer like TEGDMA. Besides, the degree of conversion of BAPD was essentially higher than the conventional monomer



### 2.5.1 Filler Size in Dental Composite Properties

The incorporation of different filler sizes has a significant impact on the dental composite properties. Different studies have shown that, the addition of smaller particles in between the larger one reduces the interparticle spacing and the quantity of the resin matrix and ultimately enhance the composite properties (Marghalani, 2010).

This different filler sizes enhance the filler fraction by allowing the smaller particles to compensate in between the larger particles and thus increase the packing density without compromising the viscosity and the handling properties (Pick *et al.*, 2011). Furthermore, the addition of smaller particles can also improve the composite strength properties by enhancing the particle surface area that results in a high surface energy at the resin-filler interface (Kundie *et al.*, 2018b). It is also reported that the larger filler particles may also harden the composite property (Zhou *et al.*, 2007).

In dental composite resin, different filler sizes have been utilized that ranges from as large as 100 $\mu$ m to very small size between 0.1 to 100nm (Beun *et al.*, 2007). Based on the fillers' particle size, dental composite is broadly classified into four types, namely macro filled composite, micro filled composite, hybrid and nanocomposite.

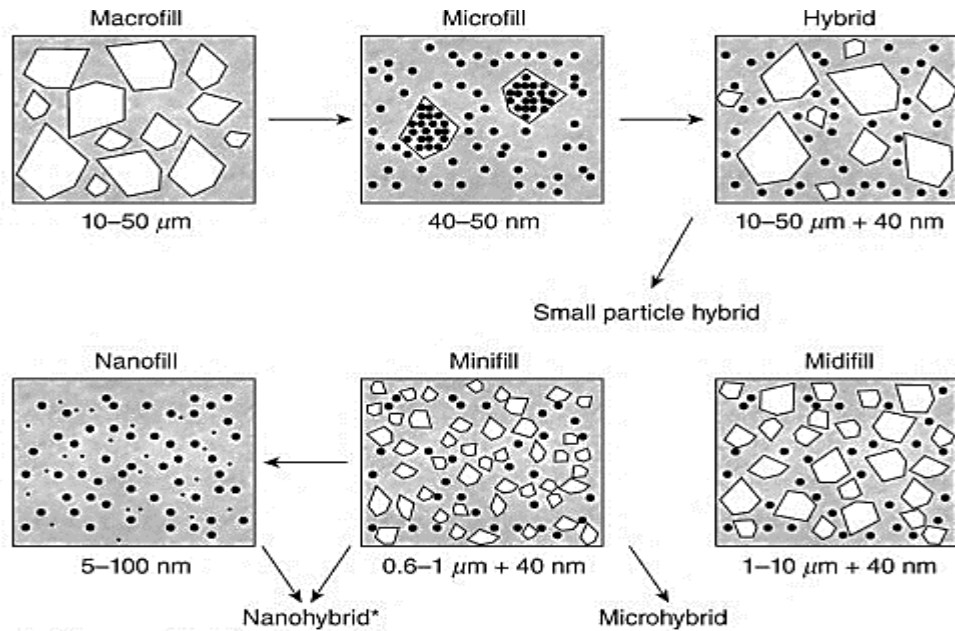


Figure 2.4: Classification of composite based on filler size

### 2.5.1(a) Macro Filled Composite

The conventional or macro filled composite was first established in the late 1950s (Anusavice *et al.*, 2003). It consists of ground, crushed or milled, splinter shaped glass and quartz filler particles ranging from 0.1-100 $\mu\text{m}$  (Ferracane and Greener, 1984). It is the materials of choice in stress bearing restorations due to its good strength properties (Sideridou *et al.*, 2009), but poor wear resistance, less polishable and discoloration limit their uses (Heuer *et al.*, 1982; Phillips *et al.*, 1972).

### 2.5.1(b) Micro Filled Composite

It was introduced in the late 1970s composed of amorphous silica particles in an organic matrix. The average filler size is about 0.02  $\mu\text{m}$ , within a range of 0.01-0.05 $\mu\text{m}$  to get the 35 wt.% of filler content (Karabela and Sideridou, 2011). In combination with a better aesthetic and smooth finish restoration, it exhibits the moderate strength property (Barakah and Taher, 2014).

### **2.5.1(c) Hybrid Composite**

Most common types of composite that are available in the commercial products consist of more than one filler size. The combination of micro filled, and macro filled composites with the average filler particles size of 15-20  $\mu\text{m}$  and 0.01-0.05  $\mu\text{m}$ . The micro filler particles permit the initial polishability and increase the surface morphology but since macro filler particles are still dislodged, roughness of the surface will occur over time. This wide filler particles size ranges may lead to high filler loading with better strength properties, enhance stiffness and wear resistance also (Barakah and Taher, 2014; Venhoven *et al.*, 1996).

### **2.5.1(d) Nano Filled Composite**

The nanotechnology prepared with nanomer and nanocluster filler particles is the recent advancement in the field of dentistry claiming superior clinical properties in both aesthetic and load-bearing situations. Nanometer-sized filler particles (approximately 0.6-1.4  $\mu\text{m}$ ) and larger groups of fused nanoclusters (approximately 25 or 70 nm size) are dispersed in a resin matrix to produce a nanocomposite. This conjugation of nanomer and nanocluster fabrications significantly decrease the interstitial spacing between the filler particles, resulting in increased filler loading consequently reduce the polymerization shrinkage and improve mechanical properties (Moszner and Klapdohr, 2004; Moszner and Salz, 2001).

## **2.5.2 Filler Shape in Composite Properties**

Nowadays, majority of the commercial fillers are either irregular or spherical shaped (Randolph *et al.*, 2016). Studies have found that, in dental composite, spherical

shaped filler influences the microfracture properties and also increases the fracture toughness and bending strength with a much higher rate for elastic modulus (Beun *et al.*, 2007). Increasing filler load is one of the most important advantages of using spherical shaped filler (Sabbagh *et al.*, 2004). According to the manufacturers' data, the spherical shape had narrow particle size distribution than the irregular shaped filler (Satterthwaite *et al.*, 2012).

### **2.5.3 Filler load in composite properties**

Filler loading plays an important role in the physio-mechanical properties of dental composite resin, more specifically by enhancing modulus, radio-opacity or by changing thermal expansion characteristics (Cramer *et al.*, 2011). Several studies reveal that by increasing filler load in dental composite, it can reduce the polymerization shrinkage and subsequently enhance the hardness, compressive strength, tensile strength and also fracture resistance properties (Moszner and Klapdohr, 2004). Increased filler load can also reduce the water sorption and also exhibit higher abrasion and wear resistance (Kim *et al.*, 2002).

### **2.5.4 Types of filler used in composite (silica, alumina & zirconia)**

In general, dental composites are composed of filler particles, resin as a matrix and silane as coupling agent. During the polymerization process, the filler particles are usually used to reduce the resin matrix shrinkage. Studies reveal that, composite with 100% resin matrix (methyl methacrylate) shows 20-25% volumetric shrinkage whereas the one with 60-70% filler can reduce the shrinkage up to 2%. In dental composite, usually 35-60 vol.% filler is carried out through the total concentration (Klapdohr and Moszner, 2005; Rüttermann *et al.*, 2007). Commonly used filler in