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Resin modified calcium silicate cements - novel formulations in clinical dental practice

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Resin modified calcium silicate cements or hybrid cements refer to the fourth generation of materials. These materials are a combination of calcium silicates and resin. The available information on novel formulation introduced hybrid cements in clinical practice is presented. The directions of the interactions of biomaterials with the dentin are discussed. The available information is critically reviewed and discussed concern basic physicochemical properties of Resin modified calcium silicate cements such as Compressive strength, solubility, dimensional changes and others. Conclusion: The data on the physicochemical properties of the presented novel formulations of resin modified calcium silicate cements in this article are limited and it are data from the manufacturers. Independent in-depth studies are needed for them in order for the dentist to select the most suitable, tailored to the individual clinical case. The examined and critically analyzed data refer mainly to the most studied representative of the 4th generation- Theracal LC.

Keywords: Pulp-capping materials, Resin modified calcium silicate cement, Physicochemical properties.

Introduction

The first bioactive calcium silicate cement MTA Original-ProRoot® MTA (Dentsply, Tulsa Dental, Johnson City, TN, USA) was developed by the Torrebinead and Dr. White team [1]. This cement was introduced in dental practice in 1998. Calcium-silicate cements (CSCs) are bioactive materials with a variety of applications in dentistry such as direct pulp capping, perforation repair, retrograde root-end filling, regenerative therapy, etc. [2]. CSC should have cell compatibility, a high antibacterial effect, ensure quality tissue healing and stimulate the formation of reparative dentin [3, 4].

Resin modified calcium silicate cements (RMCSCs) or hybrid cements are a new group of CSCs and belong to the fourth generation of materials. This type of material is a combination of calcium silicates and resin. RMCSCs are mainly used in direct and indirect pulp capping. Direct pulp capping (DPP) is a biological method, that aims preserve the vital, functional integrity of the dental pulp (DP) [5]. Pulp-capping materials are applied directly to the exposed pulp and therefore must be biocompatible. The cement has to have the capacity to withstand masticatory forces. An important advantage of vital teeth is that they have better protective resistance to masticatory force than root canal treated teeth [6].

RMCSCs are materials, that are hardened by two types of reactions: light polymerization and hydration. The hydration process is slow because the reaction requires moisture. RMCSCs are supplied with the moisture neccessary for hydration from the contact medium. But this process depends on the rate of diffusion and porosity in the material [7, 8]. Main advantages of these materials are the possibility a one visit treatment approach, less risk of infection, reduced of working time and excellent mechanical strength [9, 10].

The main representative of this generation is TheraCal LC (Bisco Inc, Schamburg, IL, USA), a material introduced in dental practice in 2017. According to the manufacturers, the chemical composition is represented by 45% Portland cement III type, 10% dicalcium silicate, a radiopaque substance from barium zirconate and 45% resin mainly from BisGMA, polyethylene glycol and photoinitiators [11, 12]. The material is a one-component paste that hardens after light polymerization for 20 seconds.

Recently, Bisco introduced a new dual resin modified calcium silicate cement TheraCal PT, with the basic indication of pulpotomy in pediatric dentistry. It consists of synthetic Portland cement, polyethylene glycol dimethacrylate (10-30%), Bis-GMA (5-10%) resin matrix, barium zirconate (1-5%) initiator (<1%)[13]. The material hardens after photopolymerization in 10 seconds.

Advances in the science of bioactive materials have led to the creation of many new materials

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Novel Formulations of Resin Modified Calcium Silicate Cements

New RMCSCs have been introduced into dental practice. There was limited knowledge of the biochemical properties of these new pulp capping materials.

Endo/Tech™ Canada has introduced NovaCal into clinical practice. According to the manufacturer, the compressive strength of this novel material is >90 Mpa, the amount of calcium ions released in 24 hours is 420 μg/cm² and it has a pH-12. It is a one-component paste that it hardens in 40 seconds by using a polymerization unit (wavelength range 400-500 nm) with a light intensity of at least 1000 mW/cm.

BisiCAL(Centrix)Dental's South Africa has also been introduced into dental practice .It is a one-component paste, that it hardens after polymerization in 40 seconds. According to information from the manufacturer of this new hybrid cement, the amount of free calcium ions in 24 hours is 420 μg/cm^2.

The new patented formula from Nexobio Korea is MTA Cem LC. It consists of tricalcium silicate particles in hydrophilic monomers and a radiopaque substance -barium sulfate.The curing time by light polymerization is 20 seconds.

Recently proposed in dental practice is another new hybrid cement - **Bright MTA** (Genoss, Suwon, Korea). It consists of calcium silicates (30-50%), polyethylene glycol dimethacrylate (10-30%), barium sulfate (15-30%), silica nanoparticles (30-60 nm), (8-15%) resin without containing Bis-GMA. Its curing time is by light polymerization for 20 seconds [14]. This material has a nanostructure composition which is the reason for its lower physical properties compared to TheraCal LC [14].

A new **BioCal Cap** was introduced into clinical practice in 2019. According to the manufacturer, the material is a combination of 35-45 wt% hydrophilic resin, Portland cement (PC) and barium sulfate (7-12%). The curing time by light polymerization is 40 seconds [15].

For the above meantiomed new materials, there is scarce information available in the literature regarding their qualities and characteristics [14, 16, 17].

Dentigrate LC (Dent ACT India) paste consists of a mixture of calcium oxide, silicon oxide, bismuth oxide, methylacrylate resins, photoinitiators and stabilizers [18].

Other newly introduced hybrid materials are: **MTA Pulp Cap** (Cumdente GmbH), **ReViCal** (Yohan, Irag), **Oxford ActiveCal PC** light-cure resin reinforced pulp capping material (GmbH, Elmshorn, Germany). For these materials there is no information in the literature.

The information presented here for most hybrid materials is mainly from the manufacturers. There is a lack of real data regarding their properties and biological potential for a healing process after their use.

The cytotoxicity of hybrid CSCs was attributed to their resin content. The presence of organic components in the composition of hybrid CSCs can have a negative impact and lead to unsatisfactory clinical effectiveness. It is possibly due to the free unpolymerized monomers. And free monomers may diffuse into the dental pulp and cause a detrimental effect [19, 20]. The degree of conversion of monomers from the composition of hybrid CSCs is related to the final mechanical properties [21, 22]. Eluted monomers negatively influence mechanical properties [23] The increase in curing time from 20 to 40 seconds led to a decrease in residual monomer content [24]. However, the increase in curing time during polymerization of light cured calcium silicate cements could lead to harmful influence on dental pulp by the heat generated [25]. So, the heat generated during the polymerization could lead to irreversible pulpal damage. The main factors affecting the amount of generated heat are the type of light sources (LS), the light intensity of LS, the position of LS, the distance between the material surface and LS, the exothermic nature of the material, curing time, remaining dentin thickness and the thickness of the layer of hybrid CSCs [26, 27]. To achieve complete polymerization, the thickness of the layer of hybrid CSCs should be small enough, about 0.65 mm. according to Nilsen, B et al. [28]. According to literature data, full polymerization of TheraCal LC requires a layer thickness of 2 mm, and for Bright MTA the appropriate layer thickness is 2.1 mm [14]. According to the manufacturer, the thickness of the layer of TheraCal LC should be 1 mm and it polymerizes in 20 seconds [13]. Gandolfi MG et al. [29] reported the appropriate TheraCal layer thickness to achieve full polymerization is 1.7 mm.

Increased distances between the tip of the LS and the surface of the material can lead to incomplete polymerization [30]. It was found that the distance of 2 mm from the tip of the LS to surface of the TheraCal LC did not increase the temperature during polymerization [31].

The available information regarding the cytotoxicity of these hybrid CSCs in the literature is conflicting

According to Bortoluzzi EA et al. [32] TheraCal LC exhibits significant damaging effects on human dental pulp stem cells in direct contact with the pulp. Similar data were reported by other authors [33]. In contrast, Buonavoglia et al. [34] did not find cytotoxicity in osteoblast cells treated with TheraCal. They reported that the cells were properly organized. In our previous research through thermogravimetric analysis it was revealed that the new hybrid material BioCal-Cap was more resistant in comparison to TheraCal LC because no volatile compounds were found. The results after gas-chromatography of organic and inorganic and mass-spectrometry SIM mode analysis revealed that the extract of TheraCal LC contained nonreacted photoinitiators, while the same analysis for BioCal-Cap revealed no nonreacted monomers of HEMA (17). And

according to the results reported by Nilsen et al. [10] TheraCal LC contains high levels of 2-(dimethylamino) ethyl methacrylate and camphorquinone, for which there is no proper, relevant information from the manufacturer.

Bioactivity of Resin Modified Calcium Silicate Cements

The success of DPC depends on different factors and conditions. A very critically important factor is the correct choice of pulp capping agent, according to its composition and qualities, refracted based on the individual characteristics of the patient. Bioactivity potential is described as the cellular effects induced by active substances in biomaterials. Dental pulp capping agents should be bioactive towards the pulp tissue and should to stimulate the DP to form tertiary dentin [35, 36]. The release rate of calcium and hydroxyl ions is a key factor for successful dentine bridge formation. [37]. The release of calcium and hydroxyl ions plays an important role in cell differentiation and proliferation of human dental pulp cells and the formation of mineralized hard tissues. It creates high alkaline media and determines the antimicrobial activity of the materials [38].

Hydroxyl ions stimulate the release of alkaline phosphatase and bone morphogen protein, which are involved in the mineralization process [39].

The available information on the release rate of calcium ions from RMCSCs in the literature is conflicting.

According to Camilleri J. [40] Theracal LC has the lowest leaching of calcium ions in comparison to Biodentine.The reason for this is the availability of a resin-modified matrix in its structure. They found that the level of released calcium ions from TheraCal LC is significantly low and stops after the 4th day of its application.

In contrast, other authors Nagham A. et al. [41] found that there were no statistically significant differences in the amount of calcium ions released from Biodentine and those from TheraCal LC over a 30-day period of comparison

Interaction Between Dentin and RMCS

Quality adhesion or interaction of biomaterials with dentin is of key importance to prevent microleakage, bacterial invasion and adverse outcomes of biological treatment [42]. Pulp capping materials should provide an adequate seal to the pulpal wound and dentin, remain in place under chewing pressure [43].

Adhesion to the dentin depends on the type of bioactive materials and the type of pretreatment of the dentin surface [42]. There is less literature on the nature of the interaction of hydraulic cements with dentine. According to Savas S et al. [31] TheraCal LC chemically bonds to the dentin, initiating the formation of hydroxylapatite crystals. Similar evidence that TheraCal LC bonds chemically to the moist dentin has been reported by others [44]. The quality of coronal restoration is another critical factor for the success of direct pulp capping.

TheraCal LC has structure, similar to that of composites and theoretically, the bonding mechanism of composites to calcium silicate pulp-capping agents is comparable with mechano-chemical adhesion to hard dental tissues [34]. It has been reported that the high shear bond strength of TheraCal is due to the hydrophilic methacrylate monomers in its structure that increase chemical adhesion to the dentin and form a strong interface between TheraCal and the bonding surface [46, 47]. In contrast, Wang et al. [48, 49] reported low adhesion strength of TheraCal LC to dentin. Similar data were found by other authors, although the authors followed the manufacturer's recommendations for applying the hybrid CSC to the wet dentin [50]. It was found that the bond strength of TherCal LC to the dentine is 1.27 kgF [51]. An in vitro study found that TheraCal LC shows a significantly larger size of internal gap at the dentinal surface compared to MTA and Biodentine [52].

Physicochemical Properties of RMCSC

The study of the physicochemical properties of pulp capping agents such as pH, solubility, water sorption, compressive strength etc. has a key role in the healing process and pulpal response [53]. Adding a small amount of polymer to the cement mixture can significantly enhance the performance of the final material. Polymers can significantly improve the tensile strength, flexural strength, flexibility, compactness and durability of cement-based composite materials [43].

In addition to their regenerative potential, biomaterials must possess sufficient physical properties, which are important for the stability and durability of the restoration. A key factor in achieving complete healing success which direct pulp capping (DPP) is the requirement that it have excellent physical-chemical properties. CSCs must hermetically seal the communication with the dental pulp (DP) but also have sufficient bond strength with both the dentin and the final restorative material. In addition, the materials must have excellent mechanical properties such as: dimensional stability, low solubility and water sorption, adequate compressive strength, etc. [43].

The properties of dental materials can be divided into three categories: physical, chemical and biological. Mechanical properties are a subgroup of physical properties. Mechanical properties refer to a material's ability to resist force [55, 56].

Compressive Strenght

The physical properties of materials are determined by

their response to the application of force and/or pressure, which leads to reversible or irreversible deformations after the removal of the action of the applied force. An important property that may affect the clinical performance of DPC is its compressive strength [57]. The basic mechanical quality of materials is compressive strength, because chewing forces are compressive in nature [58].

According to ISO 1:9917-1. the minimum compressive strength that the pulp capping agents are required to have is 50 MPa [59], but these agents must have a compressive strength similar to that of dentin and the restorative material used [60]. The ideal pulp capping material compressive strength is 300 MPa similar to dentin [60]. It is believed that the compressive strength of CSC is directly dependent on the stage and degree of hydration of the material, the content and size of the inorganic filler particles [61, 62, 64]. The immediate average compressive strength of TheraCal LC was found to be 84.47 MPa and increased after 24 hours to 114.40. The authors conclude that the upper pulp capping material has adequate compressive strength which is a condition for ensuring sufficient support of the restorative materials [63]. Similar data on significantly high compressive strength at 24 hours of 78.78 MPa was also found by Taynnara Liceski Gasperi et al. [64]. However, they found a reduction in compressive strength to 69.06 MPa on day 30. Similar data on the initial high compressive strength of TheraCal LC was also found by Ula A. Fathi [65]. A decrease in the compressive strength values of the material with time was also found by Aziz A et al. [66].

Porosity, solubility, water sorption and volume changes

CSCs are porous materials, and higher porosity of the materials is associated with greater reactivity and capacity to release biologically active molecules, but high porosity is associated with a negative effect on the mechanical resistance and marginal adaptation of the material [67, 68]. Porosity has been shown to have an impact on numerous other factors including adsorption, permeability, strength, and density [69]. The latter authors found that HCSC-TheraCal LC has low porosity and high mechanical strength compared to MTA and Biodentine. Other authors find similar data [70]. Porosity is a key properties for the microstructure of the materials. It refers to the volume of empty space in a material, which can include fluid or air, as a percentage of the total volume [70].

The solubility and water sorption of CSCs are the main physical properties that determine the stability and durability of restorations and, accordingly, the risk of degradation and failure [71-73]. Solubility is the amount of substance that will dissolve in a given amount of solvent [74]. The solubility of CSC is between 12 and 38% and depends mainly on the chemical composition [71]. According to ISO 4049 standard, the solubility of the hardened calcium silicate material must not beer than 3% exceed 40 g/mm^3. The solubility of TheraCal LC and Bright MTA was found to be low at 7.5 g/mm^3 [14]. Similar low solubility data within 3.59% of TheraCal LC was also found by Nagham A et al. [41]. These authors found water sorption for TheraCal LC at 11.53%. It was found that TheraCal LC is characterized by low water sorption, porosity and solubility [76]. The size and shape of the cement particles affect the water absorption of the material [76]. The average solubility of TheraCal LC was found to be 1.566595% over seven days [77].

Similar data for low solubility and water sorption of 2.75% and 13.96 has also been found by other authors [70]. Low solubility values of TheraCal LC of 58% and water sorption of 10.42% were found by M. G. Gandolfi et al. [29]. Similar data for low values of solubility was reported others [77].

Dimensional stability

The dimensional stability of calcium silicate cements is important. The shrinkage can lead to the loss of marginal adaptation and a gap between material and tooth with resultant bacterial leakage, whereas expansion results in cracks or the fracture of the tooth. The slight expansion may close the gaps [79]. The presence of a hydrophilic monomer (polyethylene glycol dimethacrylate) in the composition of TheraCal LC which absorbs water during the setting reaction and leads to its expansion during the setting. This expansion of TheraCal LC could enhance its penetration into dentinal tubules and hence contribute to its enhanced bond strength with dentine [64]. Hygroscopic expansion leads to better penetration in the dentinal tubules [83].

The dimensional stability of dental materials can be influenced by several factors which include the conditions for setting, the solubility levels of the material and environmental condition [79]. The expansion of the material after setting may enhance the sealing ability of MTA [80, 81]. According to Specification No.57 of ANSI/ADA, the mean linear shrinkage of a cement shall not exceed 1% or 0.1% in expansion [81, 82]. It has been reported that HCSCs expand by 0.2-6% of the initial volume [64]. Water sorption induces some expansion. The Dimensional change of TheraCal LC in 24 hours is 1.31% and it at 30 days is 3.89% [64].

Conclusion

The data on the physicochemical properties of the presented novel formulations of resin modified calcium silicate cements in this article are limited and they are from manufacturers. Independent in-depth studies are needed for them in order for the dentist to select the most suitable, tailored to the individual clinical case.

The examined and critically analyzed data refer mainly to the most studied representative of the 4th

generation TheraCal LC.

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