

Influence of bentonite addition on the handling and physical properties of tricalcium silicate cement

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Purpose: In this study, the influence of bentonite addition on the handling and physical properties of tricalcium silicate is comprehensively investigated.

Materials and Methods: Bentonite and tricalcium silicate powder were used in this study and 0, 10, 20, 30 wt% of bentonite added tricalcium silicate cements were prepared. Microstructural and morphological changes in tricalcium silicate cements are evaluated with X-ray diffraction and scanning electron microscopy. To evaluate the handling properties of the cement paste, flowability and injectability, as representative handling properties, are evaluated. The compressive strength and setting time are measured using the ISO-9917 methods.

Results: Our analysis shows that as the amount of added bentonite increases, both flowability and injectability improve significantly. Further, no significant difference is observed in the compressive strength and setting time after the bentonite addition.

Conclusion: Bentonite addition improves the handling properties of the tricalcium silicate paste without causing a significant deterioration in its physical properties. These results provide important information for the development of novel tricalcium silicate-based cements with enhanced properties.

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Key Words: bentonite, handling property, physical property, tricalcium silicate

Introduction

Mineral trioxide aggregate (MTA) was originally introduced as an endodontic material for perforation repair and root-end filling in 1993 [1,2]. Since its introduction, it has also been applied to direct pulp capping and regenerative endodontics [3-5]. MTA consists of Portland cement, bismuth oxide as a radiopacifier, and gypsum dihydrate. The main components of Portland cement are tricalcium silicate (TCS) and dicalcium silicate, which are set via the hydration reaction. MTA shows good sealing ability [6], biocompatibility [7], antibacterial activity [8], and hard tissue formation induction [9,10]. The Portland cement, which is the major component of MTA contains not only calcium silicate, calcium aluminate, and tetracalcium aluminoferrite but also several impurities such as ferric oxide, arsenic, and heavy metal elements. The contents of arsenic and other heavy elements are low, but their toxicity is of concern [11,12]. The first commercial MTA product was gray, and white MTA was developed only later to improve the esthetic appearance. White MTA was obtained by purification, which reduced the content of iron oxide, which is the main cause of the gray color, and other heavy elements (cadmium, chromium, copper, and zinc). However, the contents of arsenic, nickel, and manganese became comparatively higher than those in the gray MTA [13]. In terms of clinical properties, significantly less leakage was observed for gray MTA when used as an apical barrier than for white MTA [14]. Therefore, a filling material that would possess simultaneously improved barrier properties and low impurity content is required.

TCS is the main component of Portland cement and exhibits the self-setting property upon hydration. TCS has been applied for preparing bone cement [15], dental restorative material [16], and root filling material [17,18]. TCS can be manufactured using a conventional calcination process from a mixture of calcium carbonate and silica. Recently, TCS preparation via the sol-gel process was also reported [19]. Therefore, synthetic TCS could be a viable alternative to MTA material without impurities. However, its long setting time [20] and poor

handling properties [21] have been of concern and several efforts have been conducted to improve these properties. For example, Ber et al. [22] reported that improvement in its handling properties and setting time can be accomplished via the addition of methylcellulose and CaCl_2 . However, TCS-based cement may also suffer from similar limitations in properties, as exemplified by the fact that the final setting time of a TCS cement has been measured as 180 min [23]. An improvement in the setting time with nanosilica addition was reported for MTA and calcium silicate cement without any adverse effect on the mechanical strength [24,25]. Primus et al. [21] reported an improvement in the handling properties of the experimental TCS-based cement without a decrease in its mechanical strength. However, current MTAs and TCS-based cements are less flowable than other dental cements for fillings [21]. Therefore, the carriage of the cement to deep and narrow lesions is not easy, and an improvement in the handling properties is required.

Clay is known to be a useful additive that can improve the rheological properties of industrial materials. Bentonite (Bt), which is a type of a smectite clay, is widely used as a rheology modifier for print inks, cosmetics, and industrial cement [26,27]. In particular, Bt is an inorganic rheology modifier for water-based systems [26], and thus, the application of Bt to the TCS-based cement (also an inorganic and water-based system) should not inhibit TCS hydration or setting behavior as much as organic rheology modifiers such as glycerol [28]. Bt is a typical clay mineral consisting of montmorillonite, which is a hydrous aluminum silicate with small amounts of alkali and alkali-earth metals [26]. The unit structure of montmorillonite consists of two silica tetrahedral sheets and an alumina octahedral sheet [26]. Water is absorbed into the inter-layer space and increases the c-axis dimension of the clay. The absorbed water acts as a lubricant for the layers and the plasticity of the clay increases with the increasing amount of water absorbed [26]. Taken together, we draw a hypothesis that Bt addition into TCS-based cements improves their rheological and handling properties.

The main objective of this study is to investigate the influence of bentonite addition on the handling (i.e., flowability and injectability, as representative handling properties) and mechanical properties of synthesized TCS-based cement.

Materials and Methods

Materials

Synthetic TCS (Nippon Shika Yakuhin Co., Ltd., Shimonoseki, Japan) was used as the base material for the experimental cement. The crystal phase and the particle shape of the synthesized TCS were estimated by X-ray diffraction (XRD, Miniflex, Rigaku Corp., Tokyo, Japan) and scanning electron microscopy (SEM, S-4500, Hitachi High-Technologies Corp., Tokyo, Japan). Bt (Kaneatsu-Yuyaku, Toki, Japan) was used as the consistency control agent. The experimental cements were prepared by replacing 0, 10, 20, and 30 wt% of TCS with Bt; the corresponding cement samples are denoted as 0Bt, 10Bt, 20Bt, and 30Bt, respectively.

Relative flowability

The relative flowability was evaluated on the basis of the cement paste spreading area under compression according to a previously reported method with some modifications [29]. Each experimental cement specimen was mixed with distilled water (DW) in a water/powder ratio of 0.6 by weight. The mixture (6 mm in diameter, 1 mm in thickness) was placed at the center of a glass slide using a stainless steel mold. The amount of the cement placed was accurately weighed using a precision balance (AG245, Mettler Toledo, Greifensee, Switzerland). Subsequently, another slide glass was placed on the material and 1 kg of weight was applied on the glass slide for

60 s at room temperature (24°C). The load was applied within 2 min after mixing. After the load application, the area of the spread mixture was estimated using the flat bed scanner (600 dpi in resolution, ApeosPort-IV C3375, Fuji Xerox, Tokyo, Japan) and the image analysis software (ImageJ, Ver.1.49u, National Institute of Health, Bethesda, MD, USA). The relative flowability of the mixture was estimated as the spread area normalized relative to the weight of the mixture as follows:

$$\text{Relative Flowability} = \frac{\text{area (mm}^2\text{)}}{\text{weight of the paste (g)}}$$

Relative injectability

The relative injectability of the experimental cement mixture was estimated using a modification of the syringe push-out test described in reference [30]. Each experimental cement specimen was mixed with DW in a water/powder ratio by weight of 0.5. Approximately, 0.28 g of the mixture was filled into a 1-mL disposable syringe (SS-01T, Terumo, Tokyo, Japan). The syringe was placed in a universal testing machine (EZ LX, Shimadzu, Kyoto, Japan), and the syringe plunger was pushed with a test speed of 15 mm/min to push out the mixture from the syringe head at room temperature (24°C). The push out test was carried out until the applied force reached 50 N. The relative injectability was estimated as the percentage of the weight of the mixture ejected from the syringe tip relative to the total weight of filled mixture.

$$\text{Injectability (\%)} = \frac{w_1}{w_0} \times 100$$

where w_0 is the total weight of the filled mixture and w_1 is the weight of the mixture ejected from the syringe tip.

Compressive strength

The compressive strength was determined according to ISO 9917-1; 2007. Each experimental cement specimen was mixed with DW in a water/powder ratio by weight of 0.5. The mixture was added to a separable cylindrical stainless steel mold (4 mm in diameter and 6 mm in height). After filling the mixture, both sides of the mold were covered by two glass plates, and kept at 37°C for 1 day under 100% relative humidity (RH) until setting. The specimen was removed from the mold after setting. A part of each specimen was immersed in DW at 37°C for 28 days. Those specimens were applied for the compression test using the universal testing machine (EZ LX, Shimadzu), with a test speed of 0.75 mm/min.

Setting Time

The setting time was determined according to ISO 9917-1; 2007. Each experimental cement specimen was mixed with DW in a water/powder ratio by weight of 0.5. The mixture was added into a stainless steel mold (10 mm × 8 mm in cross section, 5 mm in depth) and kept at 37°C at 95% RH. The setting time was estimated using the modified Vicat needle penetration test (indenter mass of 400 g, needle tip has a flat circular end with 1 mm of diameter). The indenter was carefully brought into contact vertically with the mixture surface with loading. This measurement was repeated every 60 s until setting was complete. The setting time was determined as the period from the end of mixing until the time that no clear indentation mark was observed on the mixture surface after loading.

Statistical analysis

Statistical analysis was performed by 1-way analysis of variance (ANOVA) and Tukey-Kramer test was used to perform multiple comparison tests with a significance level of 0.05.

Results

Microstructure of TCS powder and set cement, and results of XRD analysis

Figure 1 shows the SEM images of TCS powder and commercial MTA (ProRoot MTA, Dentsply, Johnson City, TN, USA) powder. Particles of the TCS powder are 6-40 μm in diameter and generally spherical, as shown in Figs. 1(A) and 1(B). The MTA particles are 10-60 μm in diameter and cuboid in shape. In addition, the radiopacifier particles of MTA appear white, as shown in Figs. 1(C) and 1(D). Figure 2 shows the hardened specimens of TCS and MTA after curing for 7 days. The TCS specimen exhibits aggregated spherical particles, while the MTA specimen exhibits granular appearance together with scattered radiopacifier particles. As shown in Fig. 3, as per XRD analysis results, TCS was confirmed to be mainly composed of the tricalcium silicate (Ca_3SiO_5) phase.

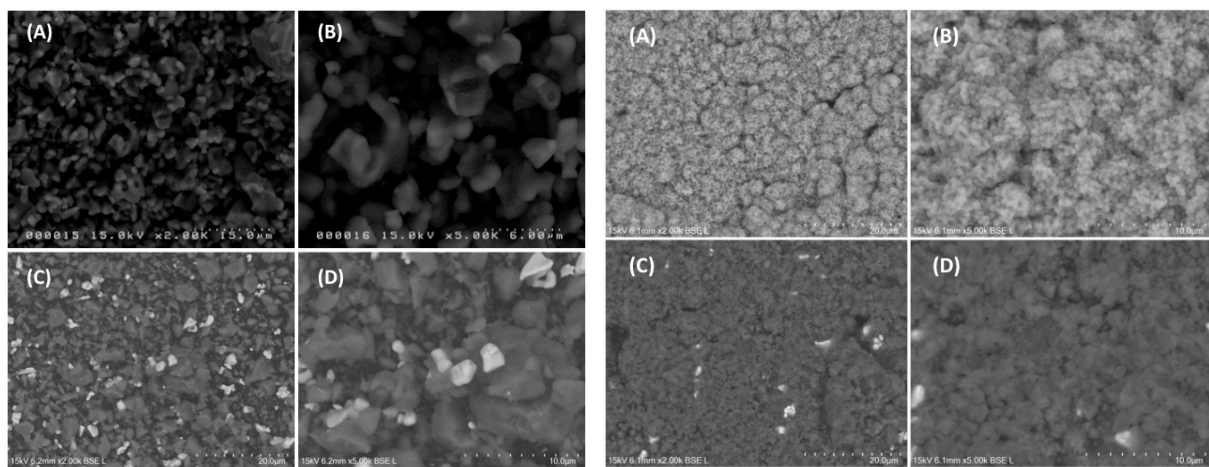


Fig. 1 (left) SEM images of (A,B) TCS and (C,D) MTA powder (ProRoot MTA). TCS particles are 6-40 μm in diameter and generally exhibit a spherical shape. MTA particles are 10-60 μm in diameter and exhibit a square shape. The radiopacifier of MTA appears as white particles in SEM images.

Fig. 2 (right) SEM images of (A,B) hardened specimen of TCS after curing for 7 days and (C,D) hardened specimen of MTA (ProRoot MTA) after curing for 7 days. Clusters of particles are visible in each image.

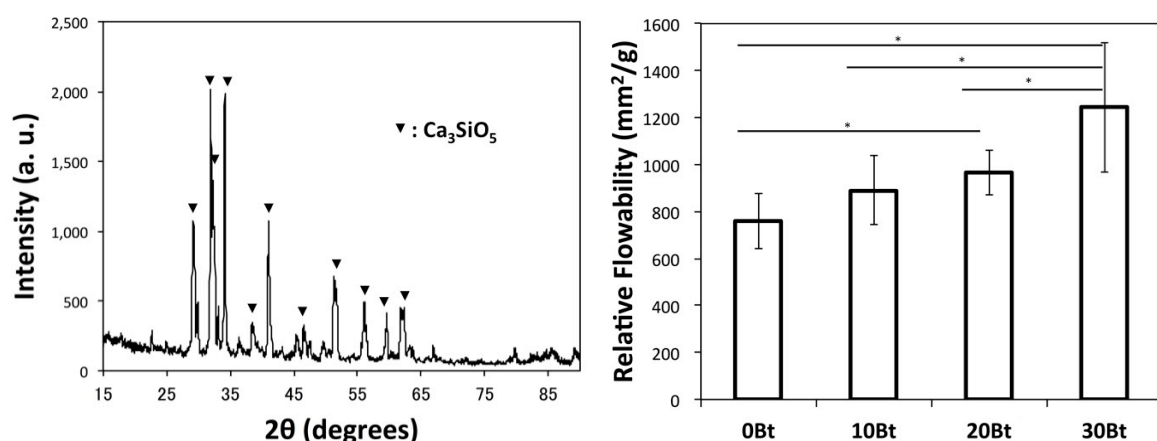


Fig. 3 (left) XRD analysis of TCS powder. ▼ indicates peaks characteristic for Ca_3SiO_5 phase.

Fig. 4 (right) Relative flowability of the cement paste. The 30Bt sample shows significantly improved flowability. All values are expressed as mean \pm standard deviation ($n = 14$). * $p < 0.05$

Relative flowability and injectability

Figure 4 shows the effect of Bt addition on the relative flowability of the experimental cement. The relative

flowability of the sample with 20 wt% of Bt (20Bt) showed small but significant increase ($p < 0.05$) compared with that of the control (0Bt); the sample with 30 wt% of Bt (30Bt), by contrast, showed significant improvement compared to all other groups ($p < 0.05$).

Figure 5 shows the typical load/plunger stroke curves of the syringe injection tests. The load required to stroke the plunger decreased with increasing wt% of Bt. In the case of the 20Bt and 30Bt samples, plateau regions were observed, which show that plunger stroke with an almost constant force had to be applied. In particular, the 30Bt sample exhibited a long plateau region and the plunger was continuously pushed at around 20 N.

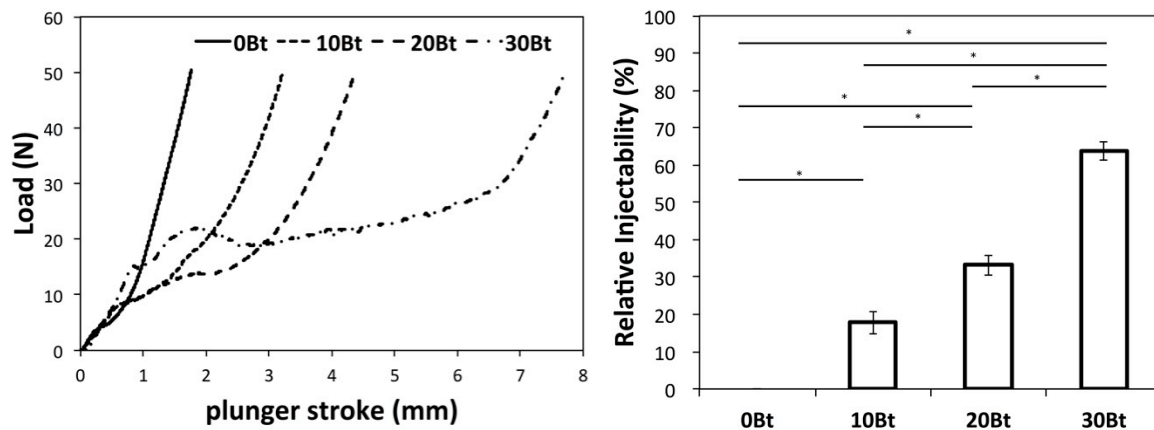


Fig. 5 (left) Load/plunger stroke curves of the cement pastes with added bentonite, as measured by the injection test. The area spanning the plateau region in the curve increased with increasing amount of Bt added.

Fig. 6 (right) Results of injectability analysis. Bt addition increases the injectability of the cement paste. All values are expressed as mean \pm standard deviation ($n = 5$). * $p < 0.05$

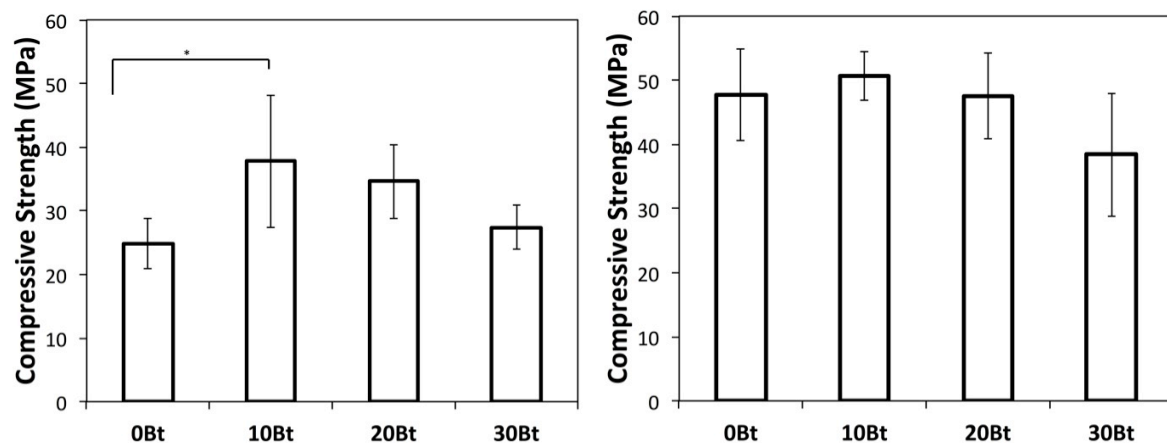


Fig. 7 (left) Results of compressive strength after 1 day. Compressive strength of the 10Bt sample after 1 day exhibited a significant increase when compared to that of 0Bt. All values are expressed as mean \pm standard deviation ($n = 5$). * $p < 0.05$

Fig. 8 (right) Results of compressive strength after 28 days. No significant differences were observed between the samples examined. All values are expressed as mean \pm standard deviation ($n = 5$). * $p < 0.05$

This tendency was also observed in Fig. 6, which shows the dependence of the relative injectability on the Bt content. Relative injectability increased significantly with increasing wt% of Bt ($p < 0.05$). In the case of the 30Bt sample, most of the mixture in the syringe could be ejected.

Compressive strength

Figures 7 and 8 show the compressive strength of TCS and Bt-added experimental cements after 1 day and 28 days. After 1 day, 10Bt showed only a slight increase in compressive strength, while 20Bt and 30Bt samples did not show a significant change. After 28 days, no significant difference was observed between the groups.

Setting time

Figure 9 shows the setting time of the cements. It is apparent that there is no significant difference between the setting times of each group.

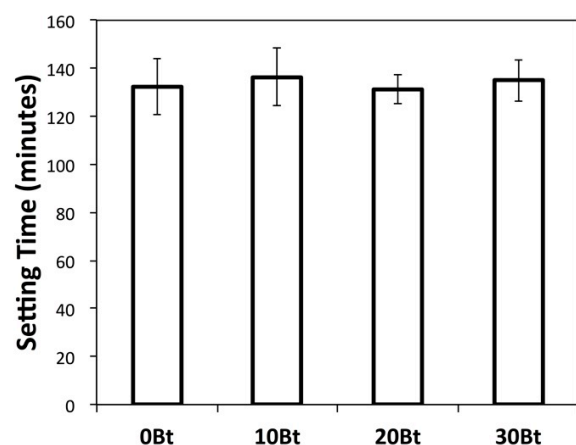


Fig. 9 The results of setting time analysis. No significant differences were observed between the samples examined. All values are expressed as mean \pm standard deviation ($n = 3$). * $p < 0.05$

Discussion

The relative flowability and injectability were estimated as representative handling properties. The results can be summarized as (1) the poor flowability of the TCS paste was improved with Bt addition of more than 20 wt% (Fig. 3); and (2) the TCS paste obtained measurable injectability by the addition of ≥ 10 wt% Bt (Fig. 4). Thus, our hypothesis that “Bt addition improves the rheological and handling properties of TCS pastes” was accepted.

Injectability was measured using a long and narrow syringe at a rapid test speed (15 mm/min) and TCS paste showed no extrusion from the syringe. On the other hand, the addition of Bt to the TCS paste at a quantity higher than 10 wt% improved the injectability. In particular, the 30Bt sample showed a clear plateau region in its injection curve under a constant load (about 20 N). Montufar et al. [30] reported the injection properties of calcium phosphate cement; in their report, the load to syringe stroke dependency appeared to show three phases. In the first phase of the injection curve, the applied load increased rapidly until a yield point was reached. After the yield point, the cement paste was extruded at a constant load (i.e., the second phase). After most of the paste was extruded, the applied load increased rapidly again in the third phase. The 30Bt sample examined in this study displayed similar three phases. Consequently, the results of the syringe tests suggest that Bt addition at over 30-wt% imparts the TCS paste with excellent injectability properties.

Both the relative flowability and injectability improved after the Bt addition, and exhibited similar trends. These improvements can be attributed to the good plasticity of wet clay. The results of the comprehensive analysis confirm that the improvements observed in the flowability and injectability of the Bt-containing TCS paste are a direct consequence of the beneficial properties of bentonite.

In this study, Bt addition caused no significant decrease in the compressive strength of TCS despite of a decrease in the TCS content which is higher compressive strength than Bt. In fact, in the 10Bt sample, a slight increase in compressive strength after curing for 1 day was observed. In the cases of the 20Bt and 30Bt samples,

there was no significant difference in the compressive strength and the handling properties improved more significantly. The compressive strength results obtained for the Bt-added cement can be attributed to the dense condensation of the cement in the mold, caused by the rheological improvements through the Bt addition. The compressive strength of commercial MTA has been reported as; 14.3 and 43.4 MPa after 24 h and 21 days, respectively, for white MTA Angelus [31]; 45.84 and 86.02 MPa after 3 and 28 days, respectively, for ProRoot MTA [32]; and 27.41, 43.65 and 30.77 MPa after 1, 3, and 7 days, respectively, for ProRoot MTA [33]. These data suggest that the setting reaction, which involves the hydration of calcium silicates, which are the main component of MTA, continues after the initial setting and leads to an increase in strength. The TCS-based cement samples examined in this study showed a relatively higher compressive strength than the commercial MTA after 24 h. However, after 28 days, their strength was lower than that of the commercial MTA. This result is directly linked to the fact that MTA cements are based on Portland cement, which consists not only of TCS but also dicalcium silicate, calcium aluminate, and other compound oxides. TCS shows a faster hydration reaction when compared to others such as dicalcium silicate [34]; consequently, the TCS-based cement in this study showed a higher compressive strength after 24 h.

Further, the Bt addition induced no acceleration or retardation effect on the TCS setting process. Bt does not undergo hydration reaction, and thus, the setting behavior depends on the TCS hydration reaction only. Therefore, the lack of dependence of the setting time on the Bt content is a reasonable and predicted outcome. Further, the setting times of commercial MTAs have been reported as 48.3 min for white MTA-Angelus [31], 140 min for ProRoot MTA white [32], and 219 min for ProRoot MTA [33]. The estimated setting time measured in this study was about 130–140 min, regardless of the Bt content and was identical to the value reported for ProRoot MTA.

The flowability and injectability of the TCS paste was improved by the addition of increasing amount of Bt. In particular, addition of 30 wt% of Bt conferred both flowable and viscous properties on the TCS paste with no significant change in the setting and mechanical properties. Thus, this study demonstrated that Bt is a promising additive of TCS-based cements for improved handling properties.

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Conflicts of Interest

The authors declare no conflicts of interest.

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