

Preparation of Micro-Crystalline Cellulose-Reinforced Polyacrylic Acid Hydrogel and its Application in Paper/Polyacrylic Acid Composites

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Paper/polyacrylic acid (PAA) composites were prepared using a micro-crystalline cellulose (MCC)-reinforced PAA hydrogel. The MCC-reinforced PAA hydrogel was prepared by *in-situ* UV light-induced polymerization of acrylic acid in the presence of MCC. The experimental results showed that the presence of MCC improved the water absorption capacity of the resulting PAA hydrogel. The addition of 1 wt% MCC (based on the dry weight of acrylic acid) in the hydrogel matrix resulted in an increase in the water absorption capacity of the PAA hydrogel by 122 g water/g hydrogel (*i.e.*, from 427 to 549 g/g). Paper/PAA composites were then made by surface loading of the MCC-reinforced PAA hydrogel fine particles. Using 2 g of MCC-reinforced PAA hydrogel particles per square meter of paper, the water absorbance of the paper/PAA composites reached 1096 g/m². Potential applications of these paper/PAA composites are also discussed in this work.

Keywords: Micro-crystalline cellulose (MCC); Polyacrylic acid (PAA); Hydrogel; Water absorbency; Paper/PAA composites

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INTRODUCTION

Cellulose has been commercially utilized in many areas, including various paper products such as printing/writing grades and various tissue grades (Hu *et al.* 2007; Beuther *et al.* 2010; Hubbe *et al.* 2013). Because of the continuing decrease in the demand for printing/writing grades, it is of commercial interest to develop new applications for cellulose materials.

Micro-crystalline cellulose (MCC), or nano-crystalline cellulose (NCC), with some unique physico-chemical properties, has generated great interest recently (Jahan *et al.* 2011; Kurihara and Isogai 2014). Their various applications have been reported in the recent literature, *e.g.*, as an additive to enhance paper strength (Ahola *et al.* 2008), surface application onto yarns after cationic modification (Zaman *et al.* 2012), as an additive to prepare thermal/pH-sensitive hydrogels (Cha *et al.* 2012), as carriers for hydrophobic molecules to enhance the photo-chromic efficiency (Sun *et al.* 2014a), application to improve the stability and efficiency of ASA internal sizing of cellulosic paper after cationic modification (Sun *et al.* 2014b), and as a carrier for triclosan to develop anti-bacterial paper products (Liu *et al.* 2015).

Superabsorbent polymers, also known as hydrogels, are commercially produced by the polymerization of acrylic acid or similar monomers. This type of material has been widely used in agriculture (Chambers *et al.* 1997), sanitary products, diapers (Zohuriaan *et al.* 2008; Zohuriaan *et al.* 2010), disposable towels and bath mats, disposable litter mats

for pets, and release control for various chemicals (Colombo 1993). The ideal products must absorb fluid rapidly and have high absorbency. In the literature, it was reported that the addition of NFC in the preparation of hydrogel will render the resulting product with enhanced bio-compatibility, biodegradability, and other environmental benefits (Güçlü *et al.* 2010; Spagnol *et al.* 2012a). There are two major polymerization processes in the manufacturing acrylic acid-based hydrogels: 1) radical polymerization using persulfate or similar chemistry (Güçlü *et al.* 2010; Spagnol *et al.* 2012a; Spagnol *et al.* 2012b; Spagnol *et al.* 2012c; Zhou *et al.* 2013), 2) UV light induced polymerization (Lu *et al.* 1997; Tehfe *et al.* 2013; Wen *et al.* 2015). Zhou *et al.* (2013) found that the addition of carboxylated cellulose nanofibrils (CCNFs) improved the water absorption capacity of the resulting superabsorbent composites using ammonium persulfate as the radical initiator. Based on the potassium persulfate radical chemistry, Spagnola *et al.* studied the use of NFC in various hydrogel composites, including chitosan-g-poly (acrylic acid) (2012a), poly (acrylamide-co-acrylate) (2012b), and starch-g-poly (sodium acrylate) (2012c). Recently, Wen *et al.* (2015) reported the results of using NFC in the PAA preparation based on the UV light induced-PAA polymerization, which is commercially practiced at Tianjin Chemole Absorbent Material Co. The advantages of the UV light PAA manufacturing process include low energy input, low operating temperature, high reaction efficiency, and high monomer conversion rate (Lu *et al.* 1997; Tehfe *et al.* 2013).

Synthetic hydrogels, such as PAA, have negligible or low biodegradability (Cooke 1990). Currently, there is a strong public demand for increasing/improving the recycling and/or biodegradability of any product/material after its use (Sannino *et al.* 2009; Feng *et al.* 2010).

The objective of this work was to develop and design paper/PAA composites that have good water absorbency and can be used in different applications, such as water retention in lawns and gardens. PAA was synthesized using a UV light-induced polymerization process, and micro-crystalline cellulose (MCC) was added during the polymerization to improve the water absorbency and potential biodegradation after use of the resulting hydrogel. The paper/PAA composites were then made by surface loading of the MCC-reinforced PAA hydrogel fine particles onto a paper surface. The potential applications of such paper/PAA composites are then illustrated in designing/testing a product that can be used for water retention in lawns and gardens.

EXPERIMENTAL

Materials

The MCC was obtained from Tianjin Haojia Cellulose Co., Ltd. (Tianjin, China), and it was prepared from a sulfite-based dissolving pulp, which was based on an acid hydrolysis process. Acrylic acid (99.9 wt%; analytical grade), sodium hydroxide, poly(ethylene glycol) diacrylate ($M_n = 250$), 1-hydroxycyclohexyl phenyl ketone, 2,2'-azobis(2-methylpropionitrile), aluminum sulfate, methanol, glycol diglycidyl ether, and poly(vinyl alcohol) (+99%; molecular weight = 85,000 to 124,000; hydrolyzed) were purchased from Sigma-Aldrich Inc. Fisher brand filter paper (P8 09-795F; 15.0 cm; coarse) was purchased from Fisher Scientific.

A K 303 Multicoater (RK Printcoat Instruments Ltd., UK) was used for the surface coating/loading of hydrogel fine particles onto the paper surface.

Preparation of MCC-Reinforced PAA Hydrogel

The preparation of the acrylic acid solution was carried out as follows: 100 g of acrylic acid was neutralized using a sodium hydroxide solution at room temperature, and its concentration was adjusted to 48% in a beaker containing a magnetic stirring rod. The solution was then cooled down to room temperature in an ice water bath. Afterwards, a pre-set amount of MCC (0.5%, or 1%, 1.5% or 2% based on the dry weight of acrylic acid) in dried powder was added to the acrylic acid solution and mixed for 3 min at 20,000 rpm using a mechanical stirrer to disperse the MCC particles.

Subsequently, 10 g of the acrylic acid solution prepared as described above was taken and placed into a beaker, to which 0.1 g of 1-hydroxycyclohexyl phenyl ketone, 0.1 g of azodiisobutyronitrile (AIBN), and 0.15 g of poly(ethylene glycol) diacrylate were added. UV light-induced polymerization was carried out under the conditions of 1000 W (at $\lambda = 365$ nm) for 8 min. The hydrogels were then cut into thin slices and dried in an oven at 105 °C. Once dried, the hydrogel samples were ground and sieved to four fractions: < 40 mesh, 40 to 60 mesh, 60 to 120 mesh, and >120 mesh. During the experiments, the focus was on the utilization of fine hydrogel particles; thus, samples of > 120 mesh were used.

Determination of Water Absorbency

A sample of the prepared hydrogel fine powder (0.5 g) was immersed in 1000 mL of water at room temperature for 30 min. Then, the samples were filtered in a Büchner funnel and weighed. The equilibrium water uptake was calculated based on the following equation,

$$S(\text{g/g}) = \frac{W_d - W_s}{W_d} \quad (1)$$

where, S (g/g) is the water absorbency of the hydrogel (g of water/g of dried sample), and W_d (g) and W_s (g) are the weights of dried and swollen samples, respectively.

Preparation of Paper/PAA Composites

A poly(vinyl alcohol) (PVA) solution was prepared by dispersing 50 g of PVA in 450 g of distilled water (for a total weight of 500 g). The PVA dispersion was stirred for 20 min first, then gelatinized in a 95 °C water bath for 30 min and cooled to room temperature. A magnetic stir bar was used for mixing the dispersion.

A 10 wt% PVA solution, prepared as mentioned above, was then coated onto a Fisher brand filter paper using a lab coater and a #8 bar at a speed of 10 m/min; the PVA coated weight was approximately 2 g/m². Subsequently, the MCC-reinforced PAA hydrogel fine particles were sprayed onto the paper by using a sieve screen (Fisher Scientific, via shaking from about 20 cm away from the paper surface; the added amount was obtained from the weight increase of the paper sheet), which was then covered by another piece of filter paper. The multilayer sandwiched sheet was subsequently pressed at 3 MPa for 2 min and then dried using a Labtech (Canada) speed dryer for 90 s at 105 °C.

The equilibrium water uptake of the sandwiched sheet was determined as follows: the sheet was cut into samples of 50 mm x 50 mm. The pre-weighted dried sample was immersed in excess distilled water at room temperature for 30 min, then taken out and dried in air for 30 min (to remove non-bound water). Finally, the samples were weighed and the equilibrium water uptake was calculated based on Eq. 1. Five trials were done for each sample, and the average was reported.

TEM and SEM Analyses of the MCC and Superabsorbent Samples

Procedures reported earlier (Wen *et al.* 2015) were followed. Briefly, for the TEM analyses, a 0.01 wt% MCC suspension was prepared. Then, a drop was transferred to a carbon-coated copper grid and air-dried overnight at room temperature. The TEM analysis was conducted using a JEOL 2010 STEM instrument (Japan) operated at an accelerating voltage of 200 keV. For the SEM analyses, glass slides containing the superabsorbent samples were covered with a layer of gold. A Hitachi SU-1510 field emission SEM operating at 3.0 kV was used.

RESULTS AND DISCUSSION

MCC-Reinforced PAA Hydrogel

The *in-situ* UV light-induced polymerization of acrylic acid was carried out in the presence of MCC. A schematic of the process is shown in Fig. 1. MCC particles were the frames within the PAA polymer matrix. It was expected that the formed hydrogel would be of porous structure and would therefore increase the water absorbency of the matrix network. The hydrogel resulting from the addition of MCC particles may resemble the structure of fruit jello, where the role of the MCC is like that of the fruit pulp. The MCC opens up the PAA matrix structure, thus imparting the resulting hydrogel with improved water absorbency capacity. Because of its porous structure, the water swelling rate of the formed hydrogel is expected to increase. In the literature, Spagnol *et al.* (2012) reported that starch-g-poly(sodium acrylate) superabsorbent hydrogel that was filled with cellulose nano-whiskers had an increased water absorbance rate *versus* the control. Furthermore, the introduction of MCC to a polymer matrix can increase the physical strength of the formed hydrogel.

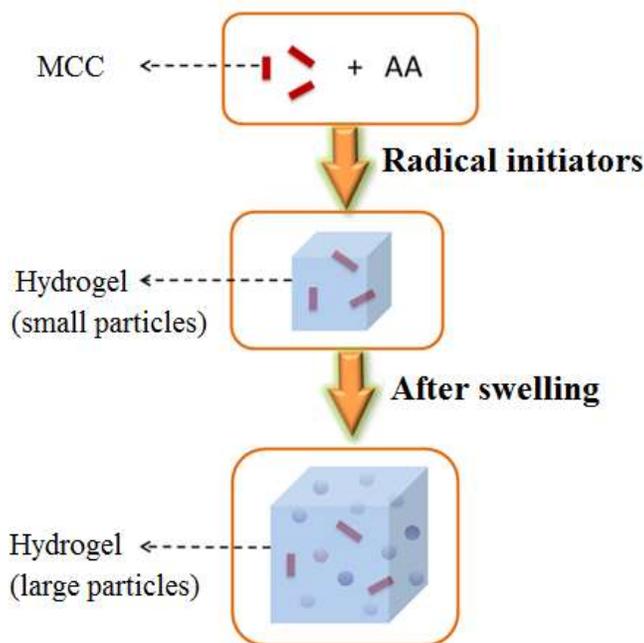


Fig. 1. Schematic illustration of the MCC/PAA composites formed by *in-situ* polymerization of acrylic acid in the presence of MCC under UV light (Wen *et al.* 2015)

Figure 2 presents the SEM images of the prepared hydrogel matrix with and without the addition of MCC. In comparison with the control (*i.e.*, PAA without MCC), it was found that a 1.0% MCC addition yielded a highly porous hydrogel structure. The presence of these rough surfaces/pores can facilitate water penetration, thus increasing the water absorbency of the polymer hydrogel matrix.

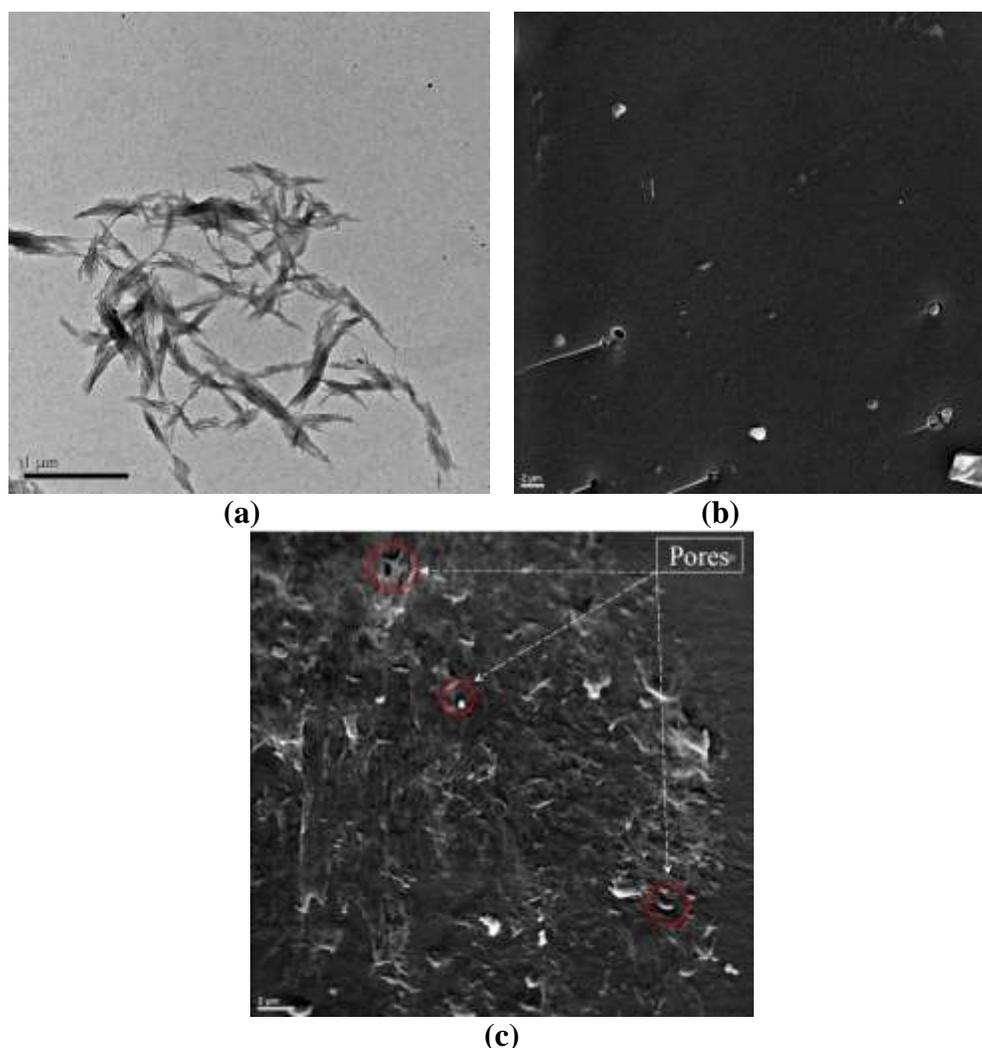


Fig. 2. (a) TEM image of MCC, (b) SEM image of superabsorbent polymer without MCC, very smooth surface with no apparent pores, and (c) SEM image of MCC-reinforced PAA hydrogel with 1.0% MCC content, a rough surface with many pores

The effect of the MCC addition level on the water absorbency of the hydrogel is plotted in Fig. 3. It was found that with the addition of 1.0% MCC, the water swelling capacity increased by 122 g water/g hydrogel, from 427 to 549 g/g. The MCC in the hydrogel matrix can increase the internal/external surface area because of the formation of many pores in the hydrogel structure (Fig. 2), which improved the water absorbency of the formed MCC/PAA hydrogel. On the other hand, when the MCC addition was greater than 1%, the water absorbance of the resulting hydrogel decreased because of the negative effects of high MCC levels on the UV light-induced polymerization process.

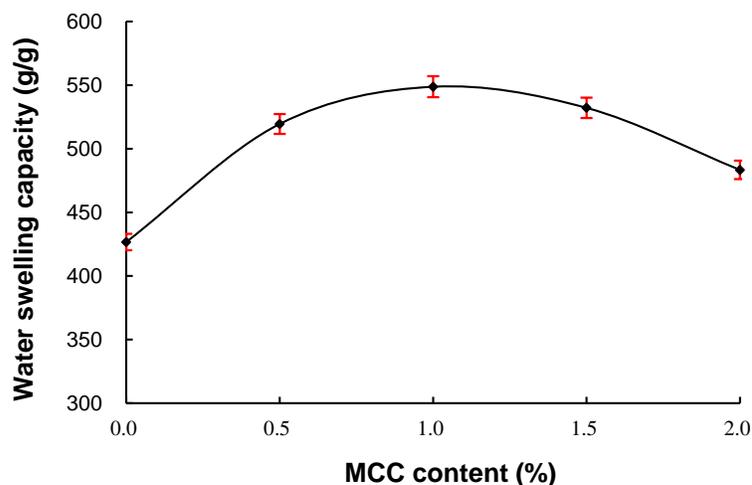


Fig. 3. Effect of MCC on the water absorbency of the MCC-reinforced poly(acrylic acid) (PAA) hydrogel

Design of Paper/PAA Composites

Very fine hydrogel particles (mesh size >120) have been reported to be unsuitable for hygienic or agricultural applications because of the gel blocking effect, and they may have to be recycled in the manufacturing process (Kabiri *et al.* 2011). In this study, these fine hydrogel particles were used to prepare paper/PAA composites. The key elements of the paper/PAA composites design included:

- 1) A layer of hydrogel with high water absorbance capacity; and
- 2) Cellulosic paper as the support layers.

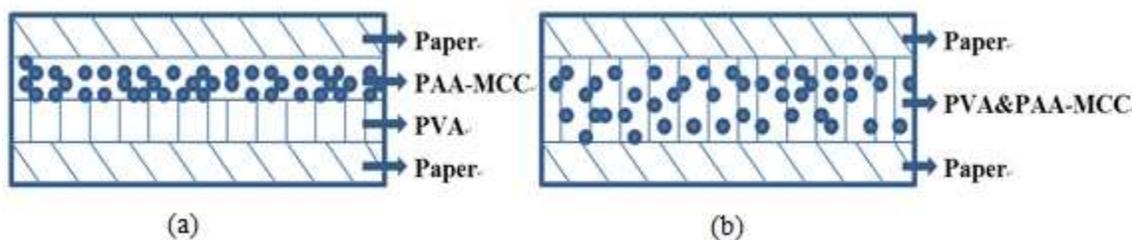


Fig. 4. Schematic representation of typical multi-layered paper/PAA composites

Figure 4 gives a cross-sectional view of the design of the paper/PAA composites. The general concept was to use paper as a recyclable/biodegradable substrate that could support the fine hydrogel particles, which would then meet the water absorbency requirements of specific end-use applications. The paper served as the supporting substrate for the fine PAA hydrogel particles. Two different methods of preparation were used: (a) the binder, such as PVA, was applied to the paper surface, and then a thin layer of the fine PAA particles was sprayed/coated on the top; and (b) the binders and fine hydrogel particles were mixed, and the mixture was then applied onto the paper surface *via* coating.

The binders that can be used in the product design are those commonly used in the paper industry, such as PVA (Jonjankiat *et al.* 2011), synthetic latexes (Piltonen *et al.* 2014), starch (Emengo *et al.* 2002; Wang *et al.* 2013), and carboxymethyl cellulose (CMC) (Ben-Zion *et al.* 1997). Other additives, such as dispersants, could also be used to enhance the product performance.

Various paper products, such as tissue paper and paperboard with various basis weights, can be utilized, and the choice would depend on the end-use specifications. The fiber sources can be virgin fibers, including chemical pulps (softwood, hardwood, or non-wood kraft fibers), mechanical pulps, and high-yield pulps. Furthermore, recycled fibers could also be used.

The concept is based on the idea that paper/PAA composites with substantial water absorption capacity can be produced using this technique. The production costs would be competitive because cellulosic fibers are usually more cost-effective than synthetic hydrogels. Such products are produced such that the high-water absorbance hydrogel is contacted to the paper surface using ordinary surface treatment processes, such as coating. In this case, the high-water absorbance hydrogel layer is applied onto the surface of the paper.

The proposed product design of paper/PAA composites in this study may have a number of advantages. It can be used to bring a high water absorbance hydrogel onto the surface of the paper using a conventional surface treatment process. It is possible to conceal the hydrogel particles under another layer of paper. It is also possible to have several paper/hydrogel layers sandwiched together in the final product so that various products with different water absorbance capacities can be produced. It would be easy to adjust the amount of the hydrogel in the paper/PAA composites by changing the amount of the coating applied to the paper substrate.

An Example of Practical Applications of the Paper/PAA Composites

The paper/PAA composites can be used to form various shapes, depending on specific product applications. For example, a multi-layered material can be folded in an appropriate manner to form a bag or an envelope. The objective of the present product design is to utilize the fine hydrogel particles that are smaller than > 120 mesh, which are otherwise usually recycled in the manufacturing process (Kabiri *et al.* 2011). Instead, the fine particles can be loaded onto a paper surface, forming paper/PAA composites that can overcome the “too small” nature of these fine particles. PVA, or other polymers, can be used as the binder. In a typical application, the loading of fine hydrogel particles of up to 2 g/m^2 of paper should be sufficient. As shown in Fig. 5, a 2 g/m^2 PAA hydrogel loading on the paper surface by following the ‘a’ type paper/ PAA composites design, resulted in a water absorbance of as high as 1096 g/m^2 .

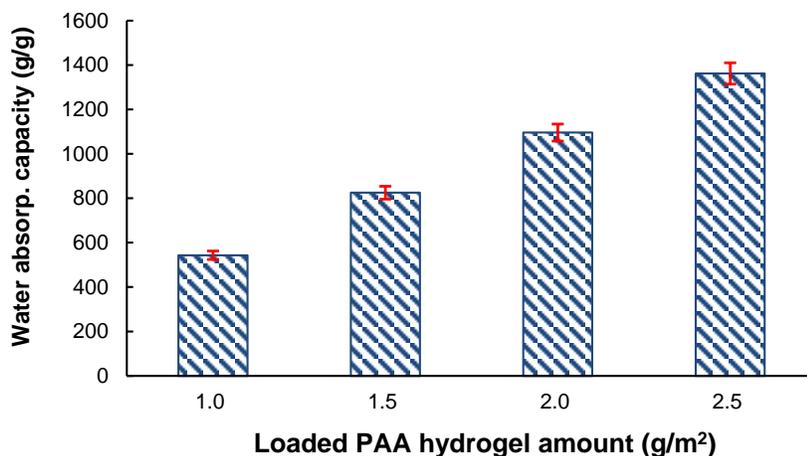


Fig. 5. Effect of coating various amounts of PAA hydrogel onto the paper substrate on the water absorbency of the resulting composites

An application example of the present paper/PAA composites is illustrated in Fig. 6. In this example, sheet forms of the paper/PAA composites can be used in the plant pot/bedding. The paper/PAA composites that are loaded with fine PAA particles can improve soil breathing, moisture retention, and other important properties.



Fig. 6. Schematic for using the paper/PAA composites in plant pot/bedding (Left: the actual paper/PAA composites with 2 g/m² PAA hydrogel loading; Middle: plant in a pot with paper/PAA composites as bedding; Right: explanation of paper/PAA composites as plant bedding)

CONCLUSIONS

1. Another application of micro-crystalline cellulose (MCC) was given in this study, and it was used for the preparation of MCC-reinforced PAA hydrogel produced by the UV light-induced polymerization of acrylic acid.
2. The addition of MCC improved the water absorbance of the resulting PAA hydrogel, and a 1.0 wt% MCC addition (added as powder, based on the dry weight of acrylic acid) led to an increase in the water absorption capacity of the PAA hydrogel by 122 g water/g hydrogel (from 427 to 549 g water/g hydrogel). The positive results from using MCC on the PAA hydrogel water absorbance were explained by the increased internal/external surface areas of the product, which were supported by the SEM results.
3. The very small particles (> 120 mesh) of the resulting MCC-reinforced PAA hydrogel, which are under-used in conventional applications, were made into a paper/poly (acrylic acid) (PAA) composites. These fine hydrogel particles were loaded onto a paper surface *via* techniques such as surface spraying and using typical binders, including PVA, starch, and CMC. The addition of 2 g of MCC-reinforced PAA hydrogel per m² of paper surface, the water absorbance of the resulting paper/PAA composites reached 1096 g/m².
4. A product design was given as an example of using such paper/PAA composites, that is, as plant pot/bedding that can improve soil breathing and moisture retention.

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