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MORTARS FOR THE ENHANCEMENT OF THE INDOOR ENVIRONMENTAL QUALITY

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ABSTRACT. Achieving a healthy indoor environment is an important concern in the construction and building materials industry. This study fits in the context of these concerns and demonstrates the possibility of developing mortars that contain colloidal silver nanoparticles with good antibacterial properties. The optimal recipe was established by replacing 50% of the amount of water in the mortar recipe with a commercial 25 mg/L colloidal silver solution. The obtained mortars have superior physical and mechanical characteristics and excellent antibacterial properties against various Gram-positive and Gram-negative bacteria compared to standard mortar.

Keywords: *mortar, colloidal silver nanoparticles, mechanical properties, antibacterial activity*

INTRODUCTION

The indoor air quality is a determining factor of health due to the fact that people spend most of their lives inside buildings. It has been observed that microbes and allergens, the lack of oxygen, inadequate temperature and humidity, mould, dust, inadequate lighting and ventilation, noise, the presence of building materials that contain noxious substances and generate toxic emissions, the functioning of equipment, etc. can seriously affect a person's health [1].

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Infections with pathogenic microorganisms are combated with antimicrobial agents, to the action of which these are vulnerable. The use of antimicrobial agents in decorative and building materials, including paints and coatings, has intensified considerably in recent years and their possible effects on human health or the environment have been increasingly studied [2]. Noble metal nanoparticles have attracted great interest due to their different characteristics from those of the macroscopic phase, which allow attractive applications in various fields, such as medicine, biotechnology, optoelectronics, biosensors, catalysis, information storage, energy conversion, and as antimicrobials [3-5].

Silver nanoparticles, an ecological alternative to organic biocides, have a high surface area, a very small size (<20 nm) and high dispersion [6]. Silver nanoparticles can be used in the form of colloidal suspensions or doping agents for a lot of composite materials with polymer matrix. Colloidal silver solutions (CSSs) arouse increased interest due to their antimicrobial properties with different applications (pharmacology, food, industry, human and veterinary medicine, etc.). The interaction of silver nanoparticles with microorganisms, such as viruses, mould, bacteria and fungi, is an expanding field of research [7, 8]. While the mechanism underlying the antibacterial actions of silver is still not fully understood, some previous studies have shown that silver ions penetrate the bacteria and interact with thiol groups of vital enzymes and inactivate them, leading to cell death [9].

Furthermore, colloidal silver solutions can be taken indefinitely because they have no side effects, the body develops no tolerance, and one cannot overdose. Unlike pharmaceutical antibiotics, which kill beneficial enzymes, colloidal silver leaves these tissue-cell enzymes intact. Therefore, colloidal silver is completely safe for humans, plants and all multicellular living organisms [10].

The purpose of this work was the development of a plastering mortar with antibacterial properties using a commercial 25 mg/L Ag solution as an additive. The study focuses on obtaining a plastering mortar with good adhesion to the substrate, low water absorption by capillarity, good mechanical and antibacterial properties.

RESULTS AND DISCUSSION

Physical-mechanical properties

The results for the physical-mechanical properties (apparent density of hardened mortar, flexural and compressive strengths, water absorption by capillarity and adhesion to the substrate) are shown in Table 1.

Table 1. Technical characteristics of the mortars

Recipe	Apparent density [kg/m^3]	Adhesion to the support layer [N/mm^2]	Flexural strength [N/mm^2]	Compressive strength [N/mm^2]	Water absorption by capillarity [Kg/(m^2·min$^{0.5}$)]
R I	2210	0.211	6.93	42.7	0.161
R II	2223	0.191	7.31	43.2	0.133
R III	2233	0.193	7.53	43.3	0.121
R IV	2237	0.211	7.61	43.7	0.135
R V	2190	0.189	7.26	42.9	0.191

Table 2 shows the influence of the commercial 25 mg/L Ag solution on the technical characteristics of plastering mortar.

The apparent density of the hardened mortars is little influenced by the concentration of the commercial 25 mg/L Ag solution, the values varying by approximately ±1% compared to the apparent density of standard mortar. The greatest variation is noted in the case of recipe IV, in which 50% of water has been replaced with the commercial 25 mg/L Ag solution (density is 1.18% higher compared to the density of the standard recipe). From the point of view of apparent density, all mortars belong to the category of heavy mortars.

Regarding *adhesion to the substrate,* recipe R IV mortar is the best, its adhesion being similar to that of standard mortar.

The flexural strength value of mortars developed with the commercial 25 mg/L Ag solution increases compared to that of standard mortar. The increase manifests with the increase in the amount of colloidal water in recipes with up to 50% commercial 25 mg/L Ag solution, in which the flexural strength is the highest, i.e. 7.8 N/mm^2 compared to 6.9 N/mm^2 for standard mortar. In the case of the 70% commercial 25 mg/L Ag solution, there is a decrease in flexural strength compared to the mentioned recipes, but this remains higher than the flexural strength of standard mortar.

The same phenomenon is seen in the case of compressive strength. The best compressive strength is that of mortar with a 50% addition of commercial 25 mg/L Ag solution, having a value of 43.7 N/mm^2, higher than that of the standard recipe, which is 42.7 N/mm^2.

The determination of *water absorption by capillarity* highlights the following observations: (*i*) the coefficient of water absorption by capillarity of standard mortar is 0.16 Kg/(m^2·min$^{0.5}$), which places this mortar in class W2; (*ii*) in the case of recipes II, III and IV, with the increase in the proportion of water replaced by the commercial 25 mg/L Ag solution, the value of the coefficient of water absorption by capillarity decreases compared to standard mortar; these mortars fall in class W2; and (*iii*) the coefficient of water absorption by capillarity in the case of recipe V increases compared to the coefficient of recipes II, III and IV, but this fits in class W2.

Given that the studied mortars have less than 1% by mass or volume of homogeneously distributed organic material, they are classified as fire reaction class A1, without requiring testing.

Antibacterial effect of mortar specimens

The antibacterial efficacy of standard mortar (without colloidal silver) and mortars containing colloidal silver in various concentrations according to Table 3 is presented in Table 2.

Table 2. Effect of colloidal silver in mortars on the survival of four bacteria after 24h incubation at 37°C

	Escherichia coli	***Pseudomonas aeruginosa***	***Enterococcus faecalis***	***Staphylococcus aureus***
R I	100	100	100	100
R II	0	80	85	25
R III	0	25	35	0
R IV	0	5	15	0
R V	0	0	0	0

As expected, the standard mortar R I has no inhibitory effect on the investigated bacteria. Already after 24 h, the lowest concentration of colloidal silver in the mortar (R II) was effective against *Escherichia coli* bacteria. In all cases, a higher colloidal silver content (R V) caused 100% inhibition of bacterial growth. The antibacterial efficacy of colloidal silver in mortars demonstrated in our study is consistent with previous studies demonstrating the antimicrobial effect of silver ions [11, 12], which makes them promising for combating the growth of bacteria on interior or exterior walls.

However, these results have to be tested under the specific conditions (moisture, temperature, surface, etc.) to which these materials would be exposed in practical use (hospitals, nursing homes, schools, food preparation and storage areas, etc.); probably a mixture with various organic biocides could show some utility and interesting results.

Silver release

The study of silver release was performed in 1 culture medium, DMEM, and ultrapure water at different time intervals (1, 3, 6, 24 and 48 h). The amounts of silver released at 48 h were comparable to those measured at 24 h and consequently only the values obtained at 24 h were taken into account for this study. The obtained results (Figure 1) are in agreement with published literature data: the leaching solution has a significant effect on the level of leaching [12, 13]. As expected, for the mortar specimen R I, no silver release was observed (<0.01 mg/l). The level of leaching was much higher for specimens that were immersed in Dulbecco's modified eagle's medium (DMEM) compared to specimens immersed in ultrapure water (UW). Furthermore, in both releasing solutions, higher amounts of colloidal silver in the mortar specimens resulted in increased releasing ions: R I > R II >R III > R IV > R V.

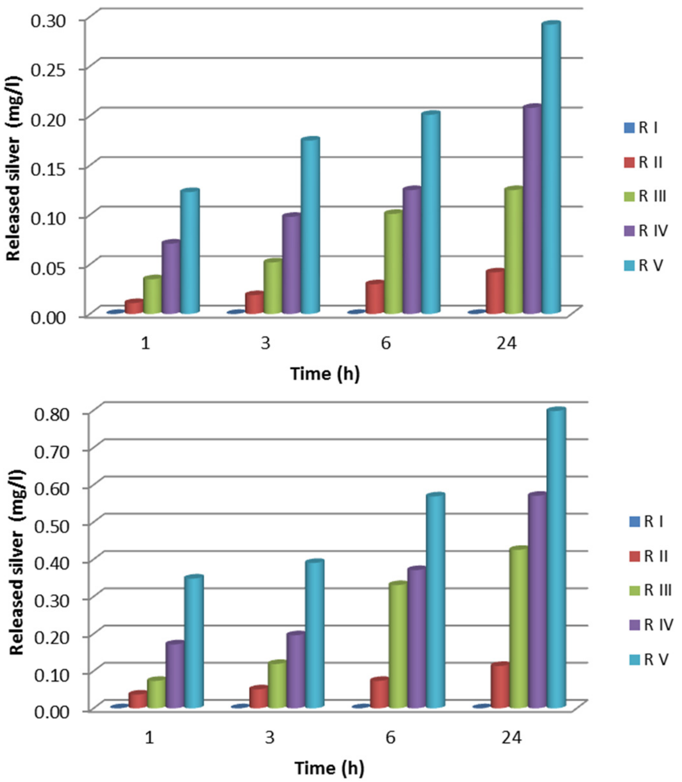

Figure 1. Silver released (mg/L) into UW (above) and DMEM (below) from mortar specimens containing colloidal silver

CONCLUSIONS

The study demonstrates the possibility to develop plastering mortars with good antibacterial properties by introducing colloidal silver instead of the water of mortar recipes. At 28 days, all mortars with colloidal silver solution addition have superior physical-mechanical characteristics and good antibacterial properties compared to the standard recipe.

It can be seen that there is an upper limit of the commercial 25 mg/L Ag solution concentration up to which these characteristics increase, after which a slight decrease occurs. From the point of view of physical-mechanical characteristics, the optimal recipe is that of mortar with 50% commercial 25 mg/L Ag solution, *i.e.* recipe R IV. According to standards in force, depending on compressive strength at 28 days, mortar falls in class CSIV and class W2 in terms of water absorption by capillarity. Under this framework, the obtained mortars can be used for the improvement of hygiene and sanitary conditions in a variety of environments such as hospitals, nursing homes, schools, food preparation and storage areas, etc.

EXPERIMENTAL SECTION

Preparation of mortar specimens

The starting materials used in this work were ordinary Portland cement 42.5, commercial silver colloidal solution (25 mg/L) introduced in various percentages in the water of the standard recipe, and (0-2) mm sand composed of four fractions [(0-0.16) mm – 150 g; (0.16-0.50) mm – 300 g; (0.5-1.00) mm – 450 g and (1.00-2.00) mm – 450 g)]. The aqueous Ag colloidal solution was purchased from a local pharmacy.

The study was performed on 5 plastering mortar recipes (Table 3). For standard mortar (recipe 1), a 1:3 binder-sand ratio was chosen, with the recipe composed of 450 g Portland cement 42.5, 1350 g sand and 225 cm^3 water. For the silver solution test samples (recipes 2-5), water in the standard mortar recipe was replaced in a 10%, 30%, 50% and 70% proportion with a commercial 25 mg/L Ag solution.

Table 3. Mortar recipes

Recipe	Cement	Water	Commercial 25 mg/L Ag solution		Sand 0-2 mm
	[g]	[mL]	[%]	[mL]	[g]
R I	450	225	-	-	1350
R II	450	202.5	10	22.5	1350
R III	450	157.5	30	67.5	1350
R IV	450	112.5	50	112.5	1350
R V	450	67.5	70	157.5	1350

Physical-mechanical properties of mortar specimens

The mortars (Figure 2) were prepared according to SR EN 1015-2:2001 [14], the raw materials were weighed, homogenized in dry state and then mixed with water (R I) or with water and the commercial 25 mg/L Ag solution (R II – R V) in the mixer.

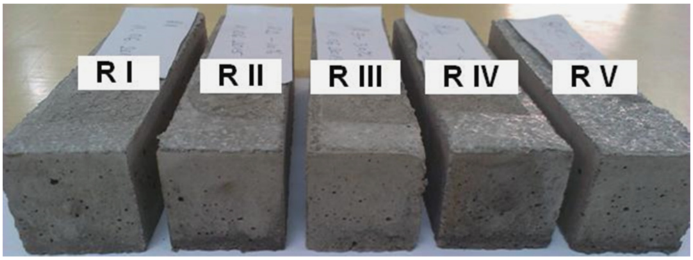

Figure 2. Mortar test samples (recipes I–V)

The consistency of standard mortar was determined with the flow table (Tecnotest, Italy), in conformity with SR EN 1015-3:2001 [15]. According to SR EN 1015-2:2001 [16], for fresh mortar with an apparent density higher than 1200 kg/m^3, the flow value is 175 ± 10 mm.

On the 16 cm x 4 cm x 4 cm prismatic test samples, cast and stored for 28 days according to standards SR EN 998-1:2011 [17], SR EN 1015-2:2001 [18], SR EN 1015-11:2002 [19], the following physical-mechanical determinations were performed:

- *the apparent density of hardened mortar* according to SR EN 1015-10:2002 [18];
- *the mechanical flexural and compressive strengths* according to SR EN 1015-11:2002 [17]. The flexural strength was tested with the automatic flexural tensile tester (Controls, Italy), and the compressive strength with the 250 KN hydraulic press (Tecnotest, Italy).

- *the water absorption coefficient due to capillary action* according to SR EN 1015-18:2003 [19].
- *the adhesion of hardened mortar to the substrate* according to SR EN 1015-12:2001 [20], with the pull-off tester 58-C0215/T (Controls, Italy).

Evaluation of antibacterial activity

The 28-day-old-mortar samples R1-R5 were cut into small 20 x 20 x 1 mm^3 prisms and dried at 80°C until a constant weight was obtained. All manipulations for this study were performed in a laminar flow hood.

Two Gram-negative bacteria strains (G^+): *Escherichia coli* ATCC 25922 and *Pseudomonas aeruginosa* ATCC 27853, and 2 Gram-positive bacteria strains (G^-): *Enterococcus faecalis* ATCC 29212 and *Staphylococcus aureus* ATCC 29213 were used. The bacteria were used at a density of 10^6 CFU/mL, grown in nutrient agar no. 2 (Sigma-Aldrich, Germany) and incubated for 24-48 h at 37°C, in darkness. The tested specimens were placed on Petri dishes with the culture medium and inoculated with 100 μL bacterial suspension, using a sterile pipette. The inhibition zones around the wells were measured (in mm) with a millimeter ruler. The Petri dishes were incubated again and measured after 48 h. Since the diameters of the microbial growth inhibition zones was comparable to that measured at 24 h, only the values obtained at 24 h were considered for this study. These experiments were repeated at least four times with similar results.

Measurements of silver release

The mortar prisms (~15 g) were submerged in 50 mL sterile centrifuge tubes containing 20 mL sterile DMEM D5796 (Sigma-Aldrich, Germany) or ultrapure water for 1, 3, 6, 24 and 48 h. At each point, the solid part was separated from the rest of the solution through centrifugation at 4000 rpm for 30 min. Solutions were collected and digested with 4 mL HNO_3 and 2 mL H_2O_2 in a closed-vessel microwave system Berghof MWS-3+ with temperature control mode (Berghof, Germany). Samples were digested according to the following 5-step program: (*i*) 5 min at 180°C and 80% power, ramp 3 min; (*ii*) 10 min at 220°C and 90% power, ramp 5 min; (*iii*-*v*) 5 min at 100°C and 10% power, ramp 1 min. After cooling down to room temperature, the resulting colorless solutions were diluted to 25 mL with deionized water. The silver content was determined by ICP-OES (OPTIMA 5300 DV, Perkin Elmer, USA). Three independent replicates were analyzed for each sample.

Calibration standards were prepared by dilution, with 2% (v/v) HNO3, of the stock ICP multielement standard solution IV (Merck, Darmstadt, Germany) 1000 mg/L. All reagents (HNO_3 65%, H_2O_2 30%) were of analytical grade and were purchased from Merck, Darmstadt, Germany. All dilutions were prepared using deionized water (18.2 MΩ/cm) obtained from a Millipore Direct-Q3 UV Ultrapure water system (Millipore, Molsheim, France). All PTFE and glass vessels were soaked in 10% (v/v) HNO_3 overnight or longer and rinsed with Milli-Q water prior to use.

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