



FILLERS IN DENTAL MATERIALS: A REVIEW

Endodontics

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ABSTRACT

The science of dental materials covers a broad range of terminology, composition, microstructure and properties used to describe or predict the performance of preventive and restorative materials. Chemical properties generally comprise the behaviour of material in a chemical action with or without any other external influences. Mechanical properties are related to behaviour of materials in response to externally applied forces or pressure. Fillers are added to a formulation to lower the compound cost and to improve these properties. Data from Pubmed and Medline databases have been extracted. Hand search and grey literature were also assessed. This review gives a comprehensive view of the various fillers used in dental materials.

KEYWORDS

Fillers, Silica

1. INTRODUCTION

Fillers are particles added to materials (plastics, composite material, concrete etc) to lower the consumption of more expensive binder material or to better some properties of the mixed material. Such materials can be in the form of solid, liquid or gas.^[1,2] Most of the materials used in dentistry contain fillers as a part of their composition. Fillers improve the chemical and mechanical properties by increasing physical resistance, thermal expansion coefficient and radiopacity. Combination of individually dispersed nano sized filler particles and nanoclusters enhance the aesthetics.^[3] Bioactive glass fillers incorporated reduce bacterial penetration into marginal gaps for composite restorations.^[4]

With the advent of modern technology, various attempts are made to modify these fillers to make them more beneficial for a better material. Strong scientific and collaborative foundations presently exist for further development and improvement of the filler system, which will certainly continue to hold a pivotal role in the dentistry.

2. CLASSIFICATION BASED ON FILLER PARTICLE SIZE^[1]

2.1 Skinner classification:

- Traditional or conventional composites: 8-12 μm
- Small particle filled composites: 1-5 μm
- Microfilled composites: 0.04–0.9 μm
- Hybrid composites: 0.6-1 μm

2.2 Bane and Heyman classification

- Megafillers or very large particle size : 0.5-2mm
- Macrofillers: 10 – 100 μm
- Midifill: 1-10 μm
- Minifill : 0.1 – 1 μm
- Microfill : 0.1 – 0.01 μm
- Nanofill : 0.005 – 0.01 μm
- Hybrid : mixed particle sizes

2.3 Philips and Lutz classification:

- Traditional composite resins: 5.30 μm earlier, 1.5 μm current
- Hybrid composite resins: 1.5 μm earlier, 0.05-0.1 μm current
- Homogeneous microfilled composites: 0.05-0.1 μm
- Heterogeneous microfilled composites: 0.05-0.1 to 1.25 μm

2.4 Based on performance:^[5]

- Functional Fillers
- Extender Fillers

2.5 Based on type:^[5]

- Particulate fillers
- Rubbery fillers
- Fibrous fillers

2.6 Classification based on chemical family^[6]

| Filler Type | Examples | Chemical Composition |
|---------------------------|------------------------------------|---------------------------------------|
| Oxides | Silica, alumina, titania, zirconia | MxOy |
| Alkaline silicate | Barium glass, Strontium glass | MxOySiO_2 |
| Biomimetic filler | Hydroxyapatite | $\text{Ca}_5(\text{PO}_4)_3\text{OH}$ |
| Organic-inorganic hybrids | ORMOCERS | SiO_2 -polymer |

3. FUNCTIONS OF FILLERS^[1]

Reinforcement: Increased filler loading generally increases physical and mechanical properties that determine clinical performance and durability such as compressive strength, tensile strength, modulus of elasticity and toughness.

Reduction of polymerization shrinkage: Increased filler loading reduces curing shrinkage in proportion to filler fraction.

Reduction in thermal expansion and contraction: Increased filler loading decreases the overall coefficient of thermal expansion of the composite that decreases interfacial stress while an individual consumes hot and cold foods and beverages.

Control of workability/viscosity: Filler loading and the range of particle sizes and shapes all affect markedly the consistency which in turn strongly affects clinical manipulation and handling properties.

Decreased water sorption: Increased filler loading decreases water sorption. Absorbed water softens the resin and makes it more prone to abrasive wear and staining.

Imparting radiopacity: Resins are inherently radiolucent. However secondary caries, poor proximal contacts and other problems cannot be detected unless adequate radiographic contrast can be achieved. Radiopacity is most often imparted by adding certain glass filler particles containing heavy metal atoms and compounds which strongly absorb x-rays. The most commonly used glass filler is barium glass.

Improve esthetics with finishing and polishing: The traditional inorganic filler particles had average diameters of about 8 to 40 μm . Currently, small particles range from 0.005 μm to 2 μm . Particles larger than the wavelength of visible light cause light scattering. Scattering increases opacity and produces a visibly rough texture when the particles are exposed at the surface. Another advantage of using small particles is that they improve esthetics with finishing and polishing.

4. DIFFERENT FILLERS USED IN DENTISTRY

4.1 Silica

Silica is an oxide of silicon with the chemical formula SiO_2 . This has a number of distinct crystalline forms in addition to amorphous forms

that includes quartz, trydamite and crystoballite. The high temperature minerals, cristobalite and tridymite, have both lower densities and indices of refraction than quartz. Since the composition is identical, the reason for the discrepancies must be in the increased spacing in the high temperature minerals.^[7,8]

4.2 Oxides and hydroxides

Various oxides and hydroxide used as fillers in dentistry, add many advantageous properties of the materials. The most commonly used fillers are aluminium hydroxide, magnesium hydroxide, zirconium dioxide, bismuth oxide and zinc oxide.^[9]

4.3 Polymers

A polymer is a large molecule composed of many repeated subunits. The consequently large molecular mass relative to small molecule compounds produces unique physical properties including toughness, viscoelasticity, and a tendency to form glasses and semi crystalline structures rather than crystals.^[10]

Polymers are of two types:^[11]

- Natural polymeric materials such as shellac, amber, wool, silk and natural rubber have been used for centuries.
- Synthetic polymers such as synthetic rubber, phenol formaldehyde resin, neoprene, nylon, polyvinylchloride, polystyrene, polyethylene, polypropylene, polyacrylonitrile, silicone.

4.4 Glass or ceramic containing heavy metals

Most commonly included are Aluminosilicates and Borosilicates.^[12]

5. VARIOUS RESTORATIVE CEMENTS

In the last 20 years there has been an explosion of different types of dental cements that have become available to practicing dentist, many of them tailored for specialized types of restoration. Different types of direct restorative materials are used in daily dental practice.

5.1 Dental amalgam

The American Dental Association (ADA) defines dental amalgam as an alloy composed of mercury, silver, tin, and copper along with other metallic elements added to improve physical and mechanical properties.^[13] It has served as an excellent and versatile restorative material for many years, despite periods of controversy.

The physical properties of the hardened amalgam depend on the relative percentages of each of the microstructural phases. Its main composition consists of silver-mercury matrix containing filler particles of silver-tin.^[14] These particles retained in the final structure act as a filler increasing its strength, corrodes the least and forms 30% of the volume. In high copper Amalgam, admixed powder is stronger than amalgam because of the silver copper particles which act as fillers in amalgam matrix, strengthening the amalgam.^[15,16]

5.2 Glass ionomer cements

Glass ionomer cements (GIC) is a hybrid of dental silicate and zinc polycarboxylate. The composition of glass ionomer cements is complex and varied. The basic component is a calcium aluminosilicate glass containing fluoride.^[1]

The set cement is a highly complex composite including gels of calcium and aluminium polyacrylates that contains fluoride. The unreacted portion of the glass powders act as filler for the cement. The glass powder is partly etched by the polyacid and the outer surface is degraded to siliceous hydrogel that contains fluorite crystallites. The set cement contains hydrogel matrix, a silica gel matrix and glass particles containing polysalt bridges between metallic ions and carboxylate groups. The fluxing action of calcium fluoride is generally supplemented by the addition of cryolite. This flux reduces the temperature at which the glass will fuse and increase the translucency of the set cement. Aluminium phosphate is also often included in the glass fusion mixtures. It improves translucency and apparently adds body to the cement paste.^[1,17] Metals, fibers and other nonreactive fillers have been evaluated in an attempt to improve the mechanical properties of GICs without compromising the handling or biological characteristics. Few of them include:

a) Amalgam alloy powder

The first attempt to increase the strength of conventional glass ionomer cements by addition of reinforcing fillers were reported by Simmons in 1983 when he added amalgam alloy powder to GIC powder

composition. Subsequently, McLean and Gasser fused and sintered amalgam powders to basic glass particles. The resulting cement particles exhibited strong bonding between the metallic and glass particles.^[17]

b) Reactive glass fibers

Lohbauer et al^[18] reported that a reactive glass fiber with 20 vol% of fiber loading had the ability to increase the fracture toughness of glass-ionomer cements. Fibers and fillers with different aspect ratios are thought to improve the mechanical properties of composite materials. Taubock et al^[19] added bioactive glass particles into the composition of GIC powder. These particles likely acted as fillers that had not adhered to the matrix of GIC leading to decreased compressive strength and modulus of elasticity. As a result its clinical usage ought to be restricted to applications where their bioactivity can be beneficial, such as root surface fillings and liners.

c) Incorporation of hydroxyapatite and zirconium oxides

In a recent study, Moshaverinia et al^[20] synthesized Nano hydroxyapatite and fluoroapatite using an ethanol based sol gel technique and incorporated the synthesized nanoparticles into commercial glass ionomer powder. Physical properties of the modified glass ionomer cements were evaluated. Results of their studies showed that after 24 h and one week of setting, the modified cement exhibited higher compressive strength, diametral tensile strength and biaxial flexural strength as compared to the control group.

d) GICs containing YbF3 and BaSO4 as filler particles

Prentice et al^[21] added nanoparticles of YbF3 and BaSO4 to conventional GIC. The BaSO4 increased the radiopacity of the cement and the YbF3 was a fluoride source that can modify both setting and working times.

e) Glass ionomers containing spherical silica filler (SSF)

Hatanaka et al^[22] incorporated silica fillers into GIC compositions and demonstrated that the addition of SSF, increased the compressive strength value by 1.1 times, while the increase of modulus of elasticity was 1.10 to 1.35 times.

f) GIOMER-The Intelligent Particle^[23]

Giomer is a fluoride releasing, resin based dental adhesive material that comprises PRG (Pre Reactive Glass) fillers that are fabricated by the acid-base reaction between fluoroaluminosilicate glass and polyalkenoic acid in the presence of water to form a wet siliceous hydrogel.

Types of PRG fillers

The PRG fillers depending on the degree of reaction with the acid are divided into two types:

S-PRG: The reaction is detected in surface-loans and are called surface reaction (surface reaction type, S-PRG fillers)

F-PRG: The reactions proceeded throughout and called are complete reactions (full reaction type, F-PRG fillers). The use of both types of PRG fillers promote rapid fluoride release through a ligand exchange within the prereacted hydrogel. The further advantage of S-PRG is that it releases five ions other than fluoride which have beneficial properties.

Modified S-PRG: Recently, improvement on the PRG technology, consists of a three layered structure with an original glass core of multifunctional fluoroboroaluminosilicate glass and two-surface layers that form a pre-reacted glass-ionomer phase on the surface of a glass core and a reinforced modified layer that covers the surface of pre-reacted glass-ionomer phase.

5.3 Composites

In materials science, a solid formed from two or more distinct phases (e.g., filler particles dispersed in a polymer matrix) that have been combined to produce properties superior to or intermediate to those of the individual constituents; also a term used in dentistry to describe a dental composite or resin-based composite.^[1]

The filler has several roles, including enhancing modulus, radiopacity, altering thermal expansion behaviour and reducing polymerization shrinkage by reducing the resin fraction. Finally, the filler-resin interface serves as a bridge by coupling polymerizable moieties to the particle surface. There have been remarkable developments in filler,

bonding, and curing technologies in esthetic restorative materials over the past 55 years.

Today, further advances in the filler component have resulted in microfilled composites, nanocomposites, hybrid composites, packable and flowable composites, just to name a few.

Many added innovations were later introduced in both the filler/reinforcing systems and the resin matrix-forming monomers. Various transparent mineral fillers are employed to strengthen and reinforce composites as well as to reduce curing shrinkage and thermal expansion. These include borosilicate, fused quartz, aluminium silicate, lithium aluminium silicate, ytterbium fluoride, barium, strontium, and zirconium and zinc glasses. The latter five types of fillers impart radiopacity because of their heavy metal atoms.^[24,25] Common fillers include the following:

Organic-inorganic hybrids

One approach was using silane-modified polymers as seeds for the initial growth of silica particles. This type of filler was first developed and patented at Fraunhofer Silicate Research Institute and was later patented under the name ORMOCER™, for organically modified ceramics. These materials were shown to have improved surface wetting properties and compressive strength compared with ordinary surface-modified silica. Fillers using the same concept in combination with traditional Bis-GMA monomers. This composite mix yielded lower shrinkage (0.8-2.2%) than other commercially available composites (2-3%). A distribution of particle sizes is used to maximize loading.^[26,27]

Nano fillers

Jandt et al.^[28] tried novel compositions of filler, adding titanium nanoparticles as well as Ag-Sn-Cu particles to assess their effect on mechanical properties as well as radiopacity, citing the routine use of these metals in posterior restorations. They found that the addition of these particles increased diametral tensile strength, fracture toughness, as well as radiopacity. Due to its advantageous mechanical properties, but adverse aesthetic properties, they suggested the possibility of its use in core materials as an alternative to the currently used materials.

Prepolymerized composite particles

In order to minimize the overall shrinkage of the composites upon photo polymerization while maintaining the advantageous properties of both large and small particles, an alternate filler preconditioning method has also been used. This method involved the mixing and prepolymerization of resin and filler before integration into the final composite.^[29]

Porous fillers

Fillers are most frequently used as solid particles, but porous particles have also been explored. These are meant to improve the bonding between the filler and the resin matrix through mechanical retention in the irregular pores of the filler. In addition to the mechanical improvements, the porous structures have been proposed as a method of producing bioactivity, releasing compounds from the pores over time.^[30]

Fibers, Nanotubes, and Whiskers

The mechanisms by which this occurs include whisker/fiber pinning and pullout, crack deflection, and bridging. The most widely used fibers in dental composites have been glass fibers.^[30,31]

Particle clusters

The size of the silica particles has always been a point of concern: larger sizes allowed higher filler loading, while smaller particles produced superior wear properties. Continuous distribution of progressively smaller particles can yield maximal filler loading.^[32,33]

5.4 Other Restorative cements

Silicate cement:

Silicate cement, the first translucent filling material, was introduced in 1878 by Fletcher. Its Powder contains commercial fillers prepared by fusing appropriate quantities of quartz, silica and alumina in acryolite/fluorite flux containing small quantities of phosphate which added strength and contributed to modulus of elasticity. All commercial liquids contains metals commonly aluminium and zinc, which are required to be soluble in phosphoric acid, nontoxic and incapable of forming coloured sulphides.^[34]

Zinc phosphate cement

Colloidal silicon dioxide, is used as an inactive filler in the powder and during manufacturing to aid in calcination process, and to improve the mechanical properties by increasing physical resistance and reduce polymerization shrinkage. Although the bismuth trioxide is believed to impart a smoothness to the freshly mixed cement mass, in large amounts it may also lengthen the setting time.^[15]

Zinc polycarboxylate cement

The powder contains reinforcing fillers such as 1% to 5% tin, magnesium oxide, 10% to 40% aluminium oxide, stannous or other fluoride may also be included to improve mechanical properties and provide leachable fluoride.^[1,35]

Zinc oxide-eugenol cement

The powder is basically zinc oxide with up to 8% of other zinc salts, accelerators and fillers which increase working time and strength. Rosin is added to reduce brittleness.

In two-paste materials used for temporary cementation, one paste contains zinc oxide mixed with mineral or vegetable oils, whereas fillers are incorporated into eugenol to form the other paste. An important improvement was the development of materials in which the liquid is a mixture of 2-ethoxybenzoic acid and eugenol, roughly in a 2:1 proportion. In these materials, alumina (30%) was added to the powder as reinforcing agent. Zinc oxide-eugenol cements reinforced with the addition of polymeric substances such as polymethyl methacrylate as fillers to the powder component significantly increases the strength of the cement have significantly higher strength than unmodified formulations.^[15,35]

Resin cements

Resin cements are composed of typically diacrylate resins containing 50-80% glass filler particles with most particles less than 1.0 µm in size. The most commonly used fillers here are quartz, silica, aluminium fluorosilicate glass, glass fibres, barium glass and various mixed oxides. The addition of filler particles into the dental adhesives should increase the viscosity, this would result in a thicker consistency.^[36]

G-CEM LinkAce is self adhesive cement which has new finer glass fillers; used to improve flow characteristics, physical properties and cement wear resistance while decreasing cement film thickness to just 3 microns. The concentration, size and shape of the filler material all directly and linearly influence the light scattering from resin composites.^[37]

A study done by Osorio et al.^[38] determined if experimental resin cements containing bioactive fillers may modulate matrix metalloproteinase-mediated collagen degradation of etched. The inclusion of Bioglass 45S5 particles exerted an additional protection of collagen during dentin remineralization.

Calcium hydroxide

Calcium hydroxide is now considered the “gold standard” for direct pulp capping agents.

The ingredients responsible for setting are calcium hydroxide and a salicylate, which react to form an amorphous calcium disalicylate. Fillers such as calcium tungstate or barium sulfate provide radiopacity. Some examples of such substances are barium sulphate and bismuth, and other compounds containing iodine and bromine.^[39]

6. DENTAL CERAMICS

Dental ceramics are nonmetallic, inorganic structures, primarily containing compounds of oxygen with one or more metallic or semi-metallic elements.^[1]

At the microstructural level, we can define ceramics by the nature of their composition of glass-to-crystalline ratio. Four basic compositional categories include:^[40]

- category 1 – glass-based systems (mainly silica),
- category 2 – glass-based systems (mainly silica) with fillers, usually crystalline (typically leucite or, more recently, lithium disilicate),
- category 3 – crystalline-based systems with glass fillers (mainly alumina)
- category 4 – polycrystalline solids (alumina and zirconia).

A study has shown Empress Esthetic LT (HC) and Empress CAD H

(LC) as the most translucent materials. These materials are categorized as high glass content ceramics with 40-50% leucite within the filler. The high glass content reduces the overall scattering, absorption, and reflection. The least translucent material was Vita In-Ceram zirconia (LC) which is a low glass content ceramic with a combination of 70% alumina/zirconia filler. Hence depending on the ceramic's matrix and filler, differences in absolute and relative translucency exist between multitudes of ceramic materials.^[41]

7. ENDODONTICS

Accomplishment of ideal root canal treatment is attributed to various essential factors such as proper instrumentation, biomechanical preparation, obturation, and ultimately depending upon the case post-endodontic restoration. Fillers play a vital role in various endodontic materials which aid in improving their physical process.

7.1 Sealers:

According to Grossman one of the ideal properties of a sealer is to be radiopaque, which is imparted by various fillers added to them. To reach an acceptable radiopacity inorganic fillers are added to the sealers.^[42] Furthermore they improve the physical, chemical and mechanical properties of a sealer such as viscosity and film thickness. Radiopacifying agents in order: Bismuth oxide, Zirconium oxide, Calcium tungstate, Barium sulfate, Zinc oxide.^[43]

7.2 Gutta percha

Gutta percha is the preferred choice as a solid, core filling material for canal obturation.

It is composed of trans-polyisoprene rubber with various additives including zinc oxide powder, barium sulfate, and modifiers to adjust the rheology for filling.^[42]

The inorganic fillers (zinc oxide and barium sulfate) form 76.4%. High zinc oxide levels are found to increase brittleness in the points and decrease the tensile strength. Metal sulfates are added to impart radiopacity, which is one of the ideal property for an obturating material.^[44]

7.3 Resilon:^[45]

Resilon™ Material is a thermoplastic synthetic polymer based root canal filling material. Based on polymers of polyester, it contains bioactive glass and radiopaque fillers. It performs like gutta-percha, has the same handling properties, and for retreatment purposes may be Resilon/Epiphany system is comprised of three components as follows:

- A self-etch primer, which contains a sulfonic acid terminated functional monomer, water and a polymerization initiator.
- A dual-curable, resin-based sealer: It contains fillers of calcium hydroxide, zirconium bismuth oxychloride, barium glass and silica. The total filler content is 70%.
- Resilon core material is a thermoplastic synthetic polymer based core contains bismuth oxychloride, bioactive glass and barium sulphate. The fillers content is 65% by weight which improves its mechanical properties and imparting radiopacity.

7.4 Antibacterial nanoparticles in endodontics

The antibacterial effect of metallic or inorganic nanoparticles such as silver, magnesium and zinc oxide against endodontic pathogens have been evaluated in many *in vitro* studies.

Chitosan nanoparticles are one of the commonly investigated polymeric nanoparticles in endodontics. The use of 45S5 bioactive glass nanoparticles was found to produce better antibacterial effects against *E. faecalis* than micro-sized bioactive glass particles.^[46,47]

8. MISCELLANEOUS DENTAL MATERIALS

8.1 Impression materials

One of the factors that enhances the accuracy of impressions is the copy ability, which is related to the filler size and viscosity of these materials.

Filler is added to impression compound to increase the viscosity at temperatures above that of the mouth and to increase the rigidity of the compound at room temperature.^[1]

In alginate, diatomaceous earth acts as a filler to increase the strength and stiffness of the alginate gel. Zinc oxide also acts as filler and has some influence on the physical properties and setting time of the gel.^[1,48]

Elastomers are formulated in several consistencies based on the filler content. They include lithopone, titanium dioxide, silica, sulfates, calcium carbonate, diatomite and fiberglass. They all have a flexible matrix that is filled with extender or filler to minimize the effects of polymerization shrinkage during setting on the overall accuracy and dimensional stability.^[49,50]

8.2 Dental waxes:

It is necessary to avoid excessive shrinkage and expansion caused by a temperature change. For this reason, organic filler is added to certain wax formulations. Such fillers should be completely miscible with the components of the inlay wax during manufacture, and they should not leave an undesirable residue after burnout. The addition of inorganic silica filler up to 10% in paraffin and beeswax blend could increase hardness and decrease melting point matrix, increase hardness, and reduce thermal expansion. Bentonite is one of inorganic filler. The application of calcium bentonite as a filler in carving wax increase physical and mechanical properties of carving wax.^[51,52]

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