### SYNTHESIS OF A NOVEL CELLULOSE-BASED HYDROGEL/NANO-HYDROXYAPATITE COMPOSITE AND POTENTIAL REGULATION OF NITROGEN FERTILIZER RELEASE

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The objective of this study was to create a nano hydroxyapatite/cellulose-based hydrogel composite as a green nitrogen fertilizer with sustained release properties. The composite was prepared by reacting cellulose, CMC-Na, citric acid as a cross-linking agent, nano hydroxyapatite, and nitrogen fertilizers KNO<sub>3</sub> and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. The shape and structure were confirmed using FT-IR and SEM. The swelling ratio was investigated in deionized water. Furthermore, the impact on soil water content was also considered. Finally, the release rates of the fertilizers in deionized water and soil were investigated. The swelling ratio of the composite reached its maximum (1000%) on day 10, and it also exhibited slow release properties. The cumulative release rates of nitrate and sulfate reached 70% and 100%, respectively, in deionized water and soil, the soil-water content was enhanced, reaching 18% on day 20. The outcomes were consistent with the standards set forth by the Committee of European Normalization. In conclusion, this novel composite has the potential for utilization in agricultural applications.

Keywords: cellulose, citric acid, CMC-Na, nano hydroxyapatite, slow-release fertilizer, soil-water content.

#### **INTRODUCTION**

The twenty-first century has brought with it a number of challenges for the agricultural sector. Chief among these is the continued growth of the global population,<sup>1</sup> which is placing increasing pressure on the world's natural resources.<sup>2</sup> This, in turn, is limiting the availability of these resources, which are essential for supporting life on Earth. The extensive use of chemicals, including fertilizers, has an adverse impact on the environment, contaminating these resources. One example of this is the leaching of nitrate into groundwater, which exacerbates global warming and climate change.<sup>3</sup> Furthermore, fertilizers represent a significant source of adverse effects on soil quality,<sup>4</sup> including salinization, degradation, waterlogging and the disruption of soil microbial flora.<sup>5</sup> Furthermore, in certain crops, particularly leafy vegetables, such as lettuce, nitrate accumulates in the leaves due to the excessive application of nitrogen fertilizers, resulting in toxic effects on human health.<sup>6,7</sup> It is therefore of significant importance to identify alternative environmentally friendly agricultural practices that enhance plant productivity, whilst reducing the impact on the surrounding environment.

Nanotechnology, a novel technology of the 21<sup>st</sup> century,<sup>8</sup> is concerned with materials at the nano scale (10<sup>-9</sup>). The distinctive physico-chemical attributes of nanoparticles, including their diminutive size, elevated surface area-to-volume ratio, ability to interact with biological membranes, high reactivity, and tendency to agglomerate, have prompted extensive scientific investigation.<sup>9</sup> Nanotechnology has been extensively employed in

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a multitude of disciplines, including medicine, industry, architecture, chemistry, physics, biology, and agriculture.<sup>10</sup> Recently, nanomaterials, such as nanopesticides and nanofertilizers, have been embraced by agricultural practices across the globe.<sup>11,12</sup>

Nanofertilizers are defined as substances that deliver nutrients to plants in one of the following ways: by coating nanoparticles or emulsions with thin polymer films; or by encapsulating the nutrients inside a nanoporous substance.<sup>13</sup> These techniques facilitate the reduction of the rate of nutrient release, minimize nutrient loss and mitigate the deleterious impact of traditional fertilizers on crops and the environment.<sup>14</sup>

Hydroxyapatite, а naturally occurring phosphorus source, is employed in conventional agriculture as a phosphorus fertilizer.<sup>15,16</sup> Nanoparticles of hydroxyapatite (HA NPs) have been the subject of extensive study in a number of different fields, including medicine (for use in bone grafting procedures),<sup>17</sup> the dental industry, and drug delivery systems.<sup>18,19</sup> Lately, it was in agriculture; the hybrid employed of hydroxyapatite coated with urea was first created as a fertilizer by Kottegoda et al. (2017). It was reported to reduce the release rate of nitrogen by 12 times and raised the nitrogen use efficiency by 50%.<sup>16</sup>

Superabsorbent hydrogels are hydrophilic polymers that possess the capacity to absorb and conserve water. They are utilized in agricultural practices to reduce the frequency of irrigation and prevent water shortages during droughts,<sup>20</sup> as well as in medical applications as drug delivery systems.<sup>21</sup> Furthermore, it has been investigated as a fertilizer carrier, with the capacity to control nutrient release. The majority of the hydrogels under examination were synthetic polyacrylamide (PAM)-based hydrogels, which are well-known biodegradable,<sup>21,22,23</sup> and and nonand have been widely employed.<sup>20,24</sup>

Cellulose-based hydrogels are organic hydrophilic polymers, including cellulose, chitin, chitosan. which possess and notable characteristics, such as biodegradability, biocompatibility, and the capacity to respond to changes in pH, time, and temperature. Additionally, they exhibit a high water absorption capacity, making them environmentally friendly and a promising alternative to synthetic polyacrylamide-based super-absorbent gels.<sup>25</sup> However, the carcinogenic epichlorohydrin was used as cross-linking agent in the synthesis of cellulose-based hydrogels.<sup>26-31</sup> In a recent study, Tarawneh *et al.* (2021) synthesized a hydrogel that was cross-linked with epichlorohydrin, which they proposed as a potential drug delivery system.<sup>32</sup> A cost-effective, non-toxic, and biodegradable hydrogel was synthesized using citric acid (CA) as a cross-linking agent,<sup>24</sup> and tested as a water reservoir agent in agricultural practices.<sup>33,34,35</sup>

A series of composite materials comprising hydrogel, nanoparticles and fertilizers were synthesized and subsequently employed in agricultural applications. A combination of nanohydroxyapatite, hydrogel, and soluble NPK (nitrogen, phosphorus, and potassium) fertilizer was created and reported to result in a reduction in nitrogen mineral content in the soil in comparison with conventional fertilizer.<sup>22</sup> In the study conducted by Olad et al. (2018), the composite comprising nano-silica. **PVP** (polyvinylpyrrolidone) hydrogel, NPK and fertilizers demonstrated a gradual release of NPK at a rate of approximately 60% over a period of 30 days, accompanied by enhanced soil-water content.<sup>21</sup> Another composite comprising a PAMbased hydrogel and urea was developed by Zhang et al. (2020), which demonstrated an acceptable release profile for urea.23

Therefore, the aim of this study is to synthesize and characterize a green cellulose-based hydrogel, comprising a nano-hydroxyapatite and nitrogen fertilizer composite, derived from commercial natural cellulose, hydroxyapatite, and carboxymethyl cellulose-sodium salt (CMC-Na), and cross-linked with citric acid. The investigation will encompass several factors, including the swelling capacity, the behavior of the fertilizer release in deionized water and a soil mix of peat moss and sand, and its effect on soil-water content.

#### EXPERIMENTAL

#### Materials

Carboxymethyl cellulose sodium salt (CMC-Na) of medium viscosity and microcrystalline cellulose (MCC) were bought from Sigma Aldrich, Germany. Citric acid (CA) M = 192.13 g/mol was bought from C.B.H. Lab Chemicals, Nottengham, UK, and hydroxyapatite (HA) – from Trans-Tech Ceramics and Advanced Materials.

## Optimization of nano-hydroxyapatite amount in the hydrogel composite

The cellulose-based hydrogel was prepared according to Zyadeh *et al.* (2023).<sup>38</sup>

In order to identify the optimal ratio of hydroxapatite (HA) to be incorporated into the hydrogel

composite, the synthesis was conducted using the previously established procedure, with the key difference being the addition of HA. This was achieved by mixing variable amounts of HA (1 g, 2 g, and 3 g) with 1 g of cellulose in deionized water, followed by the gradual addition of 3 g of CMC-Na until the mixture was fully homogenized. Subsequently, 0.75 g of anhydrous citric acid, dissolved in deionized water, was added dropwise and mixed for one hour. The mixture was then left in a water bath at 30 °C for 24 hours. On the following day, the temperature was increased to 80 °C for a further 24 hours, after which the mixtures were dried in an oven at 40 °C for 72 hours.

#### Characterization

The FTIR spectra (4000 to 400 cm<sup>-1</sup>, 4 cm<sup>-1</sup> spectral resolution, KBr pellets) were recorded using a Thermo Nicolet NEXUS 670 FT-IR spectrometer. The surface morphology of the composite was examined using scanning electron microscopy (FEI, Versa 30).

### Formulation of cellulose-based hydrogel/hydroxyapatite/nitrogen fertilizer

The mixtures, comprising cellulose, HA and CMC-Na, were loaded with predetermined amounts of the fertilizers  $KNO_3$  and  $(NH_4)_2SO_4$ , which were then mixed well before the addition of citric acid.

#### Water absorption capacity

In order to investigate the expansion dynamics of the hydrogel composite, dried samples from the prepared hydrogel composite were weighed and subsequently soaked with distilled water for a period of 24 hours. Following this, the samples were removed from the distilled water and weighed again, after the removal of any excess water by using filter paper. The swelling ratio (SR%) was calculated in accordance with Equation (1) and expressed as a percentage:<sup>24</sup>  $SR\% = \frac{WS - WD}{WD}X \, 100\%$  (1)

where WS – the swelling weight, WD – the dry weight.

#### Release behavior in deionized water

Fertilizer release behavior was confirmed by measuring  $NO_3^-$  and  $SO_4^-$  concentration using a UV-spectrophotometer (Genesys 10UV Scanning). Samples of fertilizer loaded hydrogel composite were soaked in plastic cups with 100 mL deionized water. Concentrations were assessed daily according to the standard methods of APHA (2012).<sup>35</sup>

#### Release behavior in a soil mix

A soil mixture comprising peat moss and sand in a 1:1 ratio was amended with HGH/N fertilizers and placed in a glass-centred column equipped with a valve. Subsequently, 80 mL of distilled water was added, and the columns were left at room temperature. The control group comprised identical columns filled with soil amended with a similar amount of commercial fertilizers. A 10 mL sample was taken for measuring electric conductivity using a TDS and EC meter (hold) model A1. Ten mL of distilled water was added to each sample in order to maintain constant volume.

#### Soil water content

Samples of the soil mix of peat moss and sand (weight 1:1) were amended with 1% hydrogel, then put in plastic cups and weighed (W0), irrigated to field capacity and weighed again (W1). For the control group, plastic cups were filled with the soil mix without hydrogel. Cups were left at room temperature and weighed daily (W2). Soil water content (SW %) was calculated according to Equation (2) and expressed as percentage:<sup>21</sup>

$$SW\% = \frac{W^2 - W^0}{W^1 - W^0} \dots$$
(2)

All experiments were repeated three times. Graphs, means, and standard deviation were generated using Microsoft Excel version 10.

#### **RESULTS AND DISCUSSION**

Figure 1 illustrates the impact of varying the quantity of nano-hydroxyapatite on the absorption capacity of the hydrogel. The addition of 1 g of HAp demonstrated the highest swelling percentage, reaching approximately 1000% within 24 hours. This significant increase in absorption capacity, up to approximately three-fold that of HG (data not shown), indicates the remarkable adsorption capacity of HAp. As the quantity of HAp increased, the degree of swelling decreased. This may be attributed to the brittle nature of HAp, which affects the elasticity of the hydrogel. Alternatively, the addition of HAp may fill the pores of the hydrogel, reducing its overall size. Due to its good absorption capacity, the composite made using 1 g of HA was selected for further experiments and termed HGH.

#### **Durability of HGH**

Figure 2 illustrates that the incorporation of HA into the base hydrogel forming HGH results in a threefold increase in the swelling ratio. However, the durability was found to decrease as the HGH approached its maximum swelling ratio on the tenth day, subsequently indicating the onset of degradation. The observed degradation can be attributed to the weakness of the physical crosslinking reaction.

#### **FT-IR** spectroscopy

The FTIR spectra of the hydrogel composite (HGH) are presented in Figure 3. The bands ranging from 3400 to 3571 cm<sup>-1</sup> correspond to the stretching vibration of OH group, the bands located

at 1620 cm<sup>-1</sup> indicate the stretching vibration of C=O.<sup>33</sup> The bands at 1044.5 cm<sup>-1</sup>, 1053 cm<sup>-1</sup> and 604.5 cm<sup>-1</sup>, 603.5 cm<sup>-1</sup> are related to  $PO_4^-$ 





stretching and bending vibrations of hydroxyapatite in the hydrogel HGH respectively.<sup>22</sup>



Figure 2: Effect of time on swelling ratio of the hydrogels HGH in deionized water



Figure 3: FTIR spectrum of HGH

#### SEM observation

SEM micrographs for HAp, HG and HGH are shown in Figure 4. As may be noted in the SEM micrograph in Figure 4a, HAp appears as aggregates with the size of 125 nm, while HG exhibited a compacted and layered microstructure that is highly oriented (Fig. 4b). After HA addition (HGH), the layered structure became more pronounced and took a coral shape, the structure was transformed to long slabs that are highly oriented (Fig. 4c). The average size and thickness of the slabs are 370 nm and 185 nm, respectively. In conclusion, the addition of HA to the cellulosebased hvdrogel showed а tremendous improvement in surface and alignment properties, and increased the water uptake.

#### Release behavior in deionized water

Figure 5 (a and b) displays the trend in the release rate of nitrate and sulfate of HGH loaded with fertilizers KNO<sub>3</sub> and  $(NH_4)_2SO_4$ . The initial release rate was low, at 1% and 10%, respectively, and then increased steadily, reaching 70% and 100% within one month, respectively. These results align with the definition of slow-release fertilizer set by the Committee of European Normalization.<sup>39</sup>

#### Release behavior in soil mix

Figure 6 (a and b) illustrates the release profile of KNO<sub>3</sub>/HGH and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>/HGH hydrogels in a soil mixture comprising peat moss and sand (in a 1:1 ratio by weight). The results demonstrated that the HGH/fertilizers exhibited a sustained, gradual release rate in contrast to that observed in the control groups. The initial release rate was 0% for potassium nitrate ammonium sulfate, reaching 65% and 60% by day 30. The release rate was

found to be in alignment with the definition of a slow-release fertilizer established by the Committee of European Normalization.<sup>39</sup>



Figure 4: SEM images at 5 µm scale for (a) HAp, (b) HG, and (c) HGH



Figure 5: Release rate of (a) nitrate from KNO<sub>3</sub> and HGH/KNO<sub>3</sub>, and (b) of sulfate from (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and HGH/(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in deionized water



Figure 6: Release rate of (a) KNO<sub>3</sub> and HGH/KNO<sub>3</sub>, and (b) of (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and HGH/(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> in the soil mix of peat moss and sand (weight ratio 1:1)



Figure 7: Soil-water content of the soil mix peat moss and sand (weight ratio 1:1) without hydrogel (control), and with HGH

#### Soil-water content

Figure 7 illustrates that the soil water content was improved when amended with 1% of HGH, reaching 15% on day 30. In the control group, the soil mix devoid of hydrogel exhibited a precipitous decline in water content after day 5, reaching 15% and subsequently becoming entirely depleted on day 20. In general, the addition of hydrogel composite (HGH) resulted in an increase in the water content of the soil mix. The results were comparable to those reported by Montesano *et al.* (2015). Consequently, this hydrogel composite could be employed in agricultural practices to reduce the frequency of irrigation and enhance plant performance during periods of drought.<sup>21,34</sup>

#### CONCLUSION

novel cellulose-based hydrogel/ А nanohydroxyapatite composite with slow-release properties was successfully synthesized using a CMC-Na backbone and the natural polymer cellulose. The composite was loaded with either KNO<sub>3</sub> or (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> nitrogen fertilizers. The composite exhibited an appreciable SR% (1000%) in contrast to synthetic polymer-based hydrogels. Furthermore, the HGH/N fertilizers demonstrated slow release rates in deionized water and soil mixtures, which aligns with the CEN definition of slow-release fertilizers. The findings indicate that this green nano-composite hydrogel has the potential to serve as an effective alternative to other synthetic hydrogels, with the possibility of being utilized in agricultural practices to mitigate environmental impact. The objective was to examine the impact of this newly formed composite on the growth and quality of the lettuce crop.

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