CONTEMPORARY DENTAL MATERIALS*

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Restoring the morpho-functional capacity of destroyed and compensations of lost teeth present a challenge for the synthesis of new more effective materials and the improvement of those already on the market. The aim of the manuscript was to present the advantages and disadvantages of contemporary acrylic polymers, dental ceramics, and composites, based on literature data and published research results. The choice of the adequate material is crucial for making not only dental but any product. The synthesis of new materials and the development of new technological procedures have accelerated the advancement of dentistry. Therefore, today's dental compensations faithfully imitate natural teeth and can fully perform their intended function.

Keywords: acrylates, dental ceramics, composites

Introduction

Restoring the morpho-functional capacity of destroyed teeth and the compensation for lost teeth represents a challenge for the synthesis of new, more efficient materials and improvement of the ones already present in the market. Namely, given the already established and still successfully used methods of teeth preparation and treatment, as well as the technical design of dentures, it is not pretentious to say that the development of dentistry is, in fact, the development of dental materials and new production technologies. Nowadays, there are different types of materials used in dental practice which can roughly be divided into *build-up materials* (acrylic polymers, composites, ceramics, metal alloys) and *auxiliary materials* (plaster, wax, thermoplastic, impression and refractory die mass).

The synthesis and improvement of new materials are the result of multidisciplinary action of various scientific disciplines where dentists set priorities with a clinical approach, whereas for the final product it is necessary to define the technology of making, as well as physical, mechanical and biological quality control. In this way, technology has been clearly and unequivocally incorporated into dentistry. On the other hand, the knowledge of the composition and method of making dental materials is necessary for a concise determination of their indications and contraindications in order to minimize possible errors.

The aim of the manuscript was to present the advantages and disadvantages of contemporary acrylic polymers, dental ceramics, and composites based on literature data and published research results.

Acrylates in dentistry

Acrylates are the most commonly used polymers in dentistry, mainly for making dentures and orthodontic devices. Even though this material was introduced a century ago, it is still considered modern. Excellent mechanical characteristics and optimal biocompatibility are ensured by their macromolecular structure.

Acrylic polymers are poly(acrylic acid) esters. Those are transparent matters of different viscosity, which primarily depends on the type of the substituent which is included in the composition of polymers. Based on consistency, they can be hard (brittle) and soft (resilient) [1,2]. Based on the way of the polymerization initiation, acrylates may be classified as heat-, cold-, light- and microwave-polymerized.

Acrylates used in dentistry are usually in sets containing essential components, i.e. powder and liquid. The basic substance of the liquid of the acrylic set is mainly methyl methacrylate (MMA), the carrier of the polymerization process. The adverse reaction of premature MMA binding is achieved by adding a polymerization inhibitor such as hydroquinone or methoxyphenol [1,2].

Nowadays, cross-linked acrylates whose chains are interconnected into a spatial, stereospheric network are mainly used. The most used crosslinkers are ethylene glycol, dimethacrylate (EGDM), trimethylolpropane trimethacrylate (TMPT), 1,4-butanediol dimethacrylate (1,4-BDMA) and 1,6-hexanediol dimethacrylate (1,6-HDMA) (Figure 1). Crosslinking improves physico-mechanical properties of acrylic materials. The presence of comonomer crosslinkers must not exceed 15% of the total amount of monomers [3-5]

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Figure 1. EGDMA as a crosslinking agent in a MMA polymer.

Accelerators are chemical substances which are added to the monomer, in case the acrylate binding process is stimulated chemically. Their action triggers the polymerization reaction by activating free radicals of the initiator which is involved in the composition of the acrylic powder. Tertiary amines and sulfuric acid, as well as its more stable salts, may act as accelerators. In practice, the most commonly used polymerization accelerator is N,N-dimethyl-p-toluidine [1-4].

The main ingredient of the powder is poly(methyl methacrylate) (PMMA) or poly(ethyl methacrylate) (PEMA). The polymer powder is obtained in two ways: (1) the granular type of polymer, i.e. grains of irregular shape and uneven size are obtained by mechanical shredding of larger polymer pieces; (2) the second grain type, i.e. micropearls are obtained by the monomer polymerization in water. In that case, the powder is comprised of round grains of even size which are easy and fast to polymerize [6]. A polymerization initiator, plasticizers, colours, and pigments are further added to the powder.

Polymerization initiators are peroxide or azo initiators that induce a free-radical chain reaction, most commonly dibenzoyl peroxide which is added to the powder in the amount of 0.2 to 1.5% [7]. Plasticizers provide flexibility, and phthalates such as dibutyl-, diethyl- and dicyclohexyl phthalate are among the most frequently used ones [1]. Colours and pigments are part of the composition of a polymer grain. The most commonly used are cadmium (cadmium sulfide and cadmium sulfoselenide) and iron salts, titanium oxide and zinc oxide. The addition of inorganic substances, grains and glass fibres and different inorganic particles of zirconium silicates, aluminium oxides or silicone carbides into the conventional acrylic mass improves the physical properties of the material [8-11].

The basic component of light energy-bonded acrylic polymers is urethane dimethacrylate (UDMA) in the presence of micro-silicate filler [12]. They are often available for commercial use in the form of single-component

plates or gels, which facilitates their adaptation to the model, as well as their bonding [13, 14].

According to their composition, soft acrylates are PMMA, PEMA, poly(n-butyl methacrylate) (PBUtMA), as well as a mixture of methacrylic copolymers and terpolymers. The liquid is a mixture of 30-60% of ester plasticizers (dibutyl phthalate, butyl-glycolate) and ethanol, which serves as a solvent and whose content amounts to 4-60% of the total mass [15]. Ester plasticizers make these materials soften at body temperature. Unlike other acrylic polymers, soft polymers change their composition and consistency (they harden) during their function in the orofacial system, which is in accordance with their assigned indications, oral tissue conditioning, and the inflammation treatment [15].

Certain components of acrylic materials have the ability to leave dental compensations and diffuse into saliva, via which they have a toxic or immunological effect on the oral tissues causing a local inflammatory reaction of the oral mucosa [16]. A potential cause of acrylate toxicity is its unpolymerized components, as well as by-products of the polymerization reaction [16-18]. Residual monomer represents a certain amount of monomers not bound during the polymerization procedure [17]. Incomplete polymerization reduces not only the physico-mechanical but also the biological quality of dental compensations. Namely, it has been proven that the unbound MMA allergen is also a tissue irritant [18-21]. A certain amount of residual MMA was found in dental prostheses worn for seventeen [22] or even thirty years [23]. The hydrolysis of MMA produces methacrylate acid which is proven to have a cytotoxic effect [24, 25]. Potential toxicity is also associated with butyl methacrylate (BuMA), ethyl methacrylate (EMA), and UDMA, as well as with comonomer cross-linkers (EGDMA, isobutylene methacrylate (IBMA)) [26, 27]. Eventually, residual monomer leaves the acrylic compensation by diffusing and dissolving into saliva. The amount of liberated monomer is proportional to its total residual amount in the acrylate matrix, although a certain amount of unbound monomer remains trapped in the polymer structure, never diffusing into the external environment [28]. Acrylic compensation is primarily abandoned by a residual monomer from its surface areas [29]. Acrylates impregnated by carbon, silicone or glass fibres release greater amounts of unpolymerized monomer due to porosity and faster wear and tear [2].

To stay within the limits of acceptable, the amount of residual MMA should range from 1 to 3% [28]. According to the standard (ISO 1567:1999), the maximal allowed amount of residual MMA for heat-polymerized acrylates is 2.2%, whereas for cold-polymerized acrylates it amounts to 4.5%.

An essential characteristic of acrylic materials is *glass transition* (T_g), i.e. the temperature at which amorphous polymers change from a solid, glassy state into a flexible, gummy state. It is the highest temperature dentures may be exposed to, without any risk of deformation. Regard-

ing heat-polymerized acrylates, T_g ranges from 97 °C to 115 °C, whereas it is considerably lower for cold-polymerized acrylates, 70 °C [30, 31]. The polymerization of acrylates is considered more complete if it occurs at temperatures closer to T_g [32].

Since the polymerization process of cold-polymerized acrylates is carried out very quickly, without pressure and at the temperature much lower than T_g of acrylate polymers, it is impossible to dissolve all of the polymer into the monomer [32]. Cold-polymerized acrylates have a less homogeneous and less porous structure than heat-polymerized acrylates, and therefore a higher percentage of potentially toxic substances [33-35]. Given that a higher amount of residual monomer is found in the composition of cold-polymerized acrylates than in heat and microwave-polymerized ones, its release into the oral environment is more intensive [36-39].

The amount of residual monomer in acrylic materials can be reduced using post-polymerization methods which enable additional conversion of monomers into the compact structure of polymers [40].

Microwave polymerization can reduce the amount of residual monomer up to 25% [40-42]. The reduction of the residual monomer amount is the consequence of subsequent polymerization and monomer evaporation in the microwave oven [26]. Water-bath post-polymerization may be an efficient way to diminish potential acrylate toxicity by diffusing unpolymerized monomer in water, additional polymerization at places with active radicals, or by hydrolysis to methacrylic acid [43, 44]. Kostić et al. showed higher residual monomer values in cold-polymerized acrylates, as well as the fact that its amount in post-polymerized treatments may be reduced to a clinically acceptable level [45, 46].

The same group of authors also examined the adherent characteristics of acrylates after immersion into saliva solutions and implantation into the subcutaneous and muscle tissue of experimental animals [47]. Soft acrylic material exhibited exquisite adherence to the surrounding content, which consecutively decreased its biocompatibility [47]. Kostić et al. proved the inflammatory effect of newly-made complete dentures on the oral tissues with the increase of proinflammatory cytokines in the saliva of patients [48]. On the other hand, as it can often be found in the literature, the allergic effect of heat-polymerized acrylate used for making dentures has not been proven [48]. A new generation of light and microwave-polymerized acrylates shows satisfactory biocompatibility and good physico-mechanical properties, therefore they can replace cold-polymerized acrylates in all indications.

Dental ceramics

Different types of dental ceramics have been introduced into practice in recent decades. They cover a wide range of indications, starting from veneers which are cemented to unprepared teeth, to large dental bridge constructions which serve for complete occlusal rehabilita-

tion. Dental ceramics is a material for making all types of fixed dental compensations, whether it is porcelain fused to metal by forming oxides or porcelain used individually (all-ceramic restorations).

The term "ceramics" is complex and refers to numerous materials, including metal oxides, borides, carbides, nitrides and complex mixtures of these materials. Dental ceramics is isotropic, there is no melting point, but the softening interval. The structure of ceramics is crystalline with a proper periodic arrangement of components' atoms bound by ionic or covalent bonding. Even though ceramics is a hard material it is relatively brittle at the same time, but in some cases it has mechanical properties similar to human dentine. Therefore, it is said that ceramics is strong in compression but weak in tension [49].

Dental ceramics is comprised of silicate materials, quartz, and kaolin. In most cases, orthoclase ($K_2O \cdot Al_2O_3 \cdot 6 SiO_2$) makes 75-85% of the total weight of ceramic materials [50, 51, 52]. Among other forms, silicon dioxide (SiO_2), which acts as the basic building block of glass, can appear in a non-crystalline fused form, whose high melting point is determined by a three-dimensional net of silicon tetrahedra bound by oxygen atoms (Si-O-Si) [52]. Its volume fraction amounts to 12-22%, and it is very hard, which is crucial for the mechanical stability of ceramics. Kaolin ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$) is an aluminium silicate which easily absorbs water, therefore it is easy to shape, and ensures stability. Kaolin connects all dental ceramics into one building unit and constitutes 1-4% of the ceramic powder [52].

Up to 1% of pigments are added to basic components. These pigments are metal oxides, stable at the temperature of sintering and chemically inert to other components: titanium oxide – dark yellow colour, uranium oxide – orange/yellow colour, cobalt oxide – blue colour, chromium oxide – green colour, nickel oxide – grey colour, iron oxide – dark colour, iridium oxide – black colour, etc. [53]. The addition of organic compounds (dextrin, starch, sugar) improves the plasticity of ceramics, whereas catalysts decrease the firing temperature [54].

The diversity of dental ceramics structure is seen not only in different building elements but also in different microstructure which they build inside them [55]. Dental ceramics is mostly made of the glass matrix in which different types of crystal are incorporated in a smaller or larger number, with a regular or irregular arrangement. The material that does not contain crystal is known as simple glass. If the microstructure is glassy, i.e. non-crystalline, it is transparent. If the structure is crystalline, it is opaque. The factors which also add to translucency are a particle size and density, a refraction index, etc. [56]. The material for complete ceramics compensations contains 35-90% of crystal, therefore it has better mechanical properties [57,58]. There are also types of ceramics that are dense polycrystalline structures without the glass matrix.

The quality of any ceramic material depends on the

microstructure and phase composition, mainly of the grain size and the ratio of ceramic and glass in the matrix, as well as on the preparing and processing procedure, temperatures of sintering, heating and cooling rate, etc. Therefore, from the viewpoint of practice, dental ceramics can be divided according to the microstructural composition or the processing method used in a dental laboratory.

The classification of dental ceramics according to the microstructure is based on the amount and type of the crystal phase and the composition of glass. Regardless of theoretically possible infinite variability in the material microstructure, dental ceramics can be divided into four basic categories:

Category 1: glass-based systems. This type of systems is made of silicon dioxide-based materials (silica or quartz) and different amounts of aluminium oxide (alumina). Aluminosilicates found in nature, containing different amounts of potassium and sodium are known as feldspars. Feldspar ceramics is "classic" dental ceramics of great aesthetic value, but with poor physico-mechanical properties. A large amount of the glass matrix enables colour and transparency manipulation [59].

Category 2: glass-based systems (mainly silicates) with crystalline fillers. This category has a wide range of glass-crystal ratio and different crystal types within the glass matrix: leucite, lithium-disilicate, and fluorapatite. Lithium-disilicate crystals are formed by adding lithium oxide, and leucite by increasing the potassium oxide content of aluminosilicate glass. In the literature, this type of material is known as glass-ceramics [59, 60].

Glass-ceramics is a polycrystalline material which is produced by controlled glass crystallization. It is a highly transparent material of great aesthetic value due to the glass matrix and crystal ratio [61]. When the light goes through glass-ceramics, it is less dispersed and provides an optical effect similar to the one of enamel and dentin [53]. Given that it is susceptible to cracking under heavy pressure, this type of ceramics is not used in side areas [62, 63].

Category 3: crystalline-based systems with glass fillers (mainly alumina). These materials are generally produced in the form of a porous matrix (ceramic "sponge") whose pores are further filled using a material of another phase, lanthanum aluminosilicate glass. The capillary action is used so that liquid or molten glass can flow in all pores and produce a dense material which permeates [64].

Category 4: polycrystalline solids (alumina and zirconia). Solid sintered monophasic ceramics is a group of materials formed by direct sintering of crystals together without any intervention matrices, to form a dense, polycrystalline structure without glass. There are several different processing techniques that enable the production of solid sintered aluminum oxide or zirconium materials [64, 65].

Zirconium oxide ceramics (zirconia) is crystalline zirconium dioxide (ZrO_2). Zirconium dioxide is present in

the Earth's crust in the form of a rare mineral of baddeleyite. Its mechanical properties are very similar to the properties of metals (ceramic steel), and its color is similar to the color of teeth [66]. The resistance of zirconia to traction can be as high as 900-1,200 MPa, whereas the resistance to compression is around 2000 Mpa [67]. Zirconia is considered biologically acceptable [68, 69], and due to its exceptional mechanical and physical properties, it can be used in the side sector as well as in oral implantology [65].

Zirconia is partially stabilized by adding a small amount of calcium oxide, magnesium oxide, yttrium oxide, and cerium oxide to prevent the phase transformation during sintering, and also to improve its mechanical properties [67, 70]. Zirconia is most commonly modified using yttrium oxide (Y_2O_3) which results in tetragonal zirconia polycrystal (Y-TZP).

The same forms of dental ceramics may be processed using different methods, and for didactic reasons they can be divided into:

1. Powder or liquid-based systems;
2. Blocks of glass-based systems that can be pressed;
3. CAD/CAM or dense, networked, mainly crystalline (alumina or zirconia) systems [71].

Even though the material can have the same chemistry and microstructure, the method of processing used for restoration may enhance or weaken the final properties and clinical success. In particular, the processed material blocks proved better than the same material in the form of a powder [72].

Conventional powder ceramic materials are glass-based, or the combination of glassy and crystalline components. Those are materials for veneering ceramic or metal cores, or for making entire ceramic crowns. They are manually mixed with deionized water or a special liquid for modeling. Sintering of ceramics is done in the vacuum oven, and the very process depends on the conditions of the environment, skills of technicians and a selected cycle. Sintering is the name for the process of agglomeration and consolidation of ceramic powders (particles) through heating. The slip casting method implies pouring powdered dispersions into a plaster mold after melting wax which helps remove water and create a homogenous block, i.e., a cast.

Pressed ceramic compensations are made using the injection press method. Monochromatic porcelain or glass-ceramic ingots are heated so that the material could flow under pressure into a mold which is formed using the conventional lost-wax technique.

Computer-Aided Design / Computer-Aided Manufacture (CAD/CAM) is a computer-aided technology for taking impression and making ceramic compensations using the milling technique [72]. Ceramic blocks are made of powders mixed with a binder and then pressed into a mold or extruded in the form of a block or cylinder. When heating blocks, the binder is removed and high-density ingots are obtained [58].

Dental ceramics can find its application in the produc-

tion of porcelain-fused-to-metal crowns. It is a form of conventional dental powder ceramics which is applied in layers to metal infrastructure in the form of a cap. Depending on its purpose, this type of ceramics has several layers: basic, dentin, enamel and translucent. The first layer of ceramics is especially important since it has to make a good bond with metal, and being opaque, it has to cover a dark, unnatural colour of the metal substrate [52].

Composites in Dentistry

Composites constitute a group of materials which occupy the central position in restorative dentistry owing to their aesthetic, physico-mechanical, and biological properties. They are used to fill dental defects caused by caries, fractures or morphological anomalies of the tissue, as well as in orthodontic and prosthetic therapy.

Composites are cross-linked materials which possess a three-dimensional structure comprised of two or more chemically different components with a clear borderline. They are comprised of the organic part (polymeric matrix), the inorganic part (fillers), and a silane coupling agent (adhesive) [73].

Figure 2. Organic matrix of dental composites.

The organic matrix may be based on dimethacrylate, silorane or ormocer. The most common methacrylates in the polymeric matrix are bisphenol-A-glycidyl methacrylate (Bis-GMA) and triethylene glycol dimethacrylate (TEGDMA). The Bis-GMA molecule contains two centrally located benzene rings and terminal methacrylate groups through which free radical polymerization takes place. The benzene rings provide rigidity and good mechanical properties to the composite. Bis-GMA exhibits high viscosity due to the presence of hydroxyl groups, therefore it is combined with low molecular weight monomers (diluent), such as TEGDMA or UDMA (urethane dimethacrylate) (Figure 2). The role of a linear monomer TEGDMA with two terminal methacrylic groups is cross-

networking which increases the strength of the composite and reduces its solubility in saliva [74, 75]. On the other hand, a higher percentage of TEGDMA in the organic matrix increases the polymerization contraction [76].

The silorane resin-based organic matrix, made of molecules of siloxane and an oxirane ring, is polymerized by a cation reaction. The advantage of this type of composite is the low polymerization contraction, as well as lower absorption of liquid and solubility in the oral environment [77, 78].

The ormocer matrix is a hybrid of the organic and inorganic component linked via multifunctional silane molecules [79].

A filler is the inorganic part of composites of mineral origin such as quartz, borosilicate glass, barium silicate, aluminium silicate, lithium aluminium, barium fluoride, zirconium, ceramic powder or glass particles, which are added to the organic matrix in order to improve its hardness and abrasion resistance, as well as some physical characteristics such as water absorption and solubility [80]. Filler particles are of different shape and dimensions, spherical or irregular, with micrometric (1-50 μm), submicronic (0.1-1 μm) and nanometric dimensions (5-100 nm).

The binding of the organic matrix and the filler results in an organic-inorganic compound whose molecules have double polarity, therefore they form ionic bonds (Si-O-Si) with the filler. On the other hand, they use their inorganic part to form covalent bonds with the matrix. The most commonly used binding agents are organosilanes, especially γ -methacryloxypropyltrimethoxysilane, abbreviated as silane. The percentage of silane in composite materials is between 1 and 5% [81, 82]. Composite materials also contain colour stabilizers, inhibitors and initiators, X-ray contrast agents etc. [83].

The polymerization of composite materials is initiated chemically or by light energy, most often by a blue light-emitting diode (LED) [84].

The key problem of light polymerization is the depth limit at which a satisfactory conversion of monomeric molecules into a polymeric network at 2 mm will be carried out [85]. Insufficient polymerization reduces the mechanical and biological quality of composites [81-86]. Composites are polymerized in layers, and the polymerization is often not complete especially at the site of their bonding, which leads to cracking of fillings and settling of microorganisms, especially if the shades of the material are darker [87]. To overcome this problem, the so-called bulk-fill composites have been introduced. Compared to conventional composites, they possess higher translucency which allows better light transmission even for layers up to 5 mm of thickness [85-88]. A degree of conversion of composites directly affects their mechanical properties and biocompatibility. As the conversion value is on average 55-65%, a large amount of residual monomer remains in the material [80].

Bioactive ceramics (bioceramics) are defined as "bioceramics that bond directly with bone without having

fibrillar connective tissue between them." Bioactive ceramics are hydroxyapatite (HAP), bioactive glass, and bioactive glass ceramics [89].

Conclusion

The choice of adequate material is crucial for making not only dental but any product. The synthesis of new materials and the development of new technological procedures have accelerated the advancement of dentistry. Therefore, today's dental compensations faithfully imitate natural teeth and can fully perform their intended function.

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Izvod

SAVREMENI STOMATOLOŠKI MATERIJALI

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Vraćanje morfofunkcionalnog kapaciteta destruisanih i nadoknada izgubljenih zuba predstavlja izazov za sintezu novih delotvornijih materijala i poboljšanje onih koji su već na tržištu. Cilj rada bio je da se na osnovu podataka iz literature i objavljenih rezultata istraživanja prikažu prednosti i nedostaci savremenih akrilatnih polimera, zubne keramike i kompozita. Izbor adekvatnog materijala od ključnog je značaja za izradu bilo kog, pa i stomatološkog proizvoda. Sinteza novih materijala i razvoj novih tehnoloških procedura ubrzale su napredak stomatološke struke, te današnje zubne nadoknade verno imitiraju prirodne zube i u potpunosti mogu da obavljaju njima namenjenu funkciju.

Ključne reči: akrilati, dentalne kermake, kompoziti