The Study of Water Sorption and Solubility of Differently Formulated Resin Composite Restorative Materials
A Comparative Study in vitro

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Abbreviation	Full form
HEMA	2-hydroxylethyl methacrylate
Bis-GMA	Bisphenol A glycidyl methacrylate
BPA	Bisphenol-A
CQ	Camphorquinone
C-factor	Configuration factor
DC	Degree of conversion
EBPDMA	Ethoxylated Bis-GMA
Bis-EMA	Ethoxylated bisphenol A dimethacrylate
EGDMA	Ethylene glycol dimetahcrylate
GPDM	Glycerol phosphate dimethacrylate
ISO	International organization for standardization
LED	Light emitting diode
MMA	Methyl methacrylate
SL	Solubility
TCD-urethane	Tricyclodecane-urethane dimethacrylate
TEGDMA	Tri-ethylene glycol dimethacrylate
UDMA	Urethane dimethacrylate
WS	Water sorption

Dental resin composite restorations are widely used to cope with the increased esthetic demands among adults and even children. Its esthetic properties and good clinical use have made it the preferred material for most posterior restorations[1]. One of the main disadvantages of resin composite is the polymerization shrinkage which resulted into microleakage, debonding, secondary caries and postoperative sensitivity. This polymerization shrinkage may cause interfacial debonding between composite resin and walls of the cavity with a consequent gap formation and following secondary caries[2]. To reduce polymerization shrinkage stresses, clinicians have evolved more materials and techniques to solve this problem[3]. One of these techniques is incremental packing of composite. Recently, bulk fill composites had been evolved to decrease voids accompanied by incremental technique and to facilitate use of composite to the clinicians as it is faster and more simple[4].

Flowable composite is used as pit and fissure sealant, small direct and indirect restoration, block out small undercuts in indirect cavity preparations and luting porcelain and composite resin veneers[5].

Water sorption and solubility of resin-based materials are important drawbacks of composite resin, since they influence the mechanical properties of the dental materials and longevity of composite restorations. In a wet oral environment, composites absorb water and release unreacted monomers and inorganic ions. The water uptake in dental composites had harmful effects on their mechanical/physical properties because of the hydrolytic breakdown of the bond between the silane-filler particles, filler-matrix debonding or hydrolytic degradation of the filler. In the same time, residual monomers and other components eluted from composites into the oral environment, weaken their mechanical properties[6].

On the other hand, the absorbed water may cause a hygroscopic expansion of the composite that may compensate the polymerization shrinkage and improve the restoration seal. Moreover, water sorption and solubility affect the strength, abrasion resistance and volume of resin composites.[7].

So, in order to maintain the composite restoration material in the oral cavity for a long duration, it is mandatory to study their water sorption and solubility properties to be able to predict their behavior in the oral cavity.

Dental composite

Dental resin composites were developed in response to people's demands for tooth-colored restorations[8]. Resin composite restorative materials have a number of advantages over dental amalgam including improved aesthetics, conservative cavity preparation and are adhesively bonded to the tooth with a compatible bonding system[9]. Also their favorable physical and mechanical properties, including high resistance to compression and wear, relatively low costs and simple application, recently emerged as a substitute for amalgams[10]. In the late 1950s and early 1960s,composite material was introduced to the market[11].

The first material developed for use as a direct esthetic restorative was silicate cement. It was developed in the late 1800s. Silicate cements had the disadvantage of deteriorating rapidly, because it was highly soluble in oral fluids[12]. The 1950s was the introduction of unfilled acrylic resin based on Methyl Methacrylate[13]. Robert Chang in 1969 and Henry Lee in 1970 were the first to use of composite in the paste/liquid form[13]. The late 1970s saw the development of a photo-polymerized resin composite system[14]. Such a polymerization method provided dentists with the ability to polymerize a composite at a fast rate, upon placing and contouring[13].

A composite is material that is composed of hard, pebble-like filler particles, surrounded by a hard matrix of a second material which binds the filler particles together[15]. Dental resin composites consist of a polymeric matrix based on di-methacrylate monomers, inorganic fillers for polymer reinforcement, and a coupling agent usually an organo-silane to bond the two phases[16]. An initiatoraccelerator system is also added to allow for the polymerization reaction to occur**[17]**.

Fillers incorporation greatly influence and improve material properties. Fillers increase hardness, strength, radiopacity and decrease in polymerization shrinkage, thermal expansion and contraction, water sorption, softening and staining**[18]**. There is a wide range of fillers available. Glass particles are the most common, due to their improved optical properties. Previously, quartz was favored and very commonly used. This was because of its better mechanical properties, availability and stability in contrast to other fillers. However, due to the hardness of quartz particles, enamel attrition was increased. This feature has led to the decline of quartz and increase in popularity of glass fillers. Other commonly used fillers include borosilicate glass, lithium,barium aluminium silicate, and strontium or zinc glass**[13]**.

Within each type of composite, the materials were further distinguished by the characteristics of their reinforcing fillers, and in particular their size**[19]**. Resin composite was classified according to filler size into: Macrofilled composite contain particles ranging from 1 to 50 microns (µM) in diameter. This type of composite was very strong, but difficult to polish and impossible to retain surface smoothness. It was rough which resulted in increased accumulation of plaque and stain. The poor polishability of macro-filled composites led to the development of micro-filled composite [19], based on heavily filled polymer resin blocks had milled to a particle size of 25µm.

Microfilled composite materials contain colloidal silica particles approximately 0.01 to 0.1µm in diameter, with a typical mean diameter of 0.04µm. They could be polished easily, but generally weak due to their relatively low filler content[19]. They had greater water sorption, a higher coefficient of thermal expansion and decreased elastic modulus. Moreover, they had lower fracture resistance, stiffness, and fatigue strength compared with heavily filled composites[20]. Therefore, the particle size of the conventional composites was reduced through further grinding to produce what was ultimately called hybrid composites.

These were further distinguished as “midi-fills,” with average particle sizes slightly greater than 1µm but also containing a portion of the 40 nm-sized fumed silica “micro-fillers”. Hybrid composites had a problem of not retaining a high polish for long, because of large particles. However, hybrid composites were easy to work with and were resistant to wear. They include smaller and submicron-sized particles which are more difficult to dislodge than large particles. Because of high particle density, hybrids were the first composites promoted for posterior use, and they remain one of the most wear-resistant posterior composites on the market[15].

Further refinements in the particle size through enhanced milling and grinding techniques resulted in composites with particles that were sub-micron, typically averaging about 0.4–1.0µm, which initially were called “mini-fills” and ultimately came to be referred to as “micro-hybrids[19].

The most recent development is the “nano-fill” composites, containing only nanoscale particles. They were formed by the inclusion of nanoparticles, 20 or 75 nm in size, and nanoaggregates of approximately 0.6-1.4 µm. The smallest nano particles are formed by colloidal silica which is produced by burning silica compounds such as SiCl4 within an oxygen atmosphere forming spherical, macromolecular structures[15]. These materials are generally considered to be universal composites as they can be used for most anterior and posterior applications based on their combination of strength and easy polishing and aesthetic properties[6].

Nanofilled composite particles give the restoration a better finish, which was observed in its surface texture, and the likelihood of the material's biodegrading over time was reduced. This technology had also achieved improved mechanical properties for the resin to be indicated for use in the anterior and posterior restorations. Also, lower size of the particles leads to less curing shrinkage, created less cusp wall deflection and reduced the presence of micro cracks in the enamel edges, which were responsible for marginal leakage, color changes, bacterial penetration and possible post-operative sensitivity[14].

Filler loading of each type of composite was also different. As Macrofilled composite was 70-80% by weight. Microfilled composite was 35% by weight. Hybrid composite was ranging from 8-75% by weight due to different particle size and Nanofilled composite 75% by weight.

The organic matrix of conventional resin composites is generally based on methacrylate chemistry, especially crosslinking dimethacrylates[21]. Composite formulations have continued to evolve, since bisphenol A glycidyl methacrylate (Bis-GMA) was first introduced to dentistry by Bowen in 1962, it has the viscosity of honey, and have been modified in various ways based on different properties, like viscosity or polarity[22]. Subsequent experiments incorporated triethilene glycol dimethacrylate (TEGDMA) as a diluent to reduce viscosity. This monomer combination had become one of the most widely used matrix monomer combinations for dental work. Both of these monomers contain two reactive double bonds, and when polymerized, form covalent bonds between the polymer chains known as a cross-link.

Cross-linking improved mechanical and physical properties[23]. As the resin is highly viscous, in addition to facilitating the manufacturing process and clinical handling it is diluted with other low-viscosity monomers or low molecular weight monomers which are considered viscosity controllers, such as bisphenol A dimethacrylate (Bis-DMA), ethylene glycol dimethacrylate (EGDMA), triethylene glycol dimethacrylate (TEGDMA), methyl methacrylate (MMA) or urethane dimethacrylate (UDMA)[14]. However, studies on the safety of Bis-GMA monomer had considered the possible release of Bisphenol-A (BPA) from the matrix and its adverse effects. Also, TEGDMA had a greater cytotoxic potential[24]. Urethane dimethacrylate (UDMA) had been proposed as a substitute for Bis-GMA, it has a similar molecular weight but is less viscous. It is used either alone or in combination with other monomers. In addition, it had confirmed superior mechanical properties of UDMA based composites to composites based on Bis-GMA[13]. Ethoxylated bisphenol-A-dimethacrylate (Bis-EMA) is a viscous monomer that is structurally analogous to Bis-GMA but without the two pendant hydroxyl groups. It can decrease water sorption of the resin, which is partially or completely substitute Bis-GMA in the recent formulation of dental composites.