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Super adsorbent bio-polymer additive to improve hygroscopic and acoustic properties of a conventional lime plaster

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Abstract. The paper introduces a new lime plaster composition, with a Super Bio-Polymeric Adsorbent (SABP), for interior applications to improve hygrothermal and acoustic comfort. Alginate SABP is added to a conventional lime plaster to improve hygroscopic and acoustic performance. The hygrothermal and acoustic properties of the modified plaster are compared with the ones of the original plaster with a preliminary moisture uptake test, the evaluation of the sorption isotherm, the moisture buffering value (MBV), and the acoustic absorption. The results show a significant increase in the equilibrium moisture contents and the MBV (from 0.7 to 6.2 g/(m²·%RH)). At the same time, the sound absorption coefficient is slightly improved, increasing 0.1-0.2 at frequencies higher than 500 Hz compared to the reference conventional lime plaster. The paper describes the material characterization: the sorption isotherm and the MBV are obtained using a dynamic vapor sorption (DVS) analyzer. The acoustic absorption is measured using the impedance tube method.

1. Introduction

The addition of hygroscopic materials within plaster compositions is a practice investigated by academia as a potential passive solution to improve the buffering of moisture variations of building materials for indoor applications [1][2]. Moreover, the addition of hygroscopic materials gives as the collateral effect the increase of plaster porosity, which can also lead to improved sound absorption characteristics [3][4].

The common practice adopted for measuring hygroscopic properties of building materials is the NORDTEST [2], and the evaluation of the Moisture Buffering Value (MBV) is the typical method to compare different materials. Usual plasters, such as gypsum and lime, have experienced moderate values of MBV within the range of 0.6-1.5 g/(m²·%RH) [5] and own the label of *moderate* and *good* materials according to NORDTEST. A typical strategy to increase the MBV of construction materials, like cement and limestone, is using additional hygroscopic compounds, like natural or synthetic fibers such as palm and hemp. The use of 50% of palm fiber in cement mixtures can increase the MBV up to 3.7 g/(m²·%RH) [6]. On the other hand, conventional sorbents can potentially contribute to higher moisture affinities and less use of material. Consequently, the MBV can increase easier.



The addition of zeolite, for example, increases the number of active sites to operate the moisture sorption and desorption that, together with an increase of global porosity, improve the buffering properties of the mortar. Based on this concept, He et al. [7] tested a composite mortar based on zeolite (20%wt), PCM, and gypsum, obtaining a practical MBV of 2.9 g/(m²%RH), consistently higher than pristine gypsum plaster [5].

An alternative approach belonging to the same category is using hygroscopic polymers. Fort et al. [8] realized samples of plasters with different contents of cellulose fibers and a commercial polymer named Hydropam. The MBV consistently increased from 1 to 2.9 g/(m²%RH), while thermal conductivity slightly reduced. As a counter effect, authors observed reduced compressive and flexural strength. Gonçalves et al. [9][10] studied the impact on mortars of additives to increase macro-porosity and a commercial hygroscopic polymer named Creasorb. The authors observed that adding 0.5wt% of the polymer in the mortar composition caused a 20% increase of the MBV, up to 1.3 g/(m²%RH). Other authors used the same polymers in similar concentrations with equivalent results [11]. Instead, an increased concentration of hygroscopic polymer was used in [9] and [12], between 1.5-5%wt of the composition, with an augmented MBV value of 2.55 g/(m²%RH), 113% higher than the reference sample. As in other studies, the higher concentration of hygroscopic polymer was correlated with a reduction of 62% in compressive strength. Indeed, the ionic nature of the polymers, together with their interconnected chain structure of particles, may cause an increase in water absorption during the mixing phase reducing the workability of the mortar and affecting the final result. As in Senff et al. [13], a hygroscopic polymer derived from sodium polyacrylate was incorporated into a plaster composite of hydrated lime and Portland cement at a concentration ranging from 13-27%wt. The maximum obtained MBV value was 1.08 g/(m²%RH), with a much lower increase of 17% from the reference.

Similarly to the MBV improvement strategies, different aggregates control fundamental properties, i.e., flow resistivity, porosity, pore shape factor, and tortuosity, that affect sound absorption performance. This can be optimized by designing the microstructural configuration of the mix design, knowing that by adequately controlling the void ratio and aggregate type, the tortuosity and flow resistivity [14] can be designed. In contrast, by controlling pore size, and pore aperture size, the porosity can be optimized [15]. Several reviews on the strategies that have been proposed to enhance the sound-absorbing performances of cementitious materials have been presented in [16][17].

2. Methodology

In this work, the authors added an Alginate Super Bio-Polymeric Adsorbent (SABP) to a conventional lime plaster to improve hygrothermal and acoustical performance. The hygrothermal and acoustic properties of the new plaster are evaluated and compared to the traditional lime plaster. The modified plaster preparation is illustrated in the following sections, followed by the description of each experimental activity and the respective results.

2.1. Material preparation

Recently, alginate-derived polymers have been considered building materials aggregates with different functions and mixtures. The interest is related to compressive and flexural strength [18] and the resulting concrete's self-curing property [19].

The interest of this study is the evaluation of the hygroscopic and acoustic performance of composite lime plaster with bio-polymer beads to evaluate their potential application for the indoor environment. The hygroscopic polymer used in this research is calcium alginate (CaAlg), a bio-polymer obtained through the ionotropic gelation [20] of sodium alginate monomers with calcium ions dissolved in a calcium chloride/water solution. Sodium alginate is a salt obtained from the polysaccharide compounds that make up the cell walls of brown algae [21]. Calcium Alginate was produced by mixing 1 kg of deionized water with 20g of the gelling agent, the sodium salt from the alginic acid, and rapidly stirred at high rotational speed until the complete homogeneity of the solution. The

resulting viscous gel is degassed under vacuum at relative pressure between 200-500 mbar and dripped into the crosslinking salt solution, prepared by mixing 1L of deionized water and 133g of dehydrated CaCl_2 . To complete the diffusion of Ca^{2+} , then the complete crosslinking of internal beads volume, the bath is rested for around 12 hours. Finally, crosslinked beads are drained and deeply rinsed with DI water to remove the salt excess over the external surface, then dried in a ventilated oven at a maximum of 70°C . The mortar is a composite limestone plaster based on natural hydraulic lime and recycled material with limestone and microfibrils. The bio-polymer beads of CaAlg (Figure 1a) are first homogeneously mixed with the mortar powder and then mixed with water. Two different samples were prepared: the reference material (*Ref.*) with 500g of plaster and 83g of water, while the CaAlg added selection (*Add.*) was realized with 450g of plaster, 90g of CaAlg, and 111g of water. The different mixtures were poured into a squared mold, as shown in Figure 1b, and rested at ambient temperature and humidity for about one week to complete the drying process. The final size of samples was a square of 109.5 mm and a thickness of 20.8 mm for *Ref.* and 22.4 mm for *Add.* To obtain the dry basis of each piece, an additional drying process in a ventilated oven at 70°C was carried out for 72 hours and finally weighted to get the m_0 value reported in Table 1.

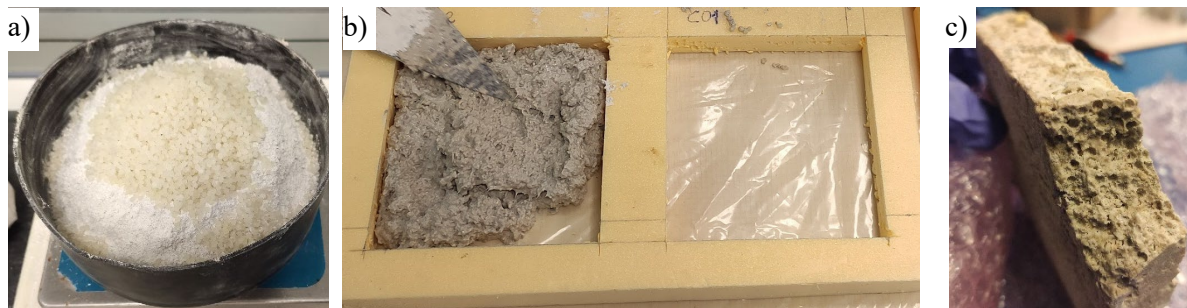


Figure 1. Samples preparation: a) Mixing the bio-polymer beads of CaAlg and mortar powder; b) Insertion of the wet mixtures into the squared molds; c) Picture of the dried alginate plaster.

3. Preliminary moisture uptake test

A preliminary moisture uptake analysis was carried out to obtain a first rough comparative evaluation of the capability of the developed material to absorb moisture from a humid environment. The objective of this first test was to obtain indications to decide whether further investigations and optimization on the developed material are worthy of being explored.

For this test, two sealed plastic containers were used. Each vessel contains 100 g of Potassium Nitrate KNO_3 saturated solution that allows reaching $\sim 93\%$ of relative humidity at 23°C . In Figure 2, the samples and the test weighing process are documented.

Samples were dried at constant mass in a stove at 60°C until a constant mass was reached, then pieces were placed in the container, and they were weighed again after 96h.

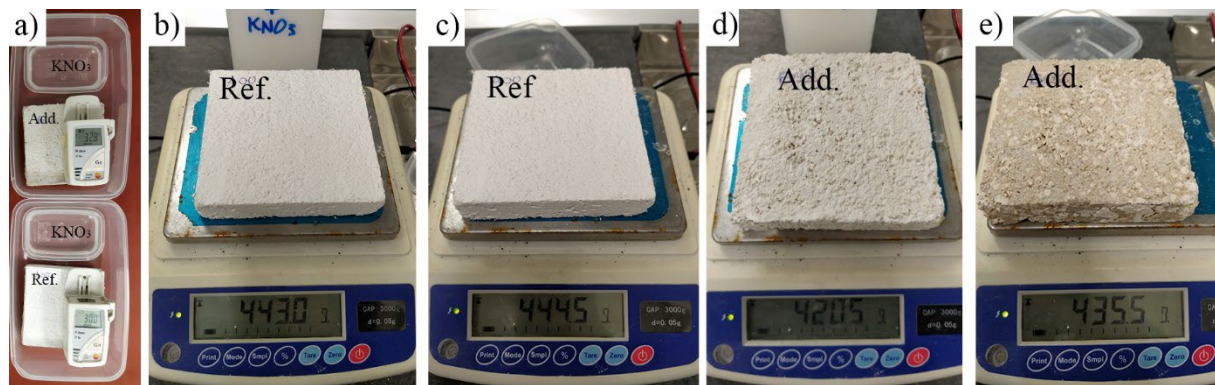


Figure 2. Test pictures: a) Samples in sealed plastic containers; b) *Ref.* (dry condition); c) *Ref.* (after 96 h at 23°C - 93% RH); d) *Add.* (dry condition); e) *Add.* (after 96h at 23°C - 93% RH).

Table 1. Results of the weighing process on dry and moist samples.

Sample	Dry mass m_0 [g]	Moisture mass m_1 [g]	Mass change Δm [g]	Moisture Uptake at 23°C, 93% RH (mass fraction) [%]
Ref.	443.0 ± 0.5	444.5 ± 0.5	1.5 ± 1.0	0.3 ± 0.2
Add.	420.5 ± 0.5	435.5 ± 0.5	15.0 ± 1.0	3.6 ± 0.2

In Table 1, the test results are summarized. As it is possible to observe, exposure to a humid environment (23°C and 93% of relative humidity) results in a mass change of ~1.5 g and ~15 g for the Reference plaster (*Ref.*) and for the plaster with the addition of alginate spheres (*Add.*). So, the newly developed materials have shown about one order of magnitude higher moisture uptake capacity than the reference plaster.

4. Sorption isotherm

Mortar cylindrical samples were prepared with a circular base of 34.5mm in diameter, with different thicknesses, and then polished to reach the height of 13mm. These samples are used to perform the measures of the sorption isotherm points and the evaluation of the moisture buffering value. The reference material sample has a dry mass of 18g, while the modified sample is 17g.

A critical limitation to using the plaster with the additive is that, beyond 80%RH, the surface of the pores begins to accumulate droplets of the solution, showing a deliquescent behavior. For this reason, the analysis is limited to the hygroscopic range up to 70%RH. The presence of a deliquescent behavior of the *Add.* The sample above that relative humidity threshold. This could be caused by salt residues from the preparation procedure and will be the object of future investigations.

The water vapor sorption isotherms are measured with the dynamic water vapor sorption analyzer system ProUmid "Vsorp Basic" at the Thermal Systems Lab (DPIA, Università degli Studi di Udine). The isotherms are obtained with the equilibrium conditions measured at 0%, 40%, 50%, 60%, and 70%RH, with a precision of $\pm 0.1\%$ RH. The samples are placed in aluminum plates that are 86mm in diameter and kept in a climatic chamber at $23 \pm 0.1^\circ\text{C}$ and variable relative humidity. The aluminum plates are kept on a rotating tray, automatically moving the plates on the scale. The rotation is also performed between the weightings to allow a uniform exposition of the air flux. The average air speed in the chamber is 0.15m/s, kept by two fans. The weightings are performed automatically every 30 minutes, with a scale reproducibility of 0.1mg, without opening the climate chamber. The air conditions are subsequently set to the relative humidity steps until the equilibrium condition is met. The equilibrium condition is obtained when the mass variation is lower than 0.001% for more than 300 minutes. This equilibrium condition is set to get the equilibrium condition defined in ISO 12571:2021, yet using a shorter period as a reference. The ISO 12571:2021 equilibrium is defined as the condition obtained after three weightings, made 24 hours apart, with a mass change lower than

0.1% of the total mass. The equilibrium condition, set in the device control software, is met when the linear regression slope of the last 300 minutes measurements is smaller than 0.001% of mass change over 300 minutes. Each equilibrium condition was reached in about 100 hours for each step. Before measuring the adsorption curve, the samples are dried in the “V sorp Basic” at 0%+0.1 RH and 23°C. The average of the last five measures of each RH step is used to calculate the points of the isotherm and the uncertainty of the measure. The uncertainties are all lower than 0.3mg, which leads to uncertainties in the moisture content lower than 0.0002% mass/mass. The results are presented in Figure 3 and Table 2. The modified plaster showed high equilibrium moisture content, up to 134 kg/m³ at 70% RH, higher than most bio-based materials such as wood.

Table 2. Equilibrium moisture contents of the samples.

RH	Ref.		Add.	
	[% m/m]	[kg/m ³]	[% m/m]	[kg/m ³]
0%	0.00	0	0.00	0
40%	0.35	5	4.99	68
50%	0.39	6	6.23	84
60%	0.42	6	7.74	105
70%	0.48	7	9.91	134

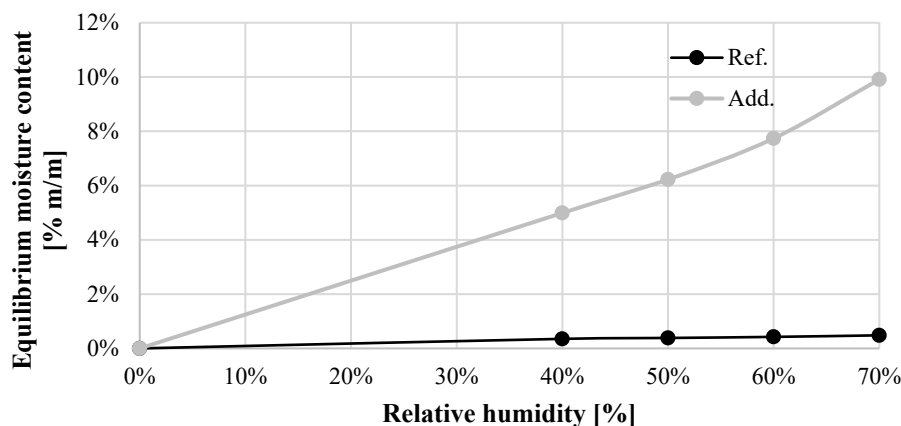


Figure 3. Sorption isotherm of the samples.

5. MBV analysis

The moisture buffering performance of the studied mortars is measured using a variation of the procedure presented in [2] for the practical moisture buffering value (MBV). The procedure followed complies with the NORDTEST protocol, except for the exposed face size smaller than 0.01m². The same samples used to measure the points of the moisture isotherm are used to perform the Moisture Buffering Value evaluations. All but one surface of the samples used to measure the sorption isotherm is covered with aluminum tape and sealed with acrylic silicone. The pieces are placed in aluminum trays with the free surface facing upwards. For this test, samples are dried at 23 °C, then conditioned to 50% RH at the same temperature. Then, the samples are exposed to the practical Moisture Buffering Value test conditions described in [2]: the temperature is set to 23°C constantly, while the RH is fixed to 75%RH for 8 hours, then 33%RH for 16 hours cyclically for seven consecutive cycles, with an average air velocity of 0.15m/s.

Figure 4a shows the total mass variation of the samples measured during the MBV test, while in 4b, the MBV values are calculated at every cycle. The average value of the practical MBV for the *Ref.* sample is 0.70±0.01, while for the *Add.* is 6.22±0.21g/(m²%RH). During the moisture buffering experiment, in the 75%RH step, deliquescent behavior is not observed.

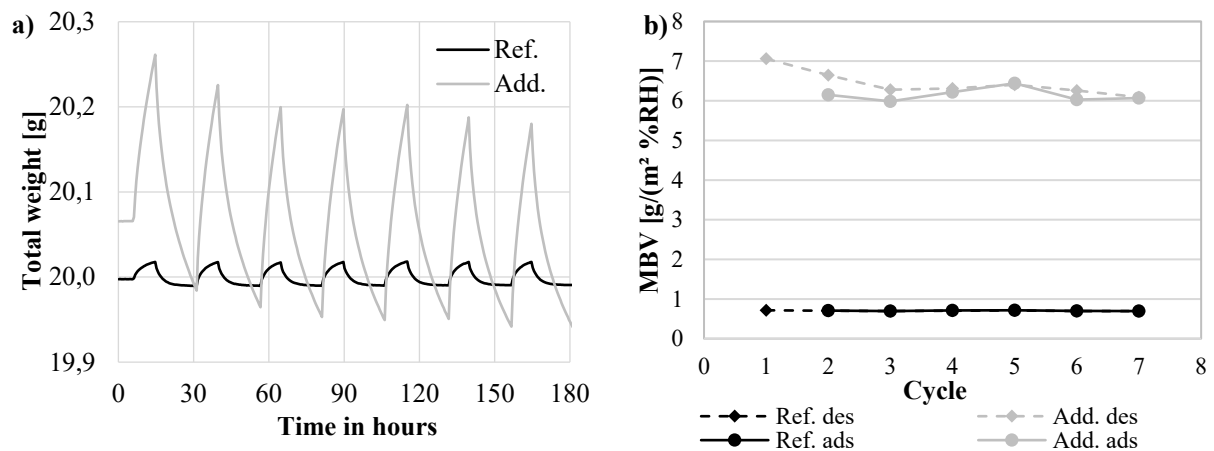


Figure 4. a) Weight variation of the samples during the MBV test; b) MVB values at every cycle.

6. Acoustic absorption

Measurements have been performed in the impedance tube by ISO 10534-2 [22] (two-microphone technique) to measure the normal-incidence absorption coefficient (α_0). The advantages of this method rely on the possibility of obtaining measurements using small samples suitable to the aim of this investigation. These measurements took place in the Applied Acoustics Laboratory (DENERG, Politecnico di Torino) using an impedance tube HW-ACT-TUBE, which has a diameter of 35 mm and is equipped with two $\frac{1}{4}$ " flush-mounted GRAS 46BD. The method allows accurate sound pressure amplitude and phase measurements in the frequency range of interest, i.e., 100-5000 Hz [22].

The tube's geometry follows the ISO standard's specifications, including the minimum distance between the microphone and source, and the microphone and test sample. A 29-mm microphone spacing is used for accurate measurements at a high-frequency range of 119 to 5700 Hz. The white noise source, i.e., a 2" aluminum driver, can produce continuous high sound levels (100 dB) inside the tube, assuring a high signal-to-noise ratio by design, and generates a flat spectrum in the 100-5000 Hz frequency range.

The measurements have been performed on single samples for each typology (*Ref.* and *Add.*). The frequency-dependent sound absorption has been evaluated and presented in third-octave bands (Figure 5). The graph shows that the *Add.* plaster has a more uniform overall distribution of frequency-dependent sound absorption. In contrast, the *Ref.* plaster presents a typical peak around 1000 Hz third-octave band. In particular, the higher sound absorption results in the developed *Add.* plaster, at both low frequency (<800 Hz) and high frequency (>1800 Hz).

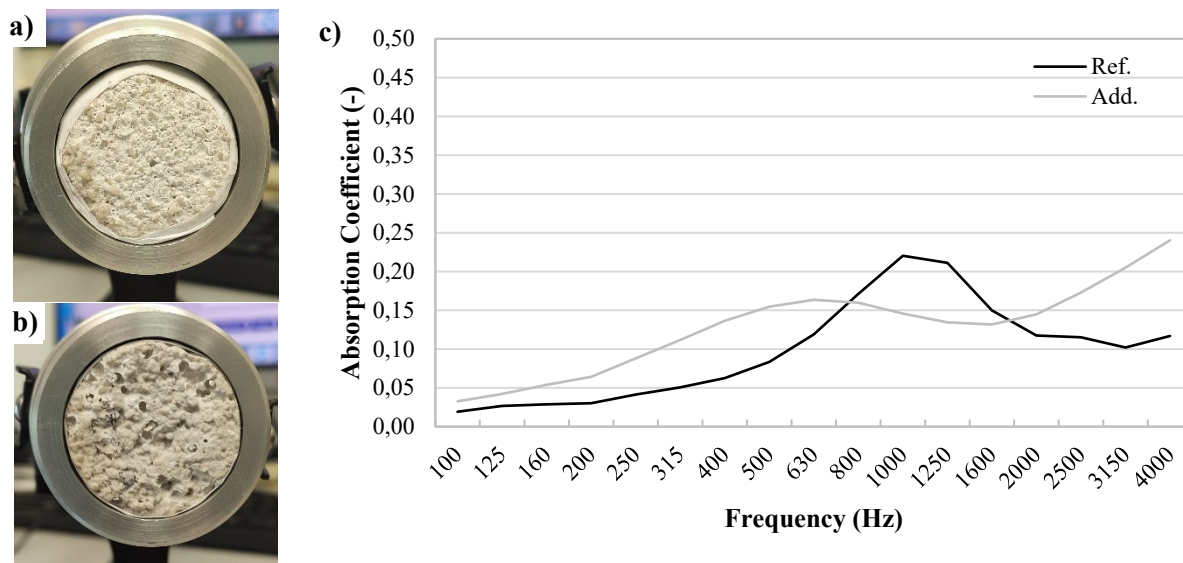


Figure 5. a) and b) are the pictures of samples *Ref.* and *Add.* inside the impedance tube HW-ACT-TUBE c) Normal-incidence sound absorption coefficient of *Ref.* and *Add.* samples.

7. Conclusion

This study developed a new lime plaster composition with a Super Bio-Polymeric Adsorbent (SABP). A comparative analysis between reference lime-based plaster formulation samples (*Ref.*) and the developed plaster with the addition of SABP (*Add.*) samples have been carried out.

After a first preliminary moisture uptake test, it was demonstrated that the newly developed SABP plaster (*Add.* sample) presents an order of magnitude higher moisture storage capabilities at 23°C and 93% relative humidity.

In a second experimental campaign, with a Dynamic Vapor Sorption Analyzer, the sorption isotherm and the practical Moisture Buffer Value MBV were measured on the *Ref.* and the *Add.* samples.

The sorption isotherm test confirmed the behavior observed in the first preliminary experimental campaign. In particular, the *Add.* sample has shown a higher moisture uptake at the different humidity conditions (between 13 and 19 times higher at 40% and 70% relative humidity, respectively). Moreover, it was demonstrated that the new lime-SABP plaster (*Add.* sample) had reached a practical MBV of up to 6.2 g/(m²·%RH), with an increase of about nine times if compared with the reference lime plaster (*Ref.*).

Acoustic measurements have been performed in the impedance tube to assess the normal-incidence sound absorption coefficient (α_0). Test results have revealed an improvement of the sound absorption showing a higher sound absorption in the developed *Add.* plaster, at both low frequency (<800 Hz) and high frequency (>1800 Hz), while slightly lower sound adsorption has been measured between 800 Hz and 1800 Hz. Overall, the *Add.* plaster shows a more even distribution of the frequency-dependent sound absorption, which makes it useful for several applications in room acoustics.

The promising results have been considered as a starting point for further investigations of the design of new optimized mixtures to maximize the moisture buffering and sound absorption performance while also assessing mechanical, aesthetic, durability, and thermal insulation properties.

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