

IMMEDIATE AND DELAYED SOLUBILITY OF MINERAL TRIOXIDE AGGREGATE AND PORTLAND CEMENT

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ABSTRACT

This study investigated the solubility of mineral trioxide aggregate (MTA) and Portland cement since its mixture until 672 hours, by means of two complimentary methods. Metal ring molds filled with the cements were covered with distilled water and, at each experimental time (3, 24, 72, 168, 336 and 672 hours), were weighed as soon as the plates in which the samples have been placed. Empty rings served as the control group (n=8). Mean weight gain and loss was determined and analyzed statistically by two-way ANOVA and Tukey's test for all pairwise comparisons. Only Portland cement showed less than 3% weight loss through 24 hours. Detached MTA residues were heavier than those of Portland cement over the 3 to 168 hours. The weight of MTA rings increased more than that of Portland rings within 672 hours (p=0.05). The findings of the present study indicate that, in an aqueous environment MTA is more soluble than Portland cement and exceeds the maximum weight loss considered acceptable by ISO 6876 standard (2001).

Key words: Solubility. MTA. Portland. Physical properties.

INTRODUCTION

Mineral trioxide aggregate (MTA) consists of a powder that, when added of water, forms a grainy and slurry mixture able to harden even in presence of complete humidity^{5,10,26,27,29}. The biocompatibility^{16,17}, dentinogenic activity^{5,11,15} and sealing ability^{2,28,30} attributed to MTA are believed to depend entirely on the physicochemical interactions of the cement with the environment²².

After setting MTA hydration produces Portlandite, which is essentially considered crystalline calcium hydroxide^{5,9,24,25}. Alkaline pH levels and calcium ions detected in the water surrounding MTA^{4,8,22} are considered essential to hard-tissue deposition^{22,26} and probably related to its dissolution in moist environment such as hard and soft tissues^{9,22}.

Recently, a white version of MTA, chemically similar to the grey version was introduced to the market^{18,4,24,25}. The

manufacturer claims that, differently from gray MTA, this form does not offer risks of tooth staining¹⁶ and gingival discoloration³ caused by iron diffusion to tissues and can applied as pulp capping material or perforation sealant in anterior teeth.

MTA has a prolonged setting time and is considered unstable when irrigated before complete hardening^{1,2,4,26}. Until now, all studies on the solubility of mineral trioxide aggregate considered it less soluble than Portland cement and under the 3% maximum weight loss determined as acceptable by International Standard Organization¹² after storage in distilled water for 24 hours^{9,13,6,21,26}. However, these investigations have been conducted based on the decrease in the mass of hardened and dried specimens^{6,9,13,21,26}, a circumstance completely different from that observed in oral tissues, where MTA is routinely used.

Although there may be a difference in particle uniformity, gypsum content and presence of radiopaque substance,

the overall composition of MTA is similar to that of Portland cement^{1,24,25}. Therefore, this study tested the hypothesis that the solubility of MTA when completely covered by water since its mixture is presumed to follow a pattern similar to that of Portland over 672 hours.

MATERIAL AND METHODS

The weight gain and loss of two commercially available materials, grey MTA[®] (Angelus, Londrina, PR, Brazil) and grey Portland cement (Votoran[®], Votorantin, São Paulo, SP, Brazil) supplemented with 20% bismuth oxide, was determined by the decline or increase in mass of specimens by storage in water. The weight of particles detached from the cements during the experiment was also recorded as recommended by the ISO 6876 standard¹² (2001).

Preparation of Ring Samples

Petri dishes and metal ring molds fixed to copper wires, having an internal diameter (i.d.) 20.0 ± 0.1 mm and a height of 1.6 ± 0.1 mm, were cleaned in an ultrasound bath of detergent for 15 minutes before being weighed on a precision scale (accuracy: 0.0001 g) alone or in sets consisting of one metal ring and one plate properly identified by codes. All weight measurements were made in grams and calculated to four decimal places.

The experimental cements were divided into portions of 1 g and mixed with 0.33 mL distilled water at a temperature of 25°C and relative humidity of $65 \pm 5\%$ according to Friland and Rosado⁹ (2003). The mixture was homogenized for 30 seconds and each mould, positioned inside a glass plate, was filled with the experimental cements to slight excess using a spatula ($n=8$).

A single operator performed all manipulations and care was taken to prevent entrapment of air bubbles or leakage of the materials from the rings.

Sample Weight

The plate with the filled ring was weighed and 60 mL distilled water, pH 7.1 (37°C), was then spread with a Luer-Lock syringe over the plate distant from the ring so that the lower and upper surfaces of the cements were completely covered with fluid (100% relative humidity). The sets were closed and placed in an oven at 37°C from which they were removed at the indicated intervals.

The control group consisted of empty rings. Eight molds were used for each experimental group and the test specimens were studied for up to 28 days. After 3, 24, 72, 168, 336 and 672 hours of immersion, the rings were hanged over the Petri dish and gently irrigated on both sides to collect the residues released into the container. After superficial drying with absorbent paper, the rings were weighed and immediately placed on new previously weighed dishes filled with 60 mL bidistilled water.

The discarded plates were kept in the oven at 65°C for 24 hours for content dehydration and the mass of the residues released by the test specimens was determined. This process

was repeated for each experimental period. The initial amount of cement inside the molds was determined by subtracting the weight of the ring-Petri dish from the weight of the filled ring-Petri dish. The weight gain or loss was recorded as the difference between the initial weight of rings and dishes and the final weight in each experimental time. The difference in mass was calculated as a percentage of the original weight of the cements. The weight gain and loss of the cements were analyzed by two-way analysis of variance and the comparison of the two methods as a function of time was performed by three-way ANOVA statistical test, both set at significance level of 5%.

RESULTS

Ring Weight Method

There was no statistically significant change in the weight of the empty and MTA filled moulds after immersion in the distilled water through the experimental intervals. Portland specimens showed the heaviest weight in the 3rd hour and the lightest in 672 hours period ($p=0.05$).

During a setting time of 3 hours, Portland cement presented a weight gain of 0.98%, statistically superior to MTA (0.32%) and to the control group. After 24 hours, there was a continuous weight gain of MTA rings statistically superior to Portland cement and empty rings in 168 hour time. In the 336th hour, Portland cement lost more weight (0.24%) than MTA (-0.08%). This difference, however, was not observed in the intervals to follow. Figure 1 shows the variations of weight gain and loss (%) detected upon weighing of the rings during each experimental intervals.

Residues Collection Weight Method

During the 3-hour collection, both Portland (5.12%) and MTA (4.23%) plates were heavier than the control plates (0.057%). A statistically significant decrease of Portland weight loss was detected from 3 to 24 hours, moment at which Portland group weight (1.46%) was lower than that observed in MTA group (3.81%) and higher than control (0.053%), a behavior that was also observed within the 72-hour period (Portland: 1.67%; MTA: 2.44%; Control: 0.067%). From the 168th hour on, the weight of residues detached from both cements was statistically equivalent and superior to that of the control group (Figure 2).

The residues collected on MTA plates in the first 24 hours were heavier than those collected in the 336- and 672-hour periods, the latter being characterized by a weight loss of approximately 1% for both cements. There was no significant change in the weight of plates containing the empty ring moulds over the experimental intervals.

Complementary Methods of Analysis

Comparing the data of ring and dish weights of both cements as a function of cement type and time, statistically significant differences were detected ($p=0.05$) (Table 1).

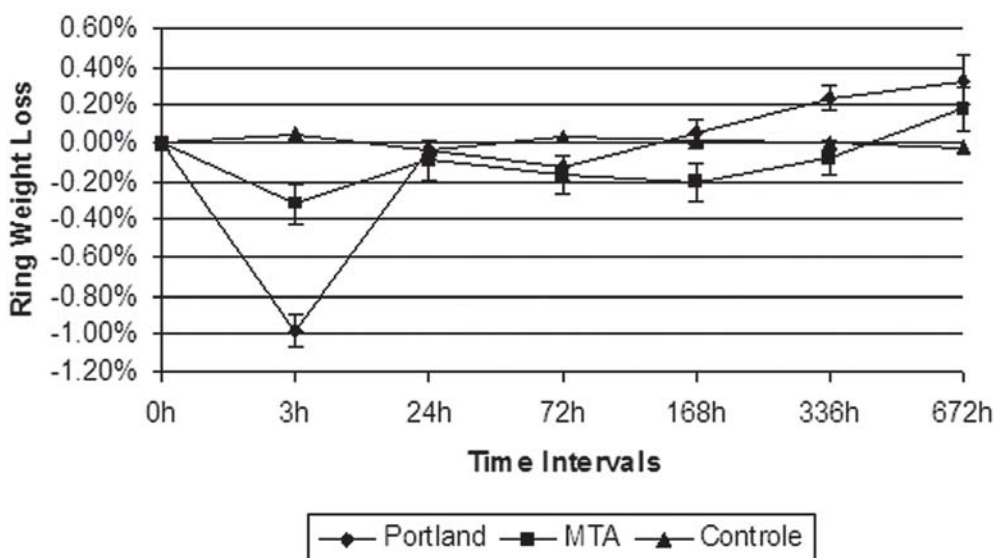


FIGURE 1- Mean weight loss ± standard error of mean of the cements during the experimental intervals determined by the ring weight method

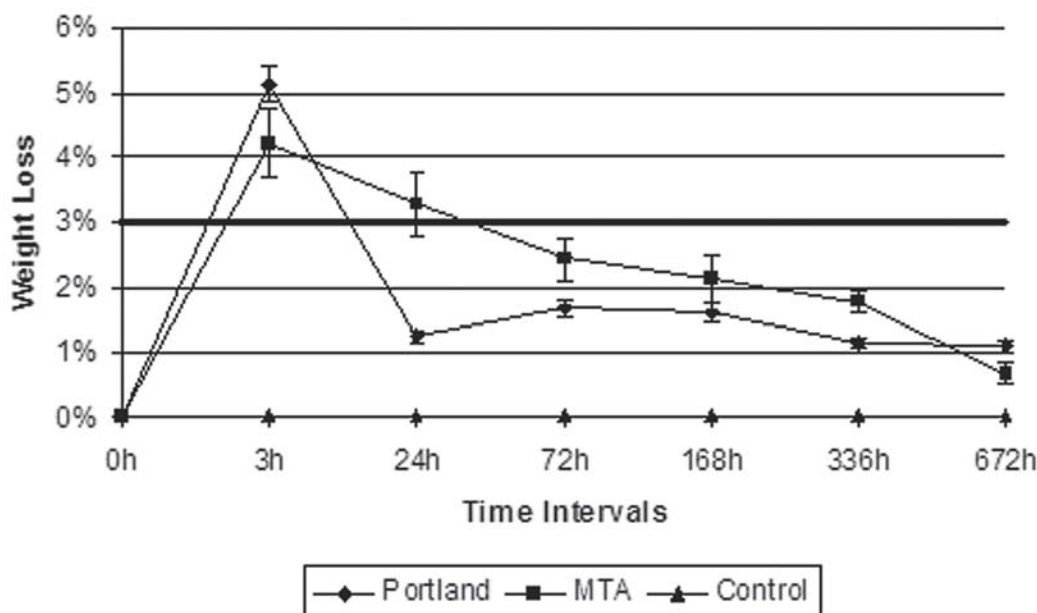


FIGURE 2- Mean weight loss ± standard error of mean of the cements during the experimental intervals determined by the residue collection method. The bold horizontal line represents the requirement for compliance with the ISO 6876 standard (2001)

TABLE 1- Three-way ANOVA for the comparison between methods, cements and time (M – methods; C – cements; Ti – Time; * statistically significant)

Effect	df Effect	MS Effect	df Error	MS Error	F	p value*
M	1	0.021240	30	0.000223	95.16363	0.000000*
C	2	0.001987	30	0.000233	8.90274	0.000922*
Ti	5	0.000071	150	0.000207	0.343510	0.885765
M-C	2	0.004054	30	0.000223	18.16431	0.000007*
M-Ti	5	0.002991	150	0.000207	14.42377	0.000000*
C-Ti	10	0.000342	150	0.000207	1.648060	0.098290
M-C-Ti	10	0.001807	150	0.000207	8.71690	0.000000*

DISCUSSION

It is well known that the humidity close to MTA determines the level at which its physical properties, such as sealing ability, push-out and flexural strength, are exerted^{10,29,30}. Differently from previous solubility studies, in the present investigation, the cements were kept immersed since the moment of their mixture. The residue mass separated from MTA-Angelus was significantly higher than that of Portland cement and above the 3% maximum weight lost considered acceptable in the first 24 hours by the International Standard Organization¹².

Ideally, a cement should block bacterial migration to pulpal and periodontal tissues to help controlling infection and preventing its recurrence. Solubility is a factor directly related to the sealing potential as well as dimensional changes. According to our results, the sealing ability of MTA in an aqueous environment might be compromised during the first 72 hours because of material's excessive solubility that may affect particle arrangement in dentinal cavity walls. Sluyk, et al.²³ (1998) suggested that 3 days is the time needed for MTA to exert its good sealing ability when used as a perforation repair material.

Differently from MTA, the solubility of Portland cement decreased to 1.46% between 3 and 24 hours after collection (Figure 2). Interestingly, *in vitro* short-term studies have not detected differences in bacterial or dye microleakage when both materials were compared as root-end fillings or perforation repair material^{2,7}. Nevertheless, the particular state of humidity in contact with specimens before and during the experimentations should be taken into account.

Clinically, the recommended provision of a wet cotton over MTA to assure proper setting and to accomplish best performance in the shortest period of time^{10,19,23,27,29} may favor the detachment of particles into pulpal and periodontal tissues and elicit inflammatory reaction. Holland, et al.¹¹ (2001) noticed small particles of Portland cement in macrophage cytoplasm 60 days after pulpotomy and pulp capping in dog's teeth.

Additionally, when the dentist accidentally pushes freshly MTA out of root limits, the tissue humidity promotes a partial dissolution of the material that, according to our results, extends at least 28 days. The released particles into adjacent structures probably elicit an undesirable chronic inflammation process¹¹ and are also, perhaps, the cause of the dark discoloration of marginal gingiva described by Bortoluzzi, et al.³ (2007) when a root perforation was sealed with gray MTA.

The overall solubility of MTA or Portland cement by itself does not contraindicate its usage as root canal sealer or orthograde apical plug in cases where intracanal exudate would impair the setting process of conventional sealers. The behavior detected with MTA, specifically the excessive initial solubility followed by a constant decrease over 672 hours, have already been described to routinely used root canal sealers AH 26 and Tubli Seal¹⁴. Nevertheless, some adjustments to turn MTA easy to handle and be inserted adequately in the root canal are still necessary^{1,9,28}.

The biological performance of MTA is attributed to its Portland content that throughout hydration is capable of releasing calcium and hydroxyl ions²⁵. It has been reported that these ions are the main component detected on soluble and insoluble residues detached from MTA^{4,8,9,22,24}. As ordinary Portland releases those ions in significantly higher quantities in the first 24 hours^{1,4,5} and considering that in this study the detachment of particles was more pronounced for MTA, it could be inferred that solubility is not the only phenomenon related to calcium and hydroxyl ions delivery in periodontal or pulp tissues.

Despite the hydration inherent pore formation, it is likely that the aqueous particle detachment also left empty spaces inside the cement matrix, which were ready to be filled by surrounding liquid. It is known that water uptake may compensate for dissolved material²⁰ during weighing. In this way, the marked 3-hour increase in ring weight, more intense for Portland cement, may be an effect of initial water entrapment in the matrix structure. Liquid sorption may influence the interaction with an immediate covering material as seen during pulp capping. In a pilot study, the glass-ionomer applied over partially set MTA dissolved it, becoming thoroughly mixed with the material¹⁹.

After few hours, the hydration process promotes an increase in volume of solids and thus decreases the porosity of the paste⁵. Theoretically, a reduction of ring weight should be expected, a phenomenon observed at the 72nd and 168th hour, respectively, with Portland and MTA, extending until the end of the experiment (Figure 1).

In the development of Portland-derived cements, it should be considered that differences in particle uniformity and reduction of gypsum content cause a delayed reduction of cement solubility, described in this study, and also lengthening of setting as well as structure weakening, as reported elsewhere^{1,5,13}.

Before suggesting MTA as a restorative material^{1,2,10,29}, studies addressing the reduction of initial solubility throughout addition of setting accelerants should be conducted to establish a possible correlation and assure invulnerability to bacterial microleakage. With regard to solubility, until its importance on cement's biocompatibility and long-term sealing property is not clarified, MTA should not be replaced by Portland cement in treatments where excessive moisture is present.

CONCLUSION

Based on the results of the present study, the null hypothesis should be rejected. MTA-Angelus was more soluble than Portland cement when completely immersed in an aqueous medium throughout 672 hours.

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