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Jacobs Journal of Dentistry and Research

Research Article

Calcium aluminate based-cements for endodontic application

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Received: 08-05-2014

Accepted: 08-12-2014

Published: 09-22-2014

Abstract

Despite the good clinical performance of MTA cement, some of its negative features must be considered. These deficiencies justify the research and development of new materials with adequate physico-mechanical and biological properties, such as calcium aluminate-based cements. Calcium aluminate based-cements have a great potential in the dental area due to their biological compatibility, mechanical strength and good physico-chemical properties. The aim of this article was to perform a comprehensive literature review regarding the development, composition, properties and application of calcium aluminate-based cements in dental area, mainly in endodontic therapy. The use of this type of cement in dentistry has increased considerably in the last years, as the number of studies evaluating its properties.

Keywords: Mineral Trioxide Aggregate; Calcium Aluminate Cement, Physico-Mechanical Properties; Physico-Chemical; Bio-Activity

Introduction

Several clinical procedures, such as root and furca perforations sealing, and retrograde filling, need a specific cement to obtain a successful treatment [1,2]. At the early 1990's, many products, such as calcium hydroxide based-cements, glass ionomer cement and amalgam, were used for such purpose [3,4]. However, none of these materials could adequately address all the required properties of a sealing cement [5].

In the same period, researchers from the Loma Linda University, (California, USA), including Dr. Mahmoud Torabinejad, developed a cement called mineral trioxide aggregate (MTA), indicated for perforations treatment, root canal retrofilling, apexification and pulp capping, being its patent required in 1995; and beginning its trading under the name of ProRoot MTA (Dentsply Tulsa Dental, Tulsa, OK, USA) [6-8].

Such cement has promoted a great transformation in endodontic therapy, not only because of its excellent physico-chemical and biological properties in comparison with the other materials used so far, but for possessing composition very similar to ordinary Portland cement (Type 1), widely used in building area [8].

Portland cement is basically classified into two sub-types: structural and non-structural [9,10]. The non-structural form has lower amount of clinker, a raw material used in the manufacture of the cement, and gypsum, at a ratio of (74-50%) relative to the structural form (100-75%) [9,10]. The clinker is responsible for a significant increase in the mechanical strength of the cement, as well as becomes it less soluble [9,10].

MTA is basically composed (% by weight) of Portland cement (75.0), plus Bi_2O_3 (20.0) to confer radiopacity to the cement; and dehydrated $CaSO_4$ (5.0) [11]. Portland cement, in turn, comprises SiO_2 (21.2), CaO (68.1), Al_2O_3 (4.7), MgO (0.48) and Fe_2O_3 (1.89) [12]. Since it is a hydraulic cement, its setting reaction begins with the addition of water, forming hydrated silica gel [13,14].

MTA cement is commercialized in two different versions, grey and white (Grey and White MTA) [12]. The main difference between the two versions is the highest concentration of iron oxide in the Grey MTA, which, according to several studies, is the main responsible for dental tissues staining when the material is used [12]. According to Bortoluzzi et al. [15], the lowest concentration of iron oxide in the white version of the cement prevents staining and gingival discoloration due to the non-diffusion of the compound into dental tissues. However, several studies have shown that both versions of the cement promote dental tissues staining [16-18].

When compared to calcium hydroxide based-cement to accidental pulp exposure treatment, the MTA cement is able to maintain a greater tissue integrity [19], with a thick mineralized barrier formation three times faster [20-22], presence of small inflammatory infiltrate, and a thin layer of pulp tissue necrosis [19,23-25]. It is believed that the mechanism of action of MTA cement is similar to the calcium hydroxide based-cements, promoting inflammation and necrosis of the pulp tissue adjacent to the area of exposure [26,27].

During MTA cement setting process, calcium disilicate and trisilicate react with calcium hydroxide to form calcium silicate hydrated gel, producing a highly alkaline pH [28]. The continuous process of cement hydration leads to calcium ions release, which are diffused through the dentinal tubules, ensuring the proper reparative cement capacity [29]. Among the other advantages of MTA in comparison with other cements used for the same purpose, its lower solubility, higher mechanical strength, better marginal adaptation and better sealing ability are outstanding [30].

Despite its excellent clinical applicability, some negative features of MTA cement should be taken into consideration, as its poor handling characteristics [31]; high cost [32]; high rates of solubility in moist conditions [5,33,34]; presence and release of arsenic [35-37] above safe limits proposed by the ISO 9917-1 standard [38], and long setting-time [6,7,39,40]. This fact justifies the development of new materials incorporating the appropriate bioactivity of MTA, however, without its undesirable properties [5].

Calcium aluminate-based cements

Calcium aluminates are the main components of calcium aluminate cements. Since its development in the early 1900s, calcium aluminate-based cement has been the subject of several studies, and due to its excellent properties has been used for different purposes [41-43]. Its main application is still in building, mainly in the manufacture of concrete as Portland cement, however, for extreme environments where greater resistance to heat and abrasion is required; and a faster setting reaction [41-43]. However, in the last decade, its application has been extended to other areas, such as Dentistry, as dental cements [41-43]. Just as the calcium silicate based-cements, calcium aluminate based-cements are considered hydraulic cements, since its setting reaction starts from the mixture of the powder with water [43]. An aluminate cement is basically composed of Al_2O_3 (43%); CaO (19%); H_2O (15%); ZrO_2 (19%), besides other components, such as magnesium, silicon, iron, titanium and alkali oxides, in proportions lower than 10% [43].Generally, such cements are composed of three main phases responsible for the hydraulic setting process: anhydrous phase CA ($CaO.Al_2O_3$), comprising from 40 to 70% of the product; phase CA_2 ($CaO.2Al_2O_3$), which is the second in proportion (>25%), and phase $C_{12}A_7$ ($12CaO.7Al_2O_3$), with approximately 10% [42,44].The hydration reaction of calcium aluminate based-cements has three distinct phases: ions dissolution, nucleation and precipitation of hydrated phases [42]. When the cement particles come in contact with water, anhydrous phases of calcium aluminate are dissociated, releasing calcium and hydroxyl ions into the aqueous medium [44]. The dissolution process continues until the concentration of Ca and hydroxyl ions is saturated, initiating their precipitation in the form of calcium aluminate hydrates, by a mechanism known as nucleation and growth [42]. Such precipitation decreases the Ca and hydroxyl ions levels below the saturation point, promoting the anhydrous phase formation. The process continues until most, or all of the anhydrous phase had been reacted [42,43].

In general, the chemical reaction responsible for the calcium aluminate based-cements formation can be described as follows:

$$CaCO_3 + Al_2O_3 = Ca(AlO_2)^2 + CO_2$$

Currently some calcium aluminate based-materials for dental application are available on the market: Ceramir Crown & Bridge (Doxa Dental AB, Uppsala, Sweden) a hybrid luting cement, composed from the mixture of a calcium aluminate based-cement and glass ionomer [45], Doxadent (Doxa Dental AB), a direct restorative cement [46], and a cement similar to MTA's clinical applications, called EndoBinder (Binderware, São Carlos, SP, Brazil) [47].

EndoBinder basically consists of (% by weight) Al_2O_3 (≥68.0), CaO (≤31.0), SiO_2 (0.3-0.8), MgO (0.4-0.5) and Fe_2O_3 (<0.3), and it is produced by the process of Al_2O_3 e $CaCO_3$ calcination, at high temperatures ranging from 1315 to 1425°C, a viable method to produce materials with a more uniform composition [47]. The calcium aluminate formed is cooled, and then ground up until obtaining a powder with proper particle size [47]. At the end of the sintering process, a radiopacifying agent is added to ensure adequate radiopacity to the cement [48], according to the ISO 6876-7.8 standard [49].

Physico-mechanical properties

The physico-mechanical properties of calcium aluminate based-cements have been described by several studies

[42,50]. Authors reported important mechanical properties, such as microhardness, compressive, flexural and diametral tensile strength, superior to MTA cement, making the material a reliable option for endodontic therapy [42,50].

Garcia et al. [50] performed a mechanical and microstructural characterization of a novel calcium aluminate based-cement for endodontic application, by compressive and diametral tensile strength tests, Vickers microhardness test and Scanning Electron Microscopy (SEM) analysis, in comparison to both versions of MTA cement (white and grey). The calcium aluminate based-cement presented higher compressive strength than white MTA after 7 and 21 days post-setting; and higher diametral tensile strength values than grey MTA (7 and 21 days), and white MTA at 21 days post-setting. As regards Vickers microhardness, it wasn't possible to obtain adequate values at 24 hours post-setting period, due to the low surface mechanical strength presented by the different cements in this initial period of evaluation. However, the calcium aluminate based-cement presented higher Vickers microhardness values than white MTA at 7 and 21 days, and grey MTA at 21 days post-setting.

In the same study [50], the SEM evaluation demonstrated several topographical accidents that formed the microstructure of the cements, such as pores and asymmetrical crystalline formation, channels and depressions. The calcium aluminate based-cement presented a crystallized structure, with aggregated particles of globular shape, homogeneous size and uniform distribution. Furthermore, the cement presented a more regular surface, with less pronounced depressions than both versions of MTA cement (Figures 1 and 2).

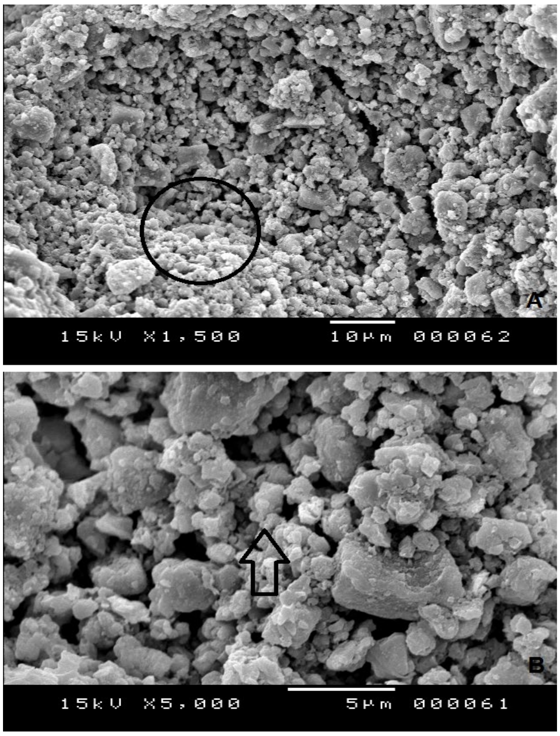

Figure 1. Representative micrographs of calcium aluminate based-cement samples 24 hours post-setting. (A) Note the cement topography, with a more regular surface than MTA cement (indication). (B) Globular-shaped aggregated particles with homogeneous size and uniform distribution (arrow).

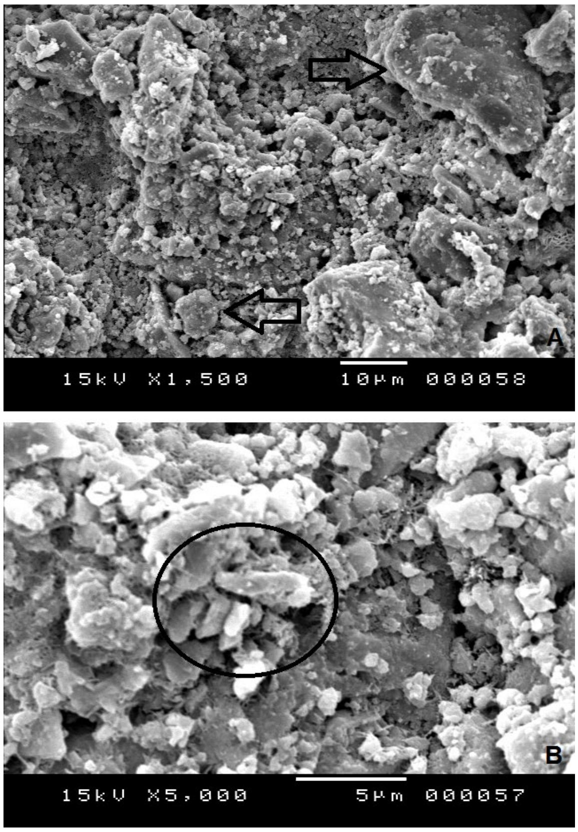

Figure 2. Representative micrographs of MTA cement samples 24 hours post-setting. (A) Note the microstructure of the cement, with irregular-shaped and asymmetric particles (arrow). (B) Aggregated particles which compose the crystalline phase of the cement, with asymmetric sizes and shapes (indication).

Oliveira et al. [42] reported in a similar study that a experimental calcium aluminate based-cement presented compressive strength values of 32.0 MPa at 24 hours post-setting, which increased significantly to 51.0 MPa after 15 days post-setting, versus 34.0 MPa of MTA. With the incorporation of a dispersant and a plasticizer to improve the handling carachteristics of the cement, the compressive strength values increased to 65.0 MPa after 15 days post-setting. Furthermore, the addition of ZnO as radiopacifying agent increased the compressive strength mean values to 81.0 MPa, whereas for MTA cement, the mean values remained to 34.0 MPa.

After MTA cement handling, bismuth oxide (radiopacifying agent) becomes part of the hydrated phase of the cement, forming a structure composed of hydrated bismuth calcium silicate, ettringite and monosulphate [51]. Such compounds are released into the medium with the calcium hydroxide formed from the calcium silicate hydration process, decreasing the precipitation of hydrated calcium hydroxide, thus, compromising the physico-mechanical properties of MTA [52,53].

Physico-chemical properties

Pires-de-Souza et al. [54], in a recente study, assessed the pH, calcium ion release and antimicrobial activity of a calcium aluminate based-cement, containing different radiopacifying agents: zinc oxide, bismuth oxide, or zirconium oxide, in comparison with MTA cement.

Samples of the tested cements were immersed into 10 ml of distilled and deionized water at 37C°, and after 2, 4, 12, 24, 48 hours; 7, 14 and 28 days, the pH was measured and calcium ion release quantified in atomic absorption spectrophotometer. MTA cement presented the highest pH values at 28 days, however, at 14 and 28 days, the calcium aluminate based-cement and MTA presented similar results, irrespective of the radiopacifying agent used.

The initial pH of MTA was higher than the calcium aluminate based-cement in its different forms. This fact could be explained by the presence of calcium chloride in MTA cement composition, used to accelerate its setting-time [55]. Several studies reported that calcium chloride increases the pH after handling, due to the calcium incorporated into the cement's formula [55-57].

Regarding to calcium ions release, when the different cements were compared in the same period of analysis, no significant difference among them was observed. The hydration process of calcium aluminate based-cements results in calcium aluminate and aluminum hydroxide hydrates [42,44], thus, calcium ions release could be attributed to the decomposition of calcium aluminate hydrate [42,44].

The antimicrobial activity of the different cements was tested against E. faecalis, S. aureus, E. coli, and C. albicans. MTA cement presented inhibition haloes similar to calcium aluminate based-cement with ZnO as radiopacifying agent for S. aureus. However, the calcium aluminate based-cement presented no antimicrobial activity against E. coli and E. faecalis with ZnO and ZrO. The other tested cements presented activity against these microorganisms, with no significant difference among them. The same was observed against C. albicans. The calcium aluminate based-cement containing ZrO and ZnO as radiopacifying agents presented no antimicrobial activity.

Garcia et al. [58] evaluated the solubility and disintegration of a novel calcium aluminate based-cement, containing the same different radiopacifying agents tested in the study cited above, in comparison with white and grey MTA. The authors reported that all tested cements presented solubility and disintegration values above the ANSI/ADA Specification No. 57 (> 3.0 %) [59]. The calcium aluminate based-cement with bismuth oxide presented the lowest mass loss, and white MTA the highest values. However, no significant difference was observed among the cements.

Several studies reported that high solubilization rates do not contraindicate the use of MTA cement [33,60]. The adequate biological performance of the cement is related to the ability to release calcium and hydroxyl ions into the medium during its continuous hydration process [61]. In addition, calcium ions are the main components detected in residues released by MTA cement, confirming that solubility is an important phenomenon in the delivery of calcium and hydroxyl ions to pulp tissue [33,62].

In the same study, Garcia et al. [58] evaluated the components released by the cements during the solubility and disintegration test. Several chemical elements were detected in the resulting solutions. Only the calcium aluminate based-cement with bismuth oxide and grey MTA accused the presence of Chromium. Calcium aluminate based-cement with zinc oxide presented the highest levels of Lead, followed by white MTA. For Arsenic, the tested cements presented different release rates. Calcium aluminate based-cement with zinc oxide presented the highest levels, and grey MTA the lowest levels. However, the amount of Arsenic and Lead released were lower than the safe limit proposed by ISO 9917-1 standard [38].

Radiopacity is an important physical property of cements used in endodontic practice. The ISO 6876-7.8 standard [49] states that endodontic cements should present a radiopacity above 3.0 mm in relation to the aluminum scale. Therefore, an adequate radiopacifying agent should be added to cement's composition to allow root canal filling evaluation, and to differentiate the material from the adjacent anatomic structures, such as dentine, bone and enamel [32].

Aguilar et al. [48] evaluated the radiopacity of a novel calcium aluminate based-cement containing three different radiopacifying agents (bismuth oxide, zinc oxide or zirconium oxide), and both versions of MTA (white and grey), in samples of different thicknesses.

The results demonstrated that the thicker the cement sample, the greater its radiopacity. The calcium aluminate based-cement with bismuth oxide; and grey MTA, presented radiopacity above 3.0 mm of the aluminum scale, irrespective of the sample thickness. When zinc oxide was used as radiopacifying agent, calcium aluminate based-cement only reached the required radiopacity with a sample 2.0 mm-thick, and with zirconium oxide, 2.5 mm-thick.

Only the calcium aluminate based-cement with bismuth oxide and grey MTA presented adequate radiopacity for all the samples thicknesses tested. The radiopacifying agent used in both cements was bismuth oxide (20% by weight), which has a higher atomic number (Z=83) in comparison with zirconium (Z=40) and zinc (Z=30), corroborating the results of Camilleri [32].

Biological properties

One of the most important properties of MTA is the reparative capacity of the cement [63-65]. MTA is able to promote reparative hard tissue formation on exposed pulp tissue faster than other cements used for such purpose [66, 67]. Several studies demonstrated that in humans, the pulp tissue repair induced by MTA is more effective than that promoted by calcium hydroxide based-cement, which causes inflammation and necrosis of the area adjacent to the exposure area [24,68].

The biological compatibility of MTA depends on its ability of releasing calcium ions, and, to the alkaline pH produced by the cement [21]. In a moist medium, MTA releases calcium oxide and calcium silicate [51]. Next, the calcium oxide released produces calcium hydroxide after get in contact with the tissue fluids, releasing the calcium ions [51].

Aguilar et al. [69] evaluated the biocompatibility of a calcium aluminate based-cement (EndoBinder) in subcutaneous tissue of rats, in comparison with the grey version of MTA. After 42 days, EndoBinder presented no inflammatory reaction, however, a mild inflammatory reaction was observed for MTA, in the same period of analysis, which denotes the presence of a chronic inflammatory process.

When materials for dental application enter in contact with living tissues, they become irritants; however, the duration of this effect on living tissues is more important than the irritant potential of such materials [70]. In a similar study, Yaltirik et al. [71], reported the same chronic inflammatory process in MTA-specimens 60 days after subcutaneous implantation of the cement. This fact could be explained by the excess of calcium ions released by MTA cement in certain circumstances, becoming irritant to living tissues, promoting a persistent inflammatory process [13].

Oliveira et al. [72] assessed the bioactivity of an experimental calcium aluminate based-cement containing accelerating additives to improve handling carachteristics and increase mechanical strength, in comparison with MTA cement. The surface of the cements was kept in contact with simulated body fluid solutions, and was analyzed by SEM, X-ray diffraction and energy-dispersive X-ray.

The authors observed that MTA cement release more ions calcium, promoting a higher pH, in comparison with the experimental calcium aluminate based-cement [72]. Such fact resulted in calcium phosphate phase's precipitation on the MTA cement surface. It was conclude that both tested cements presented bioactivity in contact with the simulated body fluid solutions, however, the solution formulation was the main responsible to define the type of phase precipitated.

Silva et al. [73] investigated the effect of cements used as of root repair sealers (calcium hydroxide, EndoBinder-calcium aluminate based-cement-and MTA) on the cytotoxicity and gelatinolytic activity of matrix metalloproteinases in 3T3 type-fibroblasts. It was observed that fibroblasts secreted MMP-2 after 24 hours in contact with calcium hydroxide, inducing MMP-2 expression in comparison with the other sealing cements. On the other hand, there was no significant difference in the cell viability results for MTA and EndoBinder. However, calcium hydroxide significantly reduced cell viability of 3T3 fibroblasts.

As regards to osteogenic potential, Castro-Raucci et al. [74] evaluated the progression of osteogenic cell cultures exposed to a novel calcium aluminate based-cement in comparison with MTA cement. After 10 days of culture, both tested cements presented osteogenic cell proliferation and adhesion, however, the cell cultures exposed to the calcium aluminate based-cement presented higher values of total cell number, total protein content and alkaline phosphatase activity. These results indicated that the calcium aluminate based-cement supported a higher differentiation of osteogenic cells than the MTA cement.

Conclusions

Despite its wide use in many areas, the application of calcium aluminate based-cements in dental practice is still recent. However, the good results obtained in different studies qualify this new material as a viable option for use in several clinical cases.

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