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WATER STABILIZATION OF CLAY BRICKS WITH IMPROVED TANNIN AND IRON MIXES

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Abstract

Weak water resistance is a big obstacle for clay materials to overcome in modern construction industry. Compared to the hydraulic stabilized additives, bio-additives have a lower carbon footprint and have been used in many vernacular construction techniques to immobilize clay. In this work, the traditional recipes of tannin and iron have been revisited, in particular, the question of pH and iron solubility has been explored. Oak tannin and $FeCl_3$ were chosen and their influence on the properties of clay materials in terms of rheological properties, compressive strength, and water resistance were characterized in the lab. Based on the results, tannin can reduce the yield stress of paste while with the addition of $FeCl_3$, the yield stress of tannin dispersed pastes increased to a value similar to the reference sample but lower than the value contain only $FeCl_3$. The increase was attributed to the complex reaction between tannin and Fe^{3+}. The iron-tannin complexes can also increase the samples' strength and water resistance. Although the complexes did not change the hydrophilic properties of the samples' surface, they prevent the ingression of water. These results are very promising as they allow the production of a fluid earth material that is water-resistant. This opens a wide range of application potentials and can help to mainstream earth materials in construction.

Keywords:

Clay materials; Water resistance; Tannin; Iron compounds; Complexation.

1 INTRODUCTION

As one of the most local and available construction materials found on the Earth, earth has been used as building materials since the Neolithic age. It can be locally sourced and used directly with almost no treatment (Hugo Houben, 2014). However, the development of urbanization expanded the need for housings for people. Leading by the urgent desire for modernity and with the invention of concrete, which is fast and robust construction materials, these traditional building techniques have been forgotten by designers and builders. As a consequence, large worldwide consumption of concrete in the building industry has brought a critical environmental impact to the earth, e.g. huge greenhouse gas (GHG) emission and great energy consumption (Habert et al., 2020). The impact caused by concrete will worsen in the following decades as concrete is still needed for the urbanization process in the developing regions. For this concern, the attention of the construction sector in recent years has been turning to the environmentally friendly alternative construction materials. Owing to its lowest environmental impact and infinite recycling possibilities, raw earth regains the interest. However, its massive application in the modern construction industry is still hindered by its time and labour intensive process as well as by its low strength and high water sensitivity, making it difficult to fulfill the nowadays building standard (Matthew R. Hall, 2012).

To improve the strength and water resistance, the most common practice is to stabilize clay materials with a hydraulic binder, such as cement or lime. Due to the hydration products formed in the matrix, a low dosage (less than 10%) of hydraulic binder can increase the strength by 3 to 4 times higher compared with the non-treated samples (Shubbar et al., 2019). However, their use as earth additive is questioned because of the generated environmental impact. Indeed, cement or lime help to increase the mechanical properties and durability of clay materials, but this increase is on the prices of higher GHG emission and large energy consumption which negatively counterbalance the environmental advantages of clay materials (Van Damme and Houben, 2018).

Looking for alternative solutions, additives with a low environmental impact that can improve the water-resistance of clay materials are available in nature. Mixing natural additives with clay to improve the strength and water resistance is not an entirely new development in earth construction and various additives have been recorded in the traditional construction recipes (Laetitia Fontaine, 2013). Recently, some of them, like casein (Chang et al., 2018, Fatehi et al., 2018), starch (Alhaik et al., 2017) or alginate (Pinel et al., 2017), have been studied thoroughly in the literature. However, until now this area is not fully explored.

In this paper, natural additives oak tannin and iron chloride were combined to modify the clay properties in terms of strength and water resistance. The influence of oak tannin (2wt% by mass of clay) and iron chloride addition (0.19, 0.57, 0.95wt% by mass of clay) on the fresh (rheology) and hardened state (compressive strength and water resistance) properties of clay pastes materials was studied.

2 EXPERIMENTAL

2.1 Materials

A pure kaolinite clay (FP80) bought from Dorfner, Germany, was used in this study. As determined in the lab, its density, specific surface area, and average size are 2.62 g/cm^3, 9.10 m^2/g, and 8.7 µm respectively. To avoid the formation of cracks, a standard sand according to EN 196-1 sourced from SNL (France) was used to prepare the mortar samples. The tannin used in this study is a commercial oak tannin (referred to here as OT) bought from Agrovin (Spain). It was extracted from lightly toasted French oak (Quercus robur and Quercus petraea). The main components of this product are hydrolyzable tannins. Iron chloride ($FeCl_3$, referred to here as IC) in powder form with a reagent grade of 97% was used (Sigma-Aldrich, Switzerland).

2.2 Sample preparation

For the rheological measurements, kaolinite pastes were prepared using the following mixing procedure: 2wt% by mass of clay of tannin was mixed manually with kaolinite clay before adding distilled water. Then the mixture was mixed with a mechanical stirrer for 2 min at 700 rpm, and mixed again for 2 min after the introduction of iron chloride. The water to clay ratio of 1 was kept constant for all the mixes. A mechanical mixer was used to prepare the mortar samples. The solid volume fraction of sand was kept constant at 50%. Only the dosage of additives varies from one sample to another. These variations are discussed in the relevant sections of the paper. All the mixes were prepared at room temperature (23±1°C).

2.3 Test methods

2.3.1 Rheological measurement

The rheology measurements were performed using a Malvern Kinexus Lab + (Malvern, Switzerland) stress-controlled rheometer equipped with a Vane geometry. The vane geometry consisted of four bladed paddle with a diameter of 25 mm, the outer cup diameter was 27.5 and its depth was 63 mm. The shear stress-rate cycle within the range of 0.1 s^{-1} to 300 s^{-1} over 300 s was applied in a shear rate-controlling regime. Only the decreasing ramps were analyzed for the determination of the yield stress, extrapolated at low shear rates through a linear regression acoording to the Herschel-Bulkley model $\tau = \tau_0 + \eta_p \gamma^n$, where τ is shear stress (Pa), τ_0 the yield stress (Pa), η_p the viscosity (Pa.s) and γ the shear rate (s^{-1}).

2.3.2 Compressive strength

The compressive strength of samples with dimensions of 40*40*160 mm^3 was measured with a hydraulic press (Matest, Switzerland). Before crushing, samples were dried at room temperature (23 ± 1°C) for 28 days and three samples were measured for each mix to minimize the deviation.

2.3.3 Water resistance

Water resistance was measured using the dip test method conforming to DIN 18945. After drying process, the prismatic sample was dipped 10cm into the water for 10 mins. The loss of materials was filtered out from the water and dried in the oven until weight stabilization. With the initial mass of the sample, the mass loss in percentage was estimated and the water resistance was evaluated according to the standard.

3 RESULTS AND DISCUSSION

3.1 Rheological properties of paste with tannin and iron chloride

The yield stress of tannin dispersed kaolinite pastes after adding iron chloride are presented in Fig. 1. To compare the effect of iron chloride on the rheological properties of pastes, results of solely using $FeCl_3$ are also plotted in the figure. As seen from the results, 2wt% OT was already enough to tune the kaolinite paste from coagulated state to highly dispersed state. The reduction of yield stress is due to the acidic and positively charged characters of tannin. As the clay rheological behaviour is controlled by their surface charge (Landrou et al., 2018), introduced tannin can increase the particles' charge density by adsorbing onto the kaolinite surface, thus disperse the colloidal particles. However, after iron chloride addition, the yield stress of pastes increases to a value similar to that of the reference sample, but lower than the pastes solely with $FeCl_3$. The pastes changed from deflocculated state to a coagulated state, even just with a small amount of iron chloride. This shift is attributed to the complexation reaction between tannin and iron chloride as displayed in Fig. 2. Tannins are prone to form stable complexes with many metal ions. The phenolic hydroxyl groups in tannin have a high affinity to chelate the metal ions, especially transition metals (E.Sieniawska, 2017, Khanbabaee and van Ree, 2001). When iron chloride was added into the mix, the free iron ions can precipitate the tannin from solutions and remove the tannin from kaolinite particles, annihilating the dispersing effect.

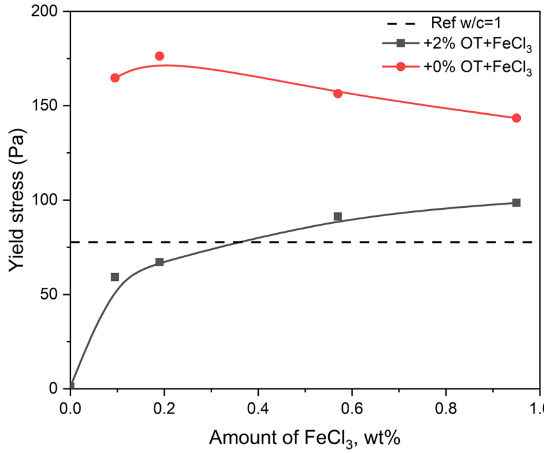

Fig. 1. Yield stress of kaolinite pastes with varied mixes.

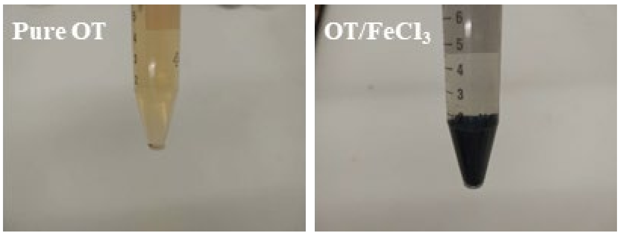

Fig. 2. Pure oak tannin solution alone (left) and with iron chloride (right). Precipitate with a dark-blue color can be seen when the tannin solution was encountered with free iron ions.

3.2 Compressive strength and water resistance

Fig. 3 presents the compressive strength and water dip test results of mortar samples after drying for 28 days. It can be seen that samples with 2wt% OT have a higher compressive strength than the reference sample. This seems to be related to the higher fluidity of the mix, which would densify the paste and reduce the final porosity (Ardant et al., 2020). It can be noted that samples with 2wt% OT have the highest dried density, followed by the reference sample and the lightest are the ones with tannin and iron chloride. However, mixes containing tannin and iron chloride together are stronger than the reference sample, suggesting that the formed complex plays a critical role in the compressive strength. Keita and Sorgho (Keita et al., 2014, Sorgho et al., 2014) have reported that the formation of tannin-iron hydroxide bond helps to increase the macroscopic strength and reduce micro-cracking. It can be observed that the maximum strength in presence of $FeCl_3$ is reached when 0.57% $FeCl_3$ is added to the dispersed clay paste. This might suggest that the maximum ratio between tannin and iron chloride for the complexation is 1: 0.29.

Thanks to the water dip test, it appears that the clay samples are getting more water-resistant when tannin and iron chloride were used. Without any treatment, the disaggregation of the reference sample is inevitable and a mass loss of around 24.4wt% was recorded. This is far from meeting the requirements of DIN 18945, where less than 5% mass loss is considered acceptable. With the introduction of the tannin, the mass loss is reduced by a factor 2. The mass loss is drastically reduced when iron chloride is added. The higher addition of $FeCl_3$, the more prominent of improvement. The samples with 2wt% OT and 0.57wt% IC remain almost intact after immersing in water for 10 mins. Clay material's water resistance is closely related to the compressive strength. Indeed, in Fig. 3, the sample with 2wt% OT and 0.57wt% IC exhibit similar strength to the sample containing 2wt% OT sample, but its mass loss is significantly lower. The improvement of water resistance when tannin only is used may be attributed to the increase of strength, linked to a reduced porosity. Nevertheless, it can be suggested that another water stabilization mechanism exists for the sample prepared with tannin and iron chloride. This might be due to the complexes accumulation and oxidation at the surface of the sample, a hypothesis that has to be verified with further characterizations.

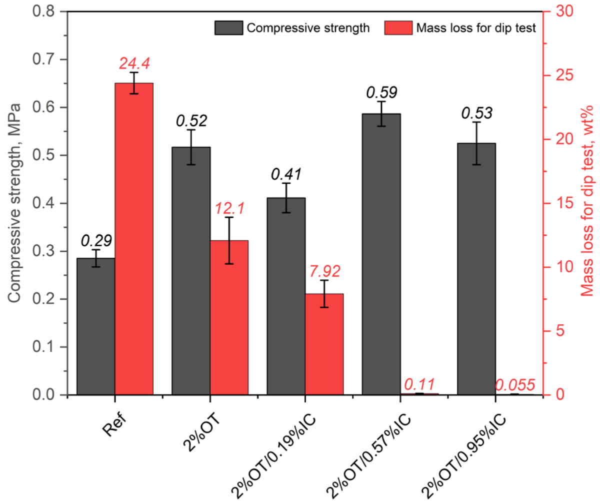

Fig. 3. Compressive strength and water dip test results of clay mortar containing different additives.

4 CONCLUSION

In this paper, an environmental-friendly additive oak tannin was combined with iron chloride to increase the water-resistance of clay materials. Their influence on the clay materials properties in terms of rheological properties, strength, and water resistance were characterized. The results show that the water-resistance can be greatly improved with the help of oak tannin and iron chloride, with a strength two times higher than the non-stabilized one. The improvement was more likely related to the iron tannates formed in the mix, which can be oxidized into a dense layer at the end. However, more analytical techniques, like TG and SEM, need to be used to support this hypothesis. Besides, since the complexation reaction between iron chloride and tannin happens very fast in the mix leading to fast coagulation of the initially deflocculated paste, further work on the influence of iron sources and their reaction kinetic needs to be explored to better master this technique before its use for poured earth.

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