Evaluating the Effect of Methyl Cellulose on Hardened State Properties in Selective Cement Activation

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Abstract. Compared with other 3D printing techniques used in construction, particle bed 3D printing by Selective Cement Activation (SCA) offers unique possibilities regarding the fabrication of high-resolution and freeform components. The printed geometry is determined by the fluid distribution in the particle bed, consisting of a dry mixture of fine aggregates and cement. The fluid intrusion behaviour and thus the shape accuracy of the printed object can be actively controlled by adding additives such as methyl cellulose (MC) to the particle bed. In this research the goal is to fundamentally understand the effect of MC varying in degree of polymerization (DOP), size and dosage on hardened state properties of SCA printed objects. Therefore, dissolution experiments of MC in the rheometer, capillary induced fluid intrusion into the particle bed as well as printing experiments are conducted. As hardened state properties of printed components, geometric precision, density as well as compressive strength are assessed.

It was found that MC granules in the particle bed increases the geometric precision and the mechanical performance of SCA objects to a certain extent. This is explained by a modified fluid intrusion behaviour in the particle bed. Here, the effect of the DOP is low compared to the effect of the particle size of the dry granules and the amount of MC. When using too much MC, the geometric precision is not further increased while the mechanical performance even decreases, which is attributed to an increase in porosity in the printed specimen.

Keywords: Additive manufacturing in construction · Selective Cement Activation · Particle bed binding · Particle bed 3D printing · Methyl cellulose · Geometric precision

1 Introduction

The construction industry has a large share on climate change as it is the largest consumer of resources, with 50% of raw materials processed and consumed worldwide being used in construction [1]. One very promising way to reduce the environmental impact of concrete components is to use less material by designing the structure more efficient. In this regard, additive manufacturing is a promising technique to meet these requirements.

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Structural optimization and usage of additive manufacturing techniques can reduce the required material input up to 70% [2]. Due to its very high resolution and the possibility to produce freeform structures, the optimized design is in particular achievable with particle bed binding. Particle bed binding encompasses a process, where the material of a particle bed is selectively bound by a liquid phase.

In this study, particle bed 3D printing by Selective Cement Activation (SCA) is investigated. In SCA, the particle bed consists of a dry mixture of fine aggregates and cement. The cement is locally activated by jetting water onto the particles, thereby creating a cement paste matrix around the aggregate particles. Then, the subsequent hardening of the matrix occurs locally. Finally, the non-bonded particles are removed in a de-powdering process. To fully scoop the possibilities of SCA, it is necessary to control fluid intrusion into the particle bed since this is key to enable high geometric precision and mechanical perfomance. In order to improve geometric precision, additives such as methyl cellulose (MC) may be used [3]. In the investigations at hand, it is the goal to fundamentally investigate the effect of MC in SCA on the hardened state properties of SCA-objects. Here, the effect of i) degree of polymerization (DOP), ii) particle size of the dry MC granule and iii) amount of MC in SCA is studied. In order to fundamentally understand the obtained results, three levels of experiments are envisaged: Investigation of dissolution rate of MC in liquid in the rheometer, investigation of capillary intrusion of water in particle beds containing MC granules and finally production and testing of hardened state properties of SCA specimen.

2 Materials and Methods

2.1 Material

The dry particle bed in the printer consists of a mix of aggregates (quartz sand, $d_{50} = 298\ \mu m$), rapid setting cement (Portland cement with Mayenite, $d_{50} = 9\ \mu m$) and MC granules. The volumetric ratio of aggregates to cement in the particle bed was 60/40. Water is used as an activator. As MC, commercially available water soluble, nonionic granules are used. Here, the MC is a methyl hydroxyethyl cellulose with various DOPs and sizes (SE Tylose):

- Low (MH 300) and high (MH 100.000) DOP
- Coarse (c, $<300\ \mu m$), medium (m, $<180\ \mu m$) and fine (f, $<100\ \mu m$) MC granules.

The DOP gives an information about the average number of anhydroglucose units per cellulose ether molecule and is correlated to the viscosity grade of a fluid with this MC [4]. The size of the granules is varied by the production side and is achieved via sieving. The nomenclature of the experiments is chosen according to the DOP and the fineness of the MC granules, e.g. MH 300_m.

2.2 Methods

Dissolution Rate of MC in the Rheometer. Water is filled in a rheometer cup and sheared at 500 rpm with a 6 bladed vane (Anton Paar MCR 502) for 300 s and the resulting torque is recorded over time. Within the first 5 s of the experiment, 5 g of MC is added to the cup. The increase in rotational torque, i.e. viscosity, starts after sufficient MC has dissolved to form a polymer structure in the fluid. Thus, the point in time of the initial torque increase is considered as a parameter to characterize the dissolution rate of the MC.

Liquid Penetration into Particle Beds. In order to experimentally transfer the effect of MC dissolved in water to the MC granules distributed in the particle bed, the following setup was used to quantify capillary penetration of water into the particle bed from bottom to top: Aggregates mixed with 0.25% and 0.5% MC granules as well as aggregates without MC as a reference are filled in a glass tube (diameter $d_i = 23.5$ mm). The bottom of the tube is covered with a lid with three holes of diameter $d = 6$ mm and filter paper. The glass tube is put into a water reservoir and the subsequent liquid penetration process over time is recorded.

Although, the boundary conditions of the experiment do not directly describe the conditions during particle bed 3D printing – where not an infinite but a locally very limited water volume is prevalent - the described experimental setup provides valuable information on capillary flow within the particle bed, which is assumed to be the dominating mechanisms for liquid intrusion in particle beds [5].

Production and Testing Hardened State Properties of SCA Samples. The density, the compressive strength as well as the geometric precision of 3D-printed specimens are investigated. For compressive strength tests, prisms with the dimensions $40 \times 40 \times 160$ mm^3 (W × H × L) were produced in the SCA-3D-printer (Progress Group). These specimens were oriented lengthwise along the travel direction of the print head. For geometric precision, cylinders ($d = 60$ mm, height: 40 mm) were produced.

The layers for the print job are applied with a height of 1.3 mm, which is then compacted to 1 mm layer height with a rotating roller. Then, water is applied with a discharge rate of 4 g/min in order to produce a w/c-ratio of 0.5. After excavation, the specimens are stored for 28 d at 20 °C/65% r. H. In order to ensure parallel planes for the mechanical testing, prisms were cut down to the precise size. The compressive strength of the specimen is measured in accordance with DIN EN 196-1.

To investigate the specimen's geometry, the cylinders are 3D scanned using a µCT. From the X-ray projections, the volumetric image is obtained which is used to determine a 3D-shape accuracy, namely the shape index S_{3D}, as also described in [3]. For this, the actually printed volume V_{act} [mm^3] is compared to the targeted volume V_{tar} [mm^3]. Thus, the shape index is determined ($S_{3D} = V_{act}/V_{tar}$). With a shape index of $S_{3D} = 1$, the printed geometry corresponds exactly with the target dimensions. The greater the difference between S_{3D} and 1, the lower the shape accuracy.

3 Results and Discussion

Dissolution Rate of MC in the Rheometer. In Fig. 1 the results of MC added to sheared water are shown. It is observed that a higher DOP of MC is accompanied by a higher torque increase in the rheometer, Fig. 1 left (compare MH100.000_m and

MH300_m). From theory, it is also expected that the increase in DOP also leads to a decrease in solubility of MC [4]. With regard to dissolution rate no significant effect of the DOP was found, as the time of initial torque increase of MC with high and low DOP are in the same order of magnitude, Fig. 1 right. Furthermore, it could be observed that the smaller the granules of MC, i.e. the higher the surface area, the shorter the time of initial torque increase, i.e. the higher the dissolution rate, Fig. 1 right. Hence, it is assumed that the granule size has a more distinct effect on liquid intrusion behaviour in particle beds than the DOP. In order to prove this assumption, the transferability of the results is studied in liquid penetration investigations on particle beds.

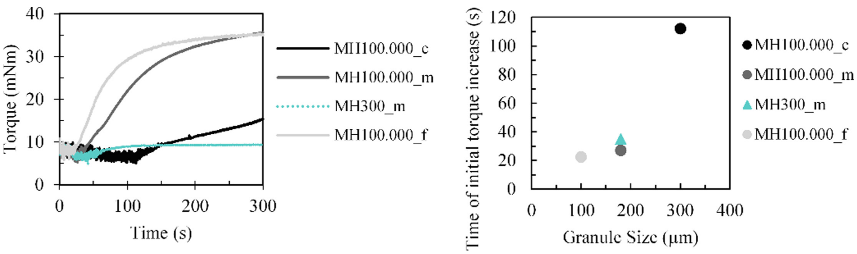

Fig. 1. Torque over time in the rheometer when MC is added to water (left); time until MC goes into solution, i.e. indicated by the point in time when an increase in torque is measured on the rheometer

Liquid Penetration into Particle Beds. Figure 2 left shows the effect of MC granules size and DOP on the liquid intrusion into the particle bed. It is evident, that the addition of MC granules in the particle bed has a severe effect on intrusion length and velocity. This can be seen by the reduced maximum intrusion length as well as the intrusion rate compared to the reference sample without MC, Fig. 2 left (empty vs. filled symbols). For capillary pressure driven liquid intrusion, which is assumed to be prevalent here, a square-root of time behaviour is expected [6]. However, the liquid intrusion behaviour into particle beds with MC deviates from a square root intrusion behaviour, which indicates (a) an alteration of the rheological properties of the intruding liquid and/or (b) a modification of the pore size and volume. When water reaches MC granules in the particle bed, it is partially and temporarily retained in swelling MC molecules. If we correlate the intrusion rate and length with the dissolution rate, we see that the higher the dissolution rate, the lower the intrusion rate and length. It is concluded, the smaller the MC granules, i.e. the higher the surface area having initial contact to water, the faster an effect is generated on the liquid intrusion length in the particle bed.

Furthermore, it is found that an increase in DOP of MC tends to decrease the intrusion velocity and length, compare dark grey circles with blue triangles in Fig. 2 left. This is explained by a higher dissolution rate of MC with a lower DOP and herewith higher "effectiveness" of the MC in the particle bed. However, this effect is inferior in the investigated range compared to the effect of the size of the MC granule, which shows a significant effect in liquid intrusion into the particle bed: the larger the granules, the smaller the surface area and herewith the lower the dissolution rate of MC, as it can be observed when comparing light grey to black circles in Fig. 2.

The described effects are becoming more distinct, when increasing the amount of MC from 0.25% to 0.5% in the particle bed (data for 0.5% not shown here).

Hardened State Properties of SCA Samples. Finally, the effects of the DOP and the size of the MC granules on the hardened properties are investigated. Therefore, two DOPs and two granule sizes are first examined with regard to geometry. We can show that the effect of DOP for 0.25% of MC in the particle bed is neglectable, as the shape index differs only slightly (S_{3D} = 1.35 and 1.33 for MH300_m and MH100.000_m, Fig. 2 right). Moreover, the distinct effect of the size of the granule was shown with the shape index for coarse (MH100.000_c, S_{3D} = 1.42) compared to medium sized granules (MH100.000_m, S_{3D} = 1.33).

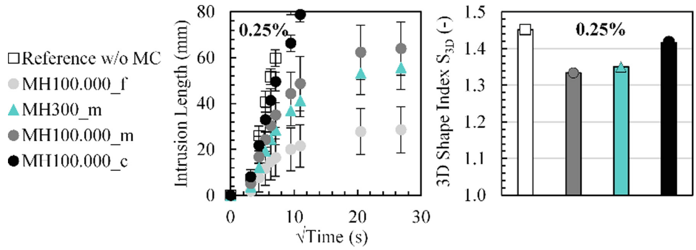

Fig. 2. Intrusion length over time in liquid intrusion experiments for MC (left) and 3D Shape Index for printed specimen (right) with a dosage of 0.25% MC in the particle bed

Finally, the effect of the amount of MC on hardened state properties, namely mechanical performance and geometry, are studied. With regard to geometric precision, an increase in MC decreases S_{3D}, i.e. increases the shape accuracy of printed objects, Fig. 3 left. For MC dosages up to 1.5 wt.-% a decrease in S_{3D} from 1.45 to 1.18 is observed. A further increase in MC does not lead to a further decrease in S_{3D}. An altered liquid penetration behavior in the particle bed containing MC may occur mainly due to the water retention in the MC inclusions, as it was also observed in the liquid intrusion experiments in the particle bed in the tubes. The more MC available, the more water is stored. However, this water retention is a time depending process and therefore S_{3D} does not decrease further, if a certain MC dosage threshold is exceeded.

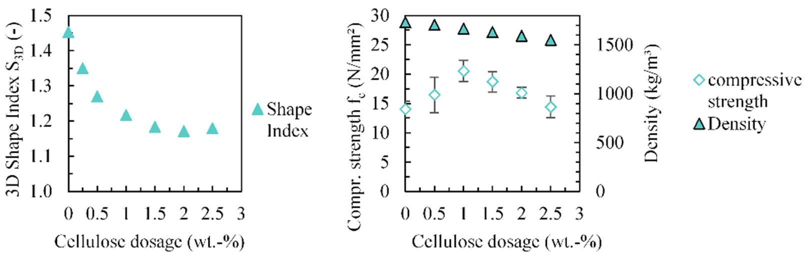

Fig. 3. Effect of metyl cellulose dosage on Shape Index S_{3D} (left) and on compressive strength and density (right) of printed objects with MH 300_m

The effect of MC on density and compressive strength is shown in Fig. 3 right. The addition of small amounts of MC, increases the compressive strength of the printed specimens from 14.0 MPa (0 wt.-% MC) up to 20.5 MPa (1.0 wt.-% MC). However, adding more MC reduces the compressive strength, as shown by the samples with 1.5 to 2.5 wt.%. The increase in MC is accompanied by a linear decrease in density of the printed elements (1.730 kg/m^3 for 0 wt.-% MC to 1.550 kg/m^3 for 2.5 wt.-% MC, triangles in Fig. 3 right). The increase in compressive strength at low MC dosages can be attributed to a temporary absorption and subsequent release of water, which may increase the degree of hydration in the vicinity of the MC inclusions [3]. A delayed and slow provision of water to the cement in the particle bed could reduce the amount of evaporating water. This effect increases with an increasing amount of MC. However, at higher MC dosages, we observed an antagonistic effect due to the reduction in density, i.e. an increase in the porosity of the printed object. In addition, a reduction in the penetration length of the water – as observed in the capillary intrusion experiments - and an associated poor layer bonding could also contribute to the reduction in compressive strength at high MC dosages.

To summarize, the addition of MC is an effective method for printing objects with high geometric precision and good mechanical performance. However, too high MC dosages do not lead to the desired effect, i.e. the compressive strength decreases without further decreasing S_{3D}. Therefore, we recommend a moderate amount - in this example 1–1.5 wt.% - for addition to the particle bed.

4 Conclusion

The effect of methyl cellulose (MC) in selective cement activation (SCA) is studied in the presented experiments. Therefore, we varied the i) degree of polymerization (DOP) (low/high), ii) size of the granule (fine/coarse) and iii) amount of MC (0 to 2.5wt.-%).

We showed that a lower DOP and smaller granules of MC are beneficial in terms of dissolution rate of MC, which finally dominates the liquid intrusion behavior in the particle bed and thus governs the shape accuracy of SCA-printed objects. However, in the investigated range the effect of the DOP is minor compared to the size of the granules. Furthermore, we showed that higher dosages of MC increase the named effects. However, for the hardened properties of SCA-objects it was found that too high MC dosages can decrease the compressive strength without further decreasing the Shape Index. Therefore, we recommend a moderate amount in the particle bed.

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