Combined Effect of Superabsorbent Polymers and Cellulose Fibers on Functional Performance of Plasters

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Abstract: Plaster has, from ancient times, been used as a decorative material. However, the advances in materials engineering such as thermal and moisture control provide new opportunities. Superabsorbent polymers (SAPs) have been found to possess passive moisture control that may find utilization in modern buildings. However, the main drawback is associated with a limited number of applicable SAPs due to mechanical strength loss. In this regard, concurrent utilization of cellulose fibers may provide additional benefits linked with the reinforcing of plaster structure and preservation of superior hygric properties. In this regard, this study investigates the combined effect of SAP and cellulose fibers on the material properties of cement-lime plaster in terms of its mechanic, thermal, and hygric properties. To access the capability of such modified plasters to control the interior moisture fluctuations, the moisture buffering value is determined. Obtained results show the effect of both applied admixtures on material performance, whilst the synergic effect was most obvious for humidity control accessed through the moisture buffer coefficient.

Keywords: cellulose fiber; superabsorbent polymer; synergy; reinforcement; mechanical performance; moisture buffering

1. Introduction

Recent developments in materials science brings new opportunities for the modification of traditional building materials, including finishing plaster [1]. While the role of this material was predominantly decorative in the past, a tailored modification may extend the functionality of this material [2–5]. This issue was subjected to several investigations aiming at improvements in terms of hygrothermal performance, durability, protective coatings, and service life. [6–9]. Due to recent challenges, the passive maintenance of indoor climates poses a major issue for new building material design, plasters included [10]. In particular, the preservation of the quality of the indoor environment has attracted intensive attention due to increased awareness of sick building syndrome and extensive energy consumption [11]. In this sense, the optimal range for indoor relative humidity lies between 35–65% [12,13]. The particular importance of indoor moisture level is shared by issues such as furniture durability and health issues. To be more specific, plasters with lightweight aggregate, silica gel, minerals, and charcoals have been designed and consequently studied to provide satisfactory indoor humidity control [14–17]. In this regard, the effect of total pore volume, shape, and pore size distribution were found to be promising research lines towards the passive modulation of indoor relative humidity [18]. Specifically, the application of perlite or vermiculite increased the pore volume by about 20%, thus increasing moisture buffering [19,20]. In this regard, the application of sodium olefine-sulphonate and superabsorbent polymers (SAPs) achieved very beneficial results in terms of shifting the moisture buffering value [21,22]. The work of Fořt et al. [23] exhibited the great potential of SAPs in the mitigation of humidity peaks during diurnal loading. As revealed, even relatively small dosages of SAPs provide substantial improvements in moisture buffering performance thanks to their great swelling capabilities [24]. On the other hand, several
drawbacks are associated with the limited workability of the fresh mixture, and a loss of mechanical strength has been revealed [25]. These findings may be viewed as a major barrier for the broader application of SAPs in conventional building materials despite the benefits linked with passive moisture control [26].

Taking into account the mechanical strength issues associated with the utilization of SAPs in plasters, the application of reinforcing fibers can be viewed as a possible solution [27]. During recent decades, various types of fibers have been studied with partial success, including with regard to their mechanical strength and thermal and hygric parameters in particular [4,28,29]. Apart from synthetic fibers such as polypropylene, glass, and carbon fibers, a wide range of natural fibers have been investigated as environmentally friendly reinforcers. For example, the application of hemp fibers provides substantial improvements in adhesion and flexural strength [4]. The work of Lee et al. [14] described that fiber incorporation has a substantial effect on material porosity, and consequently may result in the modification of water absorption capability, as well as water vapor storage properties. However, the increase in apparent porosity is accompanied by a deterioration in mechanical strength in cases where the proportion of cellulose fibers is too high [30,31].

In this regard, this study reveals the potential of the application of cellulose fibers applied together with SAP admixtures. Cellulose fibers have, in the past, been found to be a suitable component for the development of multifunctional plasters with good thermal insulation properties as well as the ability to control the moisture level in building interiors. The effect of both applied additives is described from the point of view of the thermal, hygric, and mechanical properties to provide an understanding of the humidity control capability of such modified plasters.

2. Experimental

2.1. Used Materials

The superabsorbent polymer Hydropam (Evonic Degussa International, Krefeld, Germany) was used to modify cement-lime plaster composed of cement, lime, and sand at a respective ratio of 1:1:5. The particle size of the applied SAPs (see Figure 1) ranged from 200 µm to 1000 µm, with d50 = 433 µm determined by the laser diffraction device Bettersizer S3 Plus (Liaoning, China). According to the data provided by the SAP manufacturer, Hydropam is composed of sodium salt and acrylamide/acrylic acid copolymer with a density of 690 kg/m³.

![Particle size distribution of SAP](image)

Figure 1. Particle size distribution of SAP.

The cellulose fibers came from the CIUR a.s. Company, (Brandýš and Labem, Czech Republic). The fiber lengths ranged between 2 and 4 mm. Information about the used cel-
Cellulose was obtained by Scanning Electron Microscopy (SEM). The analysis was performed by using an electron microscope JSM 6510 LV-Jeol (Tokyo, Japan) with a magnification of about 5–300,000. The SEM detail of applied fibers is shown in Figure 2. Hydrated lime CL 90-S (Mokrá Plant, Carmeuse Czech Republic) was applied together with cement CEM I 32.5 R (Cemex, Czech Republic) as base binders for the designed plasters. Particular plaster mixtures were derived from previous experiments aimed at the investigation of the effect of SAP admixture only [23]. The cellulose fibers were applied in 1, 2, and 3% dosages and compared to reference mixture and plaster without SAP admixture. The modifications of studied plasters are described in Table 1.

![SEM image of cellulose fiber.](image)

**Figure 2.** SEM image of cellulose fiber.

**Table 1.** Composition of the studied plasters.

| Mixture | SAP (%) | Cellulose (%) |
|---------|---------|---------------|
| PR      | -       | -             |
| PS0.5C1 | 0.5     | 1             |
| PS0.5C2 | 0.5     | 2             |
| PS0.5C3 | 0.5     | 3             |
| PS1C1   | 1       | 1             |
| PS1C2   | 1       | 2             |
| PS1C3   | 1       | 3             |
| PS1.5C1 | 1.5     | 1             |
| PS1.5C2 | 1.5     | 2             |
| PS1.5C3 | 1.5     | 3             |

All prepared samples were cured in a highly humid environment (RH = 95%) after sample demolding for 24 h. Such treated samples were dried in an electric oven at 65 °C until steady-state mass was reached.
2.2. Determination Methods

The bulk density, matrix density, and open porosity were determined to access the main differences in the basic material properties. The results of the dry bulk density were obtained by weighing the dried samples and volume determined by using a digital caliper. The matrix density was given by a helium pycnometer Pycnomatic ATC (Thermo Scientific, Waltham, Massachusetts, USA) device. Consequently, the total open porosity was calculated from the results of the bulk- and matrix density [32].

A hydraulic testing device, VEB WPM Leipzig, with a stiff loading frame and a maximal capacity of 3000 kN was used for the determination of the compressive and flexural strength of plaster samples after 28 days of curing at a highly humid environment on prismatic samples having dimensions of 160 mm × 40 mm × 40 mm. The compressive strength was measured on the leftover prisms broken during the bending test (loading area was 40 × 40 mm²) [33].

Regarding the thermal properties of studied plasters, the portable instrument ISOMET 21114 (Applied Precision, Raˇča, Slovakia) was employed for the determination of thermal conductivity and thermal diffusivity [34]. The measurement range of the thermal conductivity was from 0.015 to 6 W/mK, with an accuracy of 5% of reading and reproducibility of about 3% of reading in the temperature range from 0 to 40 °C.

The water vapor transmission parameters of plaster samples were studied by the dry-cup method arrangement with additional temperature/relative humidity probes placed above and under the sample for better accuracy [35]. Five circular samples with a thickness (diameter of 100 mm) of about 30 mm were sealed into aluminum cups filled with silica gel and placed in the climatic chamber. Here, conditions of about 21 °C and 50%RH were set for the whole experiment. The cups were periodically weighted to obtain the mass increase in time for the consequent calculation of water vapor transmission properties.

The liquid water transport was calculated through the water absorption coefficient derived from the following equation [36]:

\[ i = A \cdot t^{1/2} \]  

(1)

where \( i \) (kg/m²) is the cumulative mass of water, \( A \) (kg/m²s⁴) is the water absorption coefficient, and \( t \) is the time (s).

The moisture diffusivity was obtained from:

\[ \kappa_{app} = \left( \frac{A}{w_{sat} - w_0} \right)^2 \]  

(2)

where \( w_{sat} \) (kg·m⁻³) is the saturated moisture content and \( w_0 \) (kg·m⁻³) is the initial moisture content.

The ability of the samples to absorb and release water vapor was measured by determining the moisture buffering [37]. A moisture buffering test was carried out at isothermal conditions (21 °C) using a 30%/70% humidity scheme as depicted in Figure 3. The material response of particular samples was monitored by a very sensitive DVS-Advantage device for 120 h (5 = complete cycles). Consequently, the moisture buffer value was obtained from the following formula:

\[ MBV = \frac{\Delta m}{S \cdot \Delta \%RH} \]  

(3)

where \( \Delta m \) is the weight variation, \( S \) is the exposed surface of the sample, and \( \Delta \%RH \) is the change in relative humidity level—between 30% (8 h) and 70% (16 h).
3. Results and Discussion

The effect of separately incorporated SAPs and cellulose fibers have been studied in the past; however, their coeval impact on material properties has not yet been described. The results of the bulk and matrix density measurement completed by calculated values of the total open porosity are shown in Table 2. The concurrent application of both admixtures resulted in substantial modification to the material structure. Despite the preservation of the matrix density, the bulk density was significantly reduced. Specifically, the matrix density slightly varied around 2550 kg/m³, and the bulk density ranged from 1588 kg/m³ to 1185 kg/m³. As given by obtained results of bulk density and material porosity, a more critical impact is associated with the application of SAPs over cellulose fibers even at very low dosages. In this regard, the increased cellulose dosages induced only minor changes. The most obvious modification was observed for mixtures having 1.5 wt.% of SAP where some dissonance between both admixtures also revealed substantial deterioration in line with the increased content of applied cellulose fibers. On the other hand, for 0.5 wt.% SAP, the incorporation of 1 and 2 wt.% of cellulose did not cause any distinct modifications. The total open porosity varied in a similar manner. The lowest porosity was reached by the reference mix and the highest by mixture with 1.5% SAP and 3% cellulose. Such results comply with previously published research papers [25–27,29] that studied these admixtures separately. The revealed modification correlates with characteristics of used admixtures by meaning their size, swelling capability (SAP), amount of used water, and powder density [38]. The substantial shift in material porosity also depends on the amount of the used batch water that must be adjusted during the preparation of the mixture as a result of decreased workability of fresh mixtures. Similar issues were noted for concrete mixtures designed with SAP; however, the application of superplasticizers in plasters is rather sporadic compared to concrete [39]. Nevertheless, this approach should be considered in follow-up work.

![Figure 3. Time-dependent relative humidity variations during MBV experiment.](image-url)
Table 2. Basic material properties of studied plasters.

| Mixture    | Bulk Density (kg/m$^3$) | Matrix Density (kg/m$^3$) | Total Open Porosity (-) |
|------------|-------------------------|---------------------------|-------------------------|
| PR         | 1588 ± 25               | 2556 ± 11                 | 0.38                    |
| PS0.5C1    | 1546 ± 31               | 2543 ± 13                 | 0.39                    |
| PS0.5C2    | 1540 ± 26               | 2549 ± 11                 | 0.40                    |
| PS0.5C3    | 1433 ± 27               | 2561 ± 10                 | 0.44                    |
| PS1C1      | 1463 ± 30               | 2540 ± 15                 | 0.42                    |
| PS1C2      | 1408 ± 25               | 2539 ± 11                 | 0.45                    |
| PS1C3      | 1372 ± 19               | 2558 ± 12                 | 0.46                    |
| PS1.5C1    | 1377 ± 42               | 2562 ± 10                 | 0.46                    |
| PS1.5C2    | 1269 ± 27               | 2541 ± 14                 | 0.50                    |
| PS1.5C3    | 1185 ± 23               | 2550 ± 12                 | 0.54                    |

The achieved mechanical parameters of designed plasters with various SAPs and cellulose admixtures are plotted in Figure 4. As one can see, the amount of applied admixtures caused significant changes in the mechanical performance. In particular, two opposite effects can be distinguished. While the lower dosages of both admixtures had a beneficial effect on materials strength, the further increase resulted in substantial worsening in both compressive and flexural strength. Generally, the SAP admixture showed more pronounced changes despite the lower dosages compared to cellulose. Considering the material response, the application of up to 1% of SAP or 2% of cellulose caused an improvement in the mechanical strength if used together with the lowest dosage of the second admixture. Other cases resulted in substantial material deterioration that may pose a substantial barrier for the material application. To be more specific, a plaster mixture with 3% of cellulose and 1.5% of SAP reduced the compressive strength by more than 50%, even more than 67% in the case of flexural strength. In other words, the advantages of both materials can be used only in a relatively narrow interval of their amount, and exceeding the optimal dose causes significant degradation of the plaster in terms of their useful properties [40]. Such material response is probably inflicted by limited material workability and a shift in material porosity. On the other hand, the elucidation of the material strengthening lies rather in improved internal curing secured by SAPs and the reinforcing effect of applied cellulose fibers. Variations in the reinforcement effect of applied cellulose refer to limited bonds formed between the material matrix and applied cellulose fibers. The possible solution for the strength improvement can be found in coating treatment to increase the interfacial bonding between cementitious matrix and used fibers [41]. The finding of the threshold values for dosage of both admixtures, therefore, poses a very important task for the sufficient design of such modified plasters.

The effect of applied admixtures on the thermal parameters of plaster by measure of the thermal conductivity via the impulse method is shown in Figure 5. Given results represent average values obtained from 5 independent measurements. As is visible, the thermal conductivity was dropped in line with the decreased porosity from the initial 0.55 W/(mK) to 0.43 W/(mK). The utilization of both admixtures resulted in a relatively small improvement; however, according to similar research performed by Gueardia et al. [42] and Lee et al. [43], only minor changes were expected. In particular, the increased water absorption capability of SAP diminished the beneficial effect of increased open porosity on thermal resistance due to the formation of highly conductive hydrogels having higher thermal conductivity. The influence of applying cellulose fibers was rather minor, which complies with findings revealed by Nindiyasari [44]. In this regard, the achieved modifications in the thermal conductivity properties are caused by two opposing phenomena: a reduction driven by the shift in the material porosity; and an increase caused by the presence of highly conductive hydrogels.
Since the motivation of the paper is driven by the effect of applied admixtures on water transport properties, free water uptake and moisture diffusivity experiments were carried out (see results in Table 3). Looking at the presented data, one can see a slight increase in water uptake capability influenced by both applied admixtures. While the work of Senff et al. [40], did not describe any notable changes in hygric performance due to utilization of hydrophobized cellulose fibers, results depicted in the present study pointed to an increase in both monitored parameters. This effect is associated with increased material porosity, but also with the hydrophilic nature of both applied admixtures [45]. Therefore, the effect of pure cellulose fibers can be derived from the research of Nindiyasari et al. [44] or Dalmay et al. [46]. On top of that, the swelling capability of SAP further increased the water uptake in accordance with Vieira et al. [22], who described the dependency between the amount of applied SAP and water absorption. Beyond the work of You et al. [17], the substantial swelling characteristics of SAPs were almost equal to the effect of cellulose fibers, thus the moisture diffusivity was modified by both admixtures to a similar extent.
Specifically, the difference in incorporation between 1% and 3% of cellulose fibers caused variations of around 15%, and the effect between 0.5% and 1.5% of SAP was quite similar.

Table 3. Hygroic properties of studied plasters.

| Mixture     | A (kg/m²s¹/²) | K (m²/s)   |
|-------------|---------------|------------|
| PR          | 0.164         | 4.94 × 10⁻⁷|
| PS0.5C1     | 0.166         | 5.01 × 10⁻⁷|
| PS0.5C2     | 0.174         | 5.22 × 10⁻⁷|
| PS0.5C3     | 0.181         | 5.66 × 10⁻⁷|
| PS1C1       | 0.176         | 5.25 × 10⁻⁷|
| PS1C2       | 0.184         | 5.98 × 10⁻⁷|
| PS1C3       | 0.192         | 6.13 × 10⁻⁷|
| PS1.5C1     | 0.194         | 6.22 × 10⁻⁷|
| PS1.5C2     | 0.204         | 6.63 × 10⁻⁷|
| PS1.5C3     | 0.215         | 6.89 × 10⁻⁷|

The water vapor resistance factor of studied plasters is shown in Table 4. As shown, the water vapor resistance factor gradually decreased from an initial 15.33 to 11.06. The achieved trend in obtained values clearly reflected the amount of both applied admixtures as reported for other material parameters. On the other hand, our results did not decrease as sharply as those of Goncalves et al. [21]. The explanation can be found in the limited modifications of the total open porosity and different characteristics of applied SAPs [47]. The variations in chemical composition and particle size may lead to significant changes in the swelling capability driven by dissolute ions in water solution as reported in Fort et al. [48]. As the data sheets provided by the SAP producers describe, the real SAP sorption performance should be verified by additional experiments taking into account the ions concentration in the used solution. As reported by Snoeck et al. [49], this parameter may vary significantly, thus reflecting the expected results. All obtained material parameters were reflected in changes in the moisture buffering potential as accessed in Figure 6. The lowest hygroscopic capacity, as well as the potential for passive moisture moderation, was obtained for the reference plaster that can be classified as moderate (the typical classification of the majority of plasters according to the Nordtest method [50]). In contrast, the best performance was achieved by PS1.5C3 plasters, having almost three-times better capability for moisture moderation. In this regard, all plasters with 1.5% SAP dosages represent excellent moisture buffering performance. This effect is assured by the combined effect of the increased material porosity and strong water absorption capacity of SAP particles [50].

Table 4. Water vapor resistance factor.

| Mixture     | µ (-)   |
|-------------|---------|
| PR          | 15.33   |
| PS0.5C1     | 14.76   |
| PS0.5C2     | 14.38   |
| PS0.5C3     | 13.76   |
| PS1C1       | 14.43   |
| PS1C2       | 13.98   |
| PS1C3       | 13.66   |
| PS1.5C1     | 12.45   |
| PS1.5C2     | 11.79   |
| PS1.5C3     | 11.06   |
Figure 6. Moisture buffer value of modified plasters.

The correlation between the flexural strength of studied plasters and moisture buffering is illustrated in Figure 7. Samples with 0.5% SAP and up to 2% cellulose content exhibited the synergic effect in terms of improvements in both monitored parameters. On the other hand, other mixtures proved only the capability to improve the moisture buffering while the mechanical strength was reduced significantly. Interestingly, a very similar mechanical performance was obtained for mixtures PS1C3 and PS1.5C1, although the moisture buffering of plasters with higher SAP content provides more favorable moisture buffering. In other words, the effect of SAPs dominates over cellulose in the case of hygric properties. Concurrently, even such a small variation in dosage may cause a substantial deterioration in material microstructure. The observed phenomenon can be assigned to the huge impact of SAPs on fresh mixture workability. Apart from the increased material porosity, the effect of SAP, as well as cellulose, on setting time should be taken into account [22,27]. As reported in the research of Barnat-Hunek et al. [38] or Senff et al. [40], the incorporation of a hydrophilic admixture brings with it several adverse effects linked to water/binder ratio adjustments and changes in the material porosity.

Figure 7. Correlation between moisture buffering and flexural strength.
4. Conclusions
The results of the experimental analysis of the combined effect of cellulose fibers and the superabsorbent polymer were described in this work. To provide a more detailed analysis of material performance, the mechanical, hygric, and thermal properties were determined.

First, the application of cellulose fibers did not provide any notable strengthening of the plaster, only minor improvement was noted for lower dosages. Obtained results point to the necessity of additional treatment of fibers that may improve the reinforcing performance [41]. In contrast, the application of untreated cellulose fibers provides side benefits such as enhanced hygric properties. To find the rational compromise, mixture PS1C1 provides the best overall score considering the relevant increase in both selected indicators. Mixture PS1.5C1 is a material that shows satisfactory mechanical performance while the moisture buffering value was double that of the reference value. Moreover, this material exhibit significantly higher moisture buffering at the same mechanical strength level. The mixtures modified by 0.5% SAP did not reveal any significant improvements in terms of moisture properties, while also shifting the mechanical performance. The synergic effect was most obvious for humidity control performance accessed through the moisture buffer coefficient. In this regard, follow-up research aimed at in situ investigation of material performance should be carried out together with the energy balance calculation.

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