Ahmad *et al. Chem. Biol. Technol. Agric.* (2023) 10:36
https://doi.org/10.1186/s40538-023-00407-6

Chemical and Biological
Technologies in Agriculture

RESEARCH

Open Access

Eco-friendly cellulose-based hydrogels derived from wastepapers as a controlled-release fertilizer

Dayang Fazirah Binyi Abg Ahmad[1], Mohd Effendi Wasli[1], Cindy Soo Yun Tan[2], Zaki Musa[3] and Suk-Fun Chin[1*]

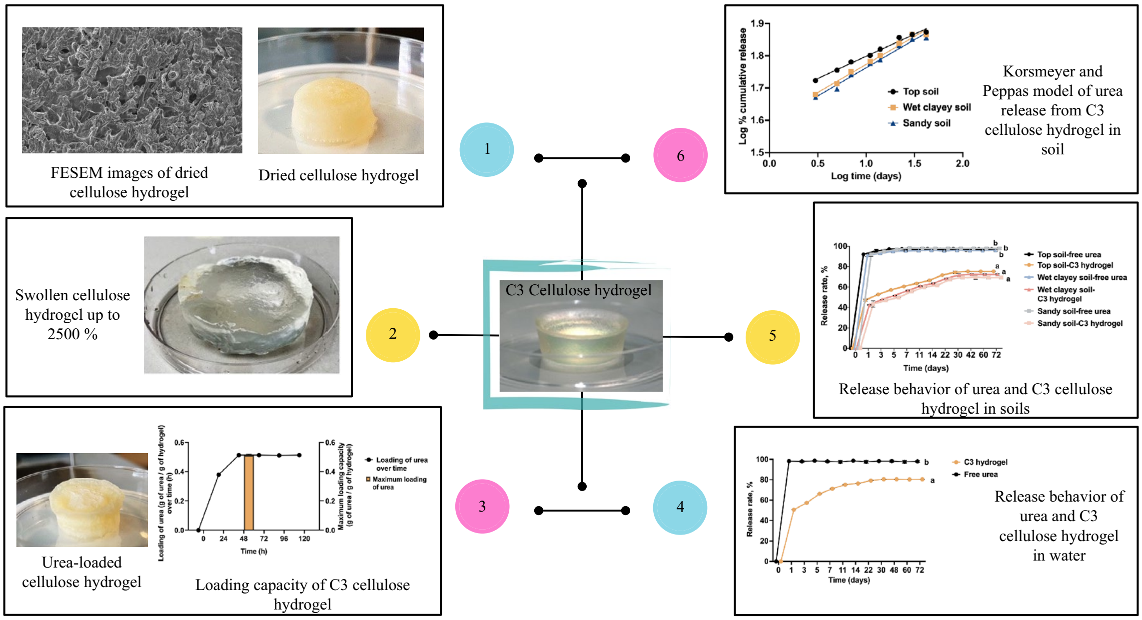
Abstract

In this study, an eco-friendly controlled release fertilizer cellulose-based hydrogel was prepared from cellulose fibers derived from wastepaper, epichlorohydrin (ECH) as a crosslinker and carboxy methyl cellulose (CMC) as a gelling agent. A maximum swelling capacity of 2000% was achieved for cellulose hydrogel with optimum composition. The soil moisture contents in the presence of optimized cellulose hydrogels were determined using the digital moisture meter. Maximum soil moisture of 36.5% was obtained in topsoil, followed by 30.1% in wet clayey soil and 23.4% in sandy soil after 7 days. Urea as a model fertilizer was loaded onto the cellulose hydrogels to control the release of fertilizer. The maximum loading capacity of urea in cellulose hydrogel is 0.51 g/g. The urea-controlled release profiles of the cellulose hydrogel in distilled water and various types of soils were investigated. The formulation of cellulose hydrogels was observed to facilitate the gradual release of urea, with about 74.71% release in topsoil, 73.37% release in wet clayey soil and 71.84% release in sandy soil within 42 days when compared to the free urea which was about 97.32%, 95.09% and 98.47% release in topsoil, wet clayey soils and sandy soils, respectively within 7 days. The result of this study shows that the urea-loaded cellulose hydrogel could be a promising controlled-release fertilizer.

Keywords Controlled-release fertilizers, Carboxymtehyl cellulose, Swelling ratio, Water retention, Sustainable agriculture

*Correspondence:
Suk-Fun Chin
sfchin@unimas.my
Full list of author information is available at the end of the article

Graphical Abstract



Introduction

Cellulose hydrogels refer to three-dimensional (3D) network structures consisting of highly crosslinked linear or branched hydrophilic cellulosic polymer chains [1, 2]. Upon contact with water, cellulose hydrogels can absorb and hold a huge amount of water in their polymeric networks and swell up to 99% [2, 3]. Due to their exciting properties, hydrogels have been used in enzymatic immobilization, solute separation, baby diapers [4], pharmaceuticals and biomedical applications [5–8], as well as soil conditioner for agriculture and horticulture [9]. Furthermore, superabsorbent hydrogels have been reported to improve soil conditions for cultivation via the enhanced water-holding capacity of the soil, reduced irrigation, high soil permeability, optimized nutrients uptake, and delayed dissolution of fertilizers, resulting in increased crop growth performance and yields [10, 11].

Fertilizers and irrigation are essential for plant growth to increase the quality and yield of any crop plants and the yield of the crops is mainly a function of fertilizer and water used [12]. However, low nutrient efficiency (< 50%) and nutrient loss are often reported by conventional fertilizers due to heavy leaching, surface run-off, undesirable reaction with organic compounds in the soils, and vaporization and volatilization processes [13–16]. A promising agriculture strategy to address these challenges is the encapsulation of fertilizers in superabsorbent hydrogels as controlled-release fertilizers to minimize fertilizer loss, extend nutrient supply for a longer period and enhance fertilizers utilization efficiency.

Currently available controlled-release fertilizers in the market are expensive due to the cost of coating materials and convoluted production process, thus deterring their wide use in agriculture [16]. Moreover, most of the fertilizer-controlled-release carriers are made of synthetic polymers, such as acrylamide-based products, making them less environmentally friendly due to their possible toxicity [16]. Cellulose-based hydrogels are desired for their renewable source, abundance, biocompatibility, biodegradability and ability to improve water retention in the soil [17].

In this work, a series of cellulose-based hydrogels were fabricated by crosslinking cellulose fibers extracted from waste papers and carboxymethylcellulose (CMC) with epichlorohydrin (ECH). The cellulose fibers were derived from wastepapers, because papers are readily available precursor materials and inexpensive. The ratio of cellulose and CMC concentrations was varied in the hydrogel fabrication and the swelling ratio of the hydrogels was compared. Urea-loaded cellulose hydrogel was prepared from the cellulose hydrogel with optimized physical–chemical properties. The release profiles of urea-loaded cellulose hydrogels were investigated in water and different types of soils to evaluate the potential use of these hydrogels as a controlled-release system.

Materials and methods

Chemical reagents and materials

Cellulose fibers extracted from wastepaper were used as the raw material. Sodium carboxymethyl cellulose (CMC, $M_w = 262.9$) was obtained from Sigma Aldrich (Wisconsin, United States of America) and epichlorohydrin (ECH, $\geq 99\%$ GC, $M_w = 92.52$) was obtained from R&M Chemicals (Essex, United Kingdom). Sodium hydroxide (NaOH, $M_w = 40$) was also obtained from Sigma Aldrich and urea (N_2H_4CO, $M_w = 60$) was purchased from HmbG Chemicals (Hamburg, Germany). Reagent solution for the Hach method was obtained from Hach Company/Hach Lange GmbH (United States of America). All chemicals were used without further purification.

Soil properties

Topsoil is classified as red–yellow podzolic soil (the soil group is classified under Sarawak Soil Classification System). The soil is acidic with pH levels ranging from 4 to 5, with a fine sandy clay-like texture (30 – 50% clay) with a CECclay of more than 24 cmolckg^{-1}. Wet clayey soil is classified as gley soil (the soil group is classified under Sarawak Soil Classification System). The soil is alkaline with pH level ranging from 8 to 10 and contains clay minerals, in which the gley layer occurs within the reach of the root systems or cultivated plants, i.e., the gleyed horizon or 50% gleying within 50 cm below O horizon. Wet clayey soil is sticky, develops a plasticity texture when wet and may turn hard, brittle and non-plastic when dry. Sandy soil is classified as arenaceous soil (the soil group is classified under Sarawak Soil Classification System) with pH levels ranging from 5.8 to 6.3. The soil includes mineral soil content with a sandy particle-size class in the upper 50 cm or more of the profile.

Cellulose extraction from wastepapers

Shredded waste papers were grounded and soaked in water and stirred continuously at 2000 rpm for 2 h. Subsequently, the filtered sample was treated with 12% (w/v) NaOH under constant stirring for 12 h [18]. The extracted cellulose pulps were washed with distilled water several times to remove excess NaOH and dried in an oven at 60 °C to constant weight.

Preparation of cellulose hydrogel

Table 1 Composition of cellulose-based hydrogels

Composition	Cellulose concentration (% w/v)	CMC concentration (% w/v)
C1	1.0	1.75
C2	2.0	1.75
C3	3.0	1.75
C4	4.0	1.75
C5	5.0	1.75
CMC2.0	3.0	2.0
CMC2.5	3.0	2.5
CMC3.0	3.0	3.0
CMC3.5	3.0	3.5
CMC4.0	3.0	4.0
CMC4.5	3.0	4.5
CMC5.0	3.0	5.0

Cellulose hydrogel was prepared by adapting the procedure used by Chin et al. [18], Zhang et al. [19], Salleh et al. [20] and Alam et al. [21]. NaOH/urea (NU) of 7:12% (w/v) solvent was prepared and precooled before use. Cellulose fibers were dissolved in the NU solution and stirred for 1 h. Various concentrations of cellulose fibers (wt%) and CMC (Table 1) were added to the NU solvent to form a suspension. For example, to prepare a C1 cellulose hydrogel, 1% (w/v) cellulose was dissolved in the NU solvent for 1 h, and 1.75% (w/v) CMC was then added. The suspension was mixed until CMC was fully dissolved. After, 5 mL of 99% ECH was added slowly into the mixture and stirred at 1800 rpm at room temperature for 15–25 min until the mixture turned into a highly viscous gel. The viscous gel was then stored in a freezer at − 20 °C overnight to allow crosslinking to form hydrogels [18]. Thereafter, the hydrogel was obtained upon thawing at room temperature, immersed in excess distilled water, and thoroughly washed to remove excess NaOH, urea and ECH. Finally, the hydrogel was oven dried at 50–60 °C and kept for further characterization.

Sample characterization

Dried cellulose hydrogel was characterized using Fourier Transform Infrared (FTIR) Spectrometer (Thermo Scientific Nicolet iS10) within the wavenumber ranging from 400 to 500 cm^{-1} using ATR technique. The surface morphology of the sample was observed using Field Emission Scanning Electron Microscope (FESEM) (JEOL JSM-IT500HR) at a magnification of 1000x. The thermal stability of the samples was analyzed using Differential Scanning Calorimetry (DSC) (Netzsch-Geratebau GmbH-DSC 200 F3 Maia Thermal Analyzer) in temperature range from − 150 °C to 600 °C.

Swelling ratio

The swelling properties of cellulose hydrogels were studied in distilled water at 26 °C. A pre-weighed, dried regular-shaped hydrogel sample was prepared and immersed in distilled water under a controlled environment. The weights of the hydrogel were recorded until the equilibrium was reached. The swelling ratio was calculated according to the following equation [22]:

$$\text{Swelling ratio} = \frac{\text{M} - \text{Mf}}{\text{Mf}} \times 100 \tag{1}$$

where M and Mf are weights of swollen and dried samples, respectively.

The soil moisture properties

The soil moisture was determined using the digital soil moisture meter (Extech Instruments-MO750). 200 g sample of each type of soil was placed in the beaker and dried in the oven at 60 °C to constant weight. After that, the optimized cellulose hydrogel was buried in the dried soils and the initial soil moisture content was recorded. The moisture content of each soil was measured again after 1 week to determine the moisture properties of cellulose hydrogels in soils. Control treatment involving soils without the optimized cellulose hydrogel was also included.

Fabrication of urea-loaded cellulose hydrogel

The optimized cellulose hydrogel was selected based on the porosity, swelling and soil moisture properties for the fabrication of urea-loaded cellulose hydrogels as a controlled-release fertilizer [11].

Urea-loaded cellulose hydrogel was fabricated by adapting the procedure used by Li et al. [23]. A pre-weighed dried regular shape hydrogel sample was immersed into 2.5% (w/v) urea solution for 48 h and placed in a controlled environment. The swollen urea-loaded hydrogels were dried to constant weight. The loading capacity (mg/g) of fertilizers were calculated using the following equation :

$$\text{Loading capacity}\left(\frac{\text{mg}}{\text{g}}\right) = \frac{(\text{Total amount of urea}) - (\text{Nonbound urea})}{\text{Weight of hydrogel}} \tag{2}$$

The total N-nutrient in the solution was measured using the Hach method 8075 (Total Kjeldahl Nitrogen (TKN), Nessler Method (150 mg/l)) [23]. The Hach method was conducted by adapting the procedure from Hach et al. [24]

Release of urea in water and soils

Release of urea in water and soil was conducted by adapting procedure used by Li et al. [23] and Liu et al. [24]. To evaluate nitrogen release, the urea-loaded, dried hydrogel was immersed in distilled water (100 mL) and incubated at 20 °C. 2 mL of the solution was collected at intervals 1, 3, 5, 7, 11, 14, 22, and 30 days or the time when cumulative N release of the fertilizers reaches more than 80%. Subsequently, 2 mL of fresh deionized water was added to keep the total liquid volume at 100 mL. Control treatment involving free urea was also conducted. The total N-nutrient concentrations of the collected solution were measured using the Hach method [25].

The dried, urea-loaded, cellulose hydrogel was placed in non-woven plastic mesh bags and buried 5 cm beneath the surface of the soil in a pot containing 200 g of dried soil to investigate the release of urea from the hydrogel in the soil. The soils were watered two times daily with 70 mL of water. Small soil samples (0.25–0.30 g) were collected at 1, 3, 5, 7, 11, 14, 22, 30 and 42 days. Control treatment involving untreated urea was also included. The N-nutrient content in the soil was determined using the Hach method [26]. Each treatment was performed in triplicates. Three types of soils were used including topsoil, wet clayey soil and sandy soil to observe the release behavior of the cellulose hydrogel in different soils.

Release kinetics of cellulose hydrogels

The release behavior was analyzed using the following Korsmeyer–Peppas equation [27]:

$$\frac{M_t}{M_\infty} = K_{KP} t^n \tag{3}$$

where K_{KP}, t, and n are diffusion constant, time and diffusion exponent indicative of the release mechanism, respectively. M_t/M_∞ is the fraction of fertilizer released at the time, t. The nutrient release mechanism is classified according to different values of n. The value of $n < 0.45$ means that the fertilizer release mechanism corresponds to a Fickian model, while $0.45 < n < 0.89$ means that the fertilizer release mechanism is a non-Fickian diffusion transport. For $n > 0.89$, then the nutrient release obeys the case II transport [27].

Statistical analysis

Significant differences of the mean were statistically analysed with ANOVA, and the differences reported throughout are significant at $p \leq 0.05$ or 0.01 using SAS software package 9.2 (SAS Institute 2010). All diagrams were drawn using GraphPad Prism 9 v9.4.1 software (2022).

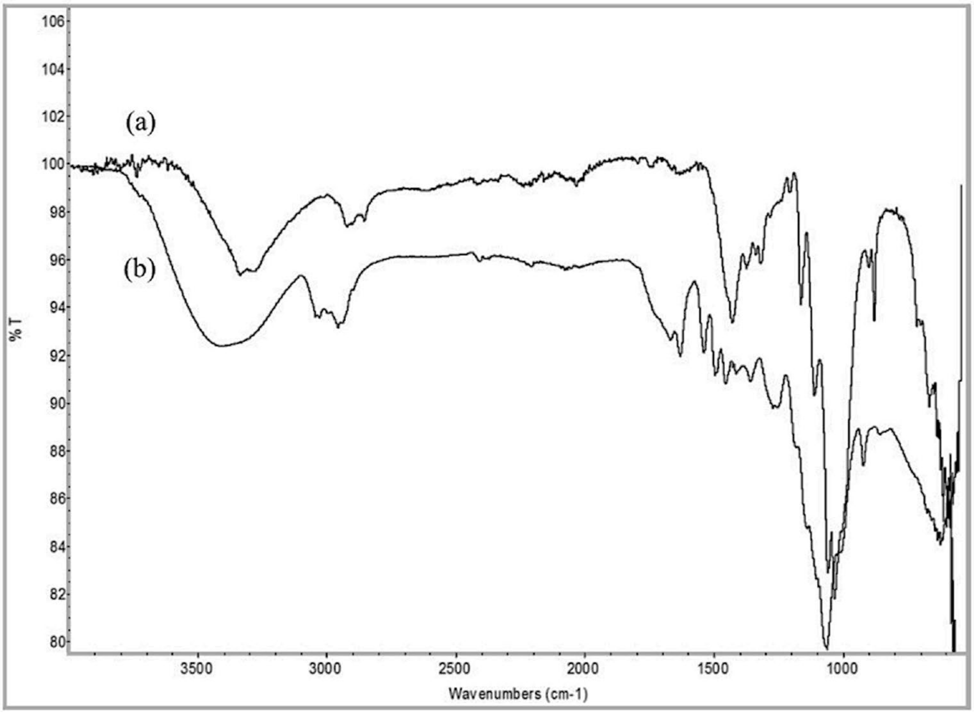


Fig. 1 FTIR spectra of **a** cellulose fibers and **b** ECH-crosslinked cellulose hydrogel

Results and discussion

FTIR analysis

Figure 1 shows FTIR spectra of (a) cellulose fibres and (b) an ECH-crosslinked cellulose hydrogel. The wavenumber range of 3360 cm^{-1} and 2900 cm^{-1} were referred to -OH and C–H bond of polysaccharides (Fig. 1a). The broad peak at 3335 cm^{-1} was due to the stretching vibration of the hydroxyl group in polysaccharides which included intermolecular and intramolecular hydrogen bonds vibration in cellulose. Figure 1b shows the crosslinking reaction in the cellulose hydrogels in which the transmittance peak at 1060 cm^{-1} was attributed to the stretching vibration of C–O–C which formed from the components of the reaction between ECH and CMC [19]. In addition, the presence of the carbonyl group (C = O) can be observed at 1720 cm^{-1} which confirmed the successful introduction of the carbonyl group in the structures [19]. This result also confirms the COOH groups in the CMC and hydrogels. Meanwhile, a new peak at 1328 cm^{-1} in the crosslinked hydrogel confirmed that crosslinking has occurred [19]. In conclusion, the expected product, a cellulose hydrogel, was well-prepared via chemical crosslinking.

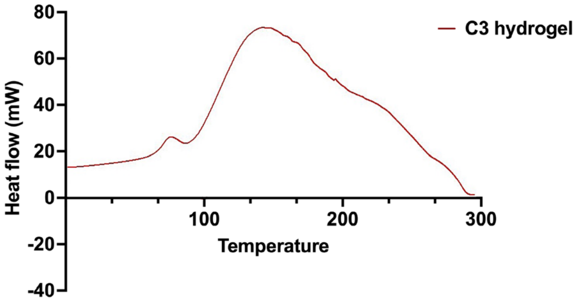


Fig. 2 DCS curve of C3 cellulose hydrogel

Differential scanning calorimetry

The DSC curve of C3 cellulose hydrogel is shown in Fig. 2. The DSC curve of C3 cellulose hydrogel shows a high and broad endothermic peak due to the presence of both cellulose and CMC. The peak > 290 °C is ascribed to dehydration or decomposition of cellulose. Meanwhile, CMC also gives a broad peak (170 °C < T > 260 °C), attributed to the lower thermal stability of CMC [28, 29]. Furthermore, this endothermic peak is also attributed to breaking hydrogen bonds [29], as the hydrogels are formed by intermolecular hydrogen bonds due to the presence of -COOH in their structure.

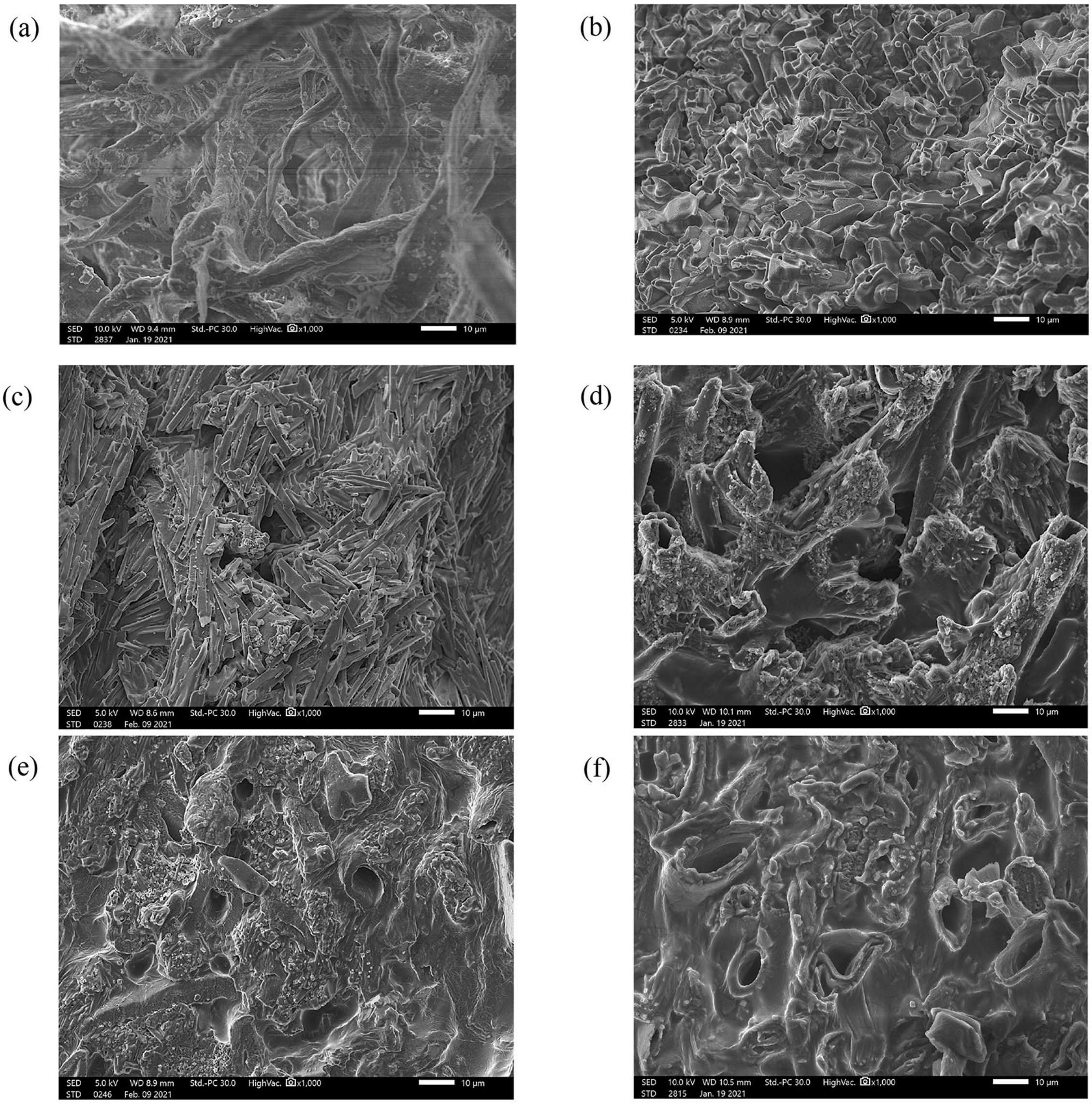


Fig. 3 FESEM images of **a** cellulose fibres, and ECH-crosslinked cellulose hydrogels of varying cellulose concentrations and 1.75% (w/v) CMC: **b** C1, **c** C2, **d** C3, **e** C4 and **f** C5 cellulose hydrogels. Magnification × 1000

Morphology and swelling behavior of cellulose hydrogel

The FESEM images of cellulose fibers and cellulose hydrogels are shown in Fig. 3. The cellulose fibers (Fig. 3a) appeared as long fibers with randomly flat shapes. The microstructure of the successfully synthesized cellulose hydrogels at different % w/v of cellulose was observed and compared in Fig. 3b–f.

The cellulose hydrogels of different compositions were further characterized for their swelling behaviors (Fig. 4a, b). C3 cellulose hydrogel consisting of 3% (w/v) cellulose and 1.75% (w/v) CMC showed the highest swelling percentage of 2055% among all formulations (Fig. 4a). The high swelling percentage of C3 cellulose hydrogel could be attributed to the highly porous microstructure with multiple channels and interconnected capillaries (Fig. 3d), thus facilitating the diffusion of water molecules into the cellulose hydrogel network [30, 31]. In the freeze–thaw preparative step, phase separation in the polymer solution occurred, allowing the formation of porous C3 assembly [32, 33]. For C4 and C5 cellulose hydrogels, reduced swelling ratios were observed, suggesting that higher cellulose content in the hydrogels promotes polymer chain entanglement via intra and inter-molecular hydrogen bonds [33]. Low swelling

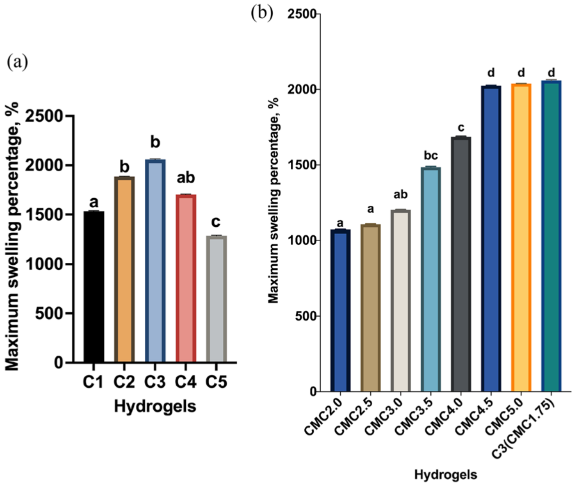


Fig. 4 Maximum swelling percentage of cellulose hydrogels consisted of different % (w/v) of (a) cellulose fibers and (b) CMC. Different letters indicate significant differences ($p \leq 0.05$) between hydrogels. Error bars show the standard deviations from triplicates ($n = 3$)

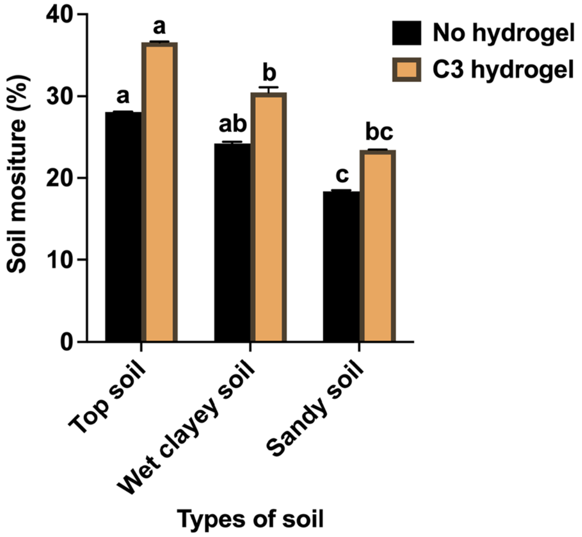


Fig. 5 Comparison of soil moisture with different types of soil using C3 cellulose hydrogel. Different letters indicate significant differences ($p \leq 0.05$) between soils. Error bars were calculated from the standard deviations from triplicates ($n = 3$)

percentages in distilled water were also observed in C1 and C2 hydrogels, due to the weaker hydrogels formed from lower cellulose concentrations, in which irregular granular surfaces and rod-like morphology were observed in their FESEM micrographs (Fig. 3b, c).

Figure 4b shows the maximum swelling percentage of cellulose hydrogels synthesized at different concentrations of CMC. In general, the swelling percentage increases with the CMC concentrations on account of its polyelectrolyte properties [34]. The hydrogel swelling could also be influenced by the presence of ions in the system. Due to the increasing amount of Na^+ ions at high concentrations of CMC (CMC4.5 and CMC5.0, Fig. 4b), no substantial changes in their water absorption were observed [35].

Herein, the swelling behavior in our cellulose hydrogels could be described using the Flory–Rehner theory, in which the equilibrium volume of the cellulose hydrogel networks is affected by the balance between the osmotic pressure and the elastic restorative force [36].

Effects of cellulose hydrogel on soil moisture

The soil moisture of three types of soils is presented in Fig. 5. The addition of the C3 cellulose hydrogel in the dried soils showed a substantial increase in the moisture content and retained the moisture for 7 days before the hydrogel dried up. The highest soil moisture was recorded in the topsoil up to 36.5% after 7 days, followed by wet clayey soil (30.1%) and sandy soil (23.4%). Topsoil showed 26.62% increased with C3 cellulose hydrogel than that of without C3 cellulose hydrogel, while wet clayey soil and sandy soil showed 22.13% and 23.6% percentage increase. Compared to the topsoil, the lower moisture content in the wet clayey soil was most likely attributed to the slow infiltration of water from cellulose hydrogel through the compact soil profile of clayey soil with small particle size. It is expected that the lowest soil moisture content was observed in the sandy soil at 23.4% in presence of the C3 hydrogel. Typically, the moisture of sandy soil ranges between 3 and 10% from the driest condition to the wettest condition [37]. These findings indicate that the presence of C3 cellulose hydrogel enhances the moisture content in soils and reduces water transpiration. Application of C3 cellulose hydrogel in soil potentially reduces farming irrigation frequencies, and enhanced the survival of plants during drought seasons and thus crop yield.

Loading and release profiles of urea in water and soils

Based on the analysis reported in this work as included in the previous section, optimal formulation to prepare cellulose hydrogel was observed in C3 composition. Therefore, C3 composition was further selected for the fabrication of urea-loaded cellulose hydrogels.

The urea loading capacity of C3 cellulose hydrogel is shown in Fig. 6a. The urea loading for C3 cellulose hydrogel was rapid and equilibrium was achieved within 48 h. The maximum loading capacity of urea (N-nutrient content) of C3 cellulose hydrogel was 0.51 g/g. Rapid encapsulation of urea by C3 cellulose hydrogel was due to the high initial concentration gradient of urea in the water

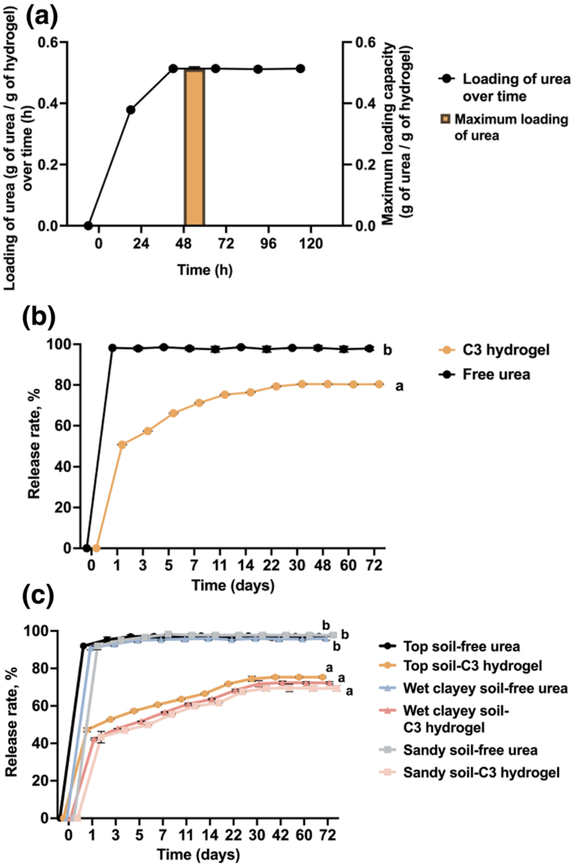


Fig. 6 **a** Loading capacity of C3 cellulose hydrogel. Error bars were calculated from the standard deviations of triplicates ($n = 3$). **b** Release behavior of free urea and C3 cellulose hydrogel in water. Different letters indicate significance differences ($p \leq 0.05$) between samples. **c** Release behavior of urea and C3 cellulose hydrogel in three types of soils. Different letters indicate significance differences ($p \leq 0.05$) between samples in different soils. Error bars were calculated from the standard deviations from triplicates ($n = 3$)

and the facile diffusion of urea into the porous structures of C3 cellulose hydrogel [38].

Urea release profiles of C3 cellulose hydrogel in water and soils are shown in Fig. 6b, c, respectively. In control studies, 98.7% free urea was released in water within 1 day (Fig. 6b). In contrast, more controlled and slower release of urea from C3 cellulose hydrogel in water was observed, with only 50.77%, 71.26% and 80.46% release after 1 day, 1 week and 1 month, respectively. "Burst release" was observed at the initial stage of the studies (within 1 h) for all types of fertilizer due to the high initial concentration gradient of loaded urea in the cellulose hydrogels and the release of urea near the surface [39].

The controlled-release behaviors of free urea and C3 cellulose hydrogel in different soils were further investigated to evaluate the potential application of C3 cellulose hydrogel for precision agriculture (Fig. 6c). While more than 95% free urea (control) was released in topsoil, wet clayey soil and sandy soil within 1 week, the urea release by C3 cellulose hydrogels in those soil samples was significantly reduced to below 60% after 7 days. For the wet clayey soils, the N-nutrient content released was 42.14% (1 day), 56.32% (1 week) and 73.37% (1 month), which were the lowest % release among all the types of soil added with C3 cellulose hydrogels. Meanwhile, in topsoil, the N-nutrient release in C3 cellulose hydrogels released 47.70%, 56.34% and 74.71% within 1 day, 1 week and 1 month, respectively. A similar urea release trend was demonstrated by C3 cellulose hydrogels in sandy soils, with 45.98%, 55.17% and 71.84% urea release in the soils at 1 day, 1 week and 1 month, respectively.

Table 2 Parameters of diffusion Korsmeyer and Peppas model for urea release from C3 cellulose hydrogels in water and soil

Kinetic model	Parameter	Water	Topsoil	Wet clayey Soil	Sandy soil
Korsmeyer–Peppas	R^2	0.9565	0.9734	0.9935	0.9575
	n	0.1452	0.1284	0.1245	0.2574
	K_{KP}	1.7103	1.653	1.6727	1.3608

The possible N-nutrient release mechanism of C3 cellulose hydrogels involves a combination of absorption, dissolution and diffusion [39]. High urea release (> 70%) from C3 cellulose hydrogels within 1 month might be attributed to the excellent hydrophilic properties of the extracted cellulose from blended wastepaper and CMC polyelectrolyte, facilitating water absorption of the hydrogel networks from the soils, causing the hydrogels to swell. The dynamic exchange of free water is expected between the cellulose hydrogels and the urea solution [8, 10]. This gradually enhances the urea dissolution within the hydrogels and speeds up the diffusion of urea from the porous hydrogels to the surroundings. Differences in the osmotic pressure between the cellulose hydrogels and their surroundings are the main driving forces of the N-nutrient release of C3 cellulose hydrogels. Based on our preliminary findings, the release profiles of urea from C3 cellulose hydrogels in water and various soil conditions (< 80%) within 30 days show promising compliance with the standards of slow-release fertilizers of the Committee of European Normalization (CEN) [10, 40].

Kinetics studies of urea release

The kinetic model proposed by Korsmeyer–Peppas was applied to analyze the urea release behavior of C3 cellulose hydrogels in both water and soils [30, 38]. The kinetics release parameters in water and soils were calculated based on Eq. 3 (Table 2) and plotted in Fig. 7a, b, respectively.

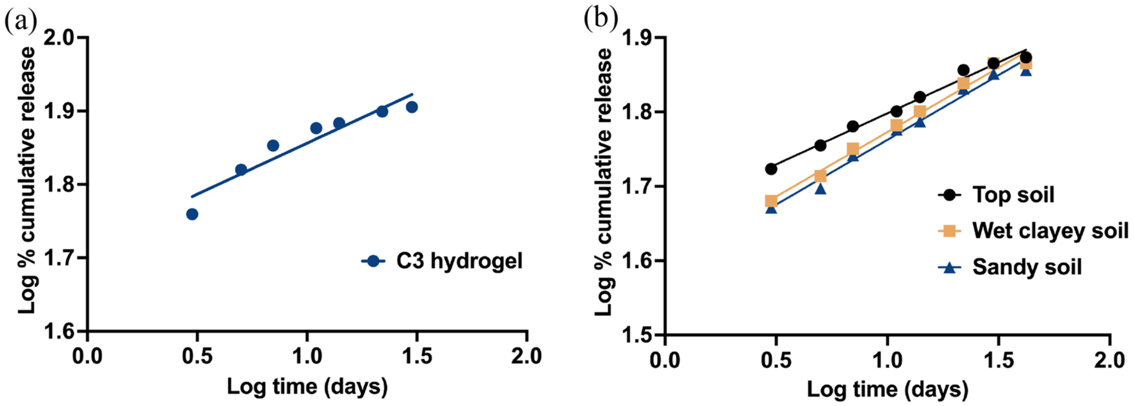


Fig. 7 Korsmeyer and Peppas model of urea release from C3 cellulose hydrogels in (**a**) water and (**b**) three types of soils

It can be deduced from Table 2 that the Fickian diffusion model is the most suitable to describe the control-release mechanism of fertilizer in water and soils ($R^2 > 0.85$ and $n < 0.45$). This indicates that the fertilizer release of C3 cellulose hydrogels was driven by diffusion through their pores and channels which was influenced by the concentration gradient and degree of swelling of the hydrogels [34, 41, 42].

Conclusion

Cellulose hydrogels were successfully prepared from varying ratios of cellulose fibers derived from paper wastes, CMC and ECH, using inexpensive and biodegradable materials. FTIR result confirmed that an ECH-crosslinked cellulose hydrogel was performed successfully. Cellulose hydrogels based on C3 composition exhibited porous networks with a high swelling ability as well as enhanced soil moisture retention soils, promising its utilization in sandy soils, as well as arid and semi-arid land. C3 cellulose hydrogel showed favorable performance as a controlled-release fertilizer based on its high loading capacity of urea as the model fertilizer and controlled release behavior in different types of soil. Moreover, the fertilizer release kinetic studies revealed that C3 cellulose hydrogel corresponds to the Korsmeyer–Peppas kinetic model in both water and soil media. Our findings suggested the high potential of C3 cellulose hydrogel possesses high potential as an eco-friendly and cost-effective slow-release fertilizer improve the utilization efficiency of fertilizer and water conservation in agricultural applications.

Acknowledgements
The authors would like to thank the Sarawak Research and Development Council (SRDC) (Grant No. RDCRG/CAT/2019/26; GL/F07/SRDC/02/2020) for the financial support.

Author contributions

Conceptualization: Suk-Fun Chin, Effendi Wasli, Cindy Soo-Yun Tan and Zaki Musa. Investigation and original manuscript drafting: Dayang Fazirah. Writing, reviewing, and editing: all authors. All authors read and approved the final manuscript.

Funding
Open Access funding provided by Universiti Malaysia Sarawak. This work was financially supported by the Sarawak Research and Development Council (SRDC) (Grant No. RDCRG/CAT/2019/26; GL/F07/SRDC/02/2020).

Availability of data and materials

The data sets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate

This manuscript is an original paper and has not been published in other journals. The authors agreed to keep the copyright rule.

Consent for publication

The authors revised the manuscript and agreed with the publication.

Competing interests

The authors declare no competing interests.

Author details

[1]Faculty Resource Science and Technology, Universiti Malaysia Sarawak, Malaysia, 94300 Kota Samarahan, Sarawak, Malaysia. [2]Faculty of Applied Science, Universiti Teknologi MARA, 94300 Kota Samarahan, Sarawak, Malaysia. [3]Malaysian Agricultural Research and Development Institute (MARDI), Jalan Santubong, Petra Jaya, 93050 Kuching, Sarawak, Malaysia.

Received: 6 February 2023 Accepted: 3 May 2023

References

1. Kabir SMF, Sikdar PP, Haque B, Bhuiyan MAR, Ali A, Islam MN. Cellulose-based hydrogel materials: chemistry, properties and their prospective applications. Prog Biomater. 2018;7:153–74. https://doi.org/10.1007/s40204-018-0095-0.
2. Behera S, Mahanwar PA. Superabsorbent polymers in agriculture and other application: a review. Polym Plast Technol Mater. 2019;59(4):341–56.

3. Wei P, Chen W, Song Q, Wu Y, Xu Y. Superabsorbent hydrogels enhanced by quaternized tunicate cellulose nanocrystals with adjustable strength and swelling ratio. Cellulose. 2021;28:3723–32. https://doi.org/10.1007/s10570-021-03776-z.
4. Liu M, Liang R, Zhan F, Liu Z, Niu A. Synthesis of a slow-release and superabsorbent nitrogen fertilizer and its properties. Polym Adv Technol. 2006;17(6):430–8.
5. Shen X, Shamshina JL, Berton P, Gurau G, Rogers RD. Hydrogels based on cellulose and chitin: fabrication, properties, and application. Green Chem. 2016;18(1):53–75.
6. Zainal SH, Mohd NH, Suhaili N, Anuar FH, Lazim AM, Othaman R. preparation of cellulose-based hydrogels: a review. J Mater Res Technol. 2021;10:935–52. https://doi.org/10.1016/j.jmrt.2020.12.012.
7. Chin SF, Romainor AN, Pang SC, Lihan S. Antimicrobial starch-citrate hydrogel for potential applications as drug delivery carriers. J Drug Deliv Sci Technol. 2019. https://doi.org/10.1016/j.jddst.2019.101239.
8. Chin SF, Jimmy FB, Pang SC. Size controlled fabrication of cellulose nanoparticles for drug delivery applications. J Drug Deliv Sci Technol. 2018;43:262–6. https://doi.org/10.1016/j.jddst.2017.10.021.
9. Arbona V, Iglesias DJ, Jacas J, Primo-Millo E, Talon M, Gomez-Cadenas A. Hydrogel substrate amendment alleviates drought effects on young citrus plant. Plant Soil. 2005;270:73–82. https://doi.org/10.1007/s11104-004-1160-0.
10. Olad A, Zebhi H, Slarai D, Mirmohseni A, Tabar AR. Slow-release NPK fertilizer encapsulated by carboxymethyl cellulose-based nanocomposite with the function of water retention in soil. Mater Sci Eng C Mater Biol Appl. 2018;90:333–40. https://doi.org/10.1016/j.msec.2018.04.083.
11. Abd El-Rehim HA. Characterization and possible agricultural application of polyacrylamide/sodium alginate crosslinked hydrogels prepared by ionizing radiation. J Appl Polym Sci. 2006;101(6):3572–80.
12. Elbarbary AM, Ghobashy MM. Controlled release fertilizers using superabsorbent hydrogel prepared by gamma radiation. Radiochim Acta. 2006;105(10):1–12.
13. Savci S. Investigation of effect of chemical fertilizers on environment. APCBEE Procedia. 2012;1:287–92. https://doi.org/10.1016/j.apcbee.2012.03.047.
14. Hashim MMA, Yusop MK, Othman R, Wahid SA. Field evaluation of newly-developed controlled release fertilizer on rice production and nitrogen uptake. Sains Malays. 2017;46(6):925–32.
15. Versino F, Urriza M, Garcia MA. Eco-compatible cassava starch films for fertilizer controlled release. Int J Biol Macromol. 2019;134:302–7. https://doi.org/10.1016/j.ijbiomac.2019.05.037.
16. Lawrencia D, Wong SK, Low DYS, Goh BH, Goh JK, Ruktanonchai UR, Soottitantawat A, Lee LH, Tang SY. Controlled release fertilizers: a review on coating materials and mechanism of release. Plants. 2021;10(2):238–63.
17. Lin Q, Chang J, Gao M, Ma H. Synthesis of magnetic epichlorohydrin cross-linked carboxymethyl cellulose microspheres and their absorption behavior for methylene blue. J Environ Sci Health A. 2017;52(2):106–16.
18. Chin SF, Jong SJ, Yeo YJ. Optimization of cellulose-based hydrogel synthesis using response surface methodology. Biointerface Res Appl Chem. 2021;12(6):7136–46.
19. Zhang H, Yang M, Luan Q, Tang H, Huang F, Xiang X, Yang C, Bao Y. Cellulose anionic hydrogels based on cellulose nanofibers as natural stimulants for seed germination and seedling growth. J Agric Food Chem. 2017;65(17):3785–91.
20. Salleh KM, Zakaria S, Sajab MS, Gan S, Chia CH, Jaafar SNS, Amran UA. Chemically crosslinked hydrogel and its driving force towards superabsorbent behaviour. Int J Biol Macromol. 2018;118:1422–30. https://doi.org/10.1016/j.ijbiomac.2018.06.159.
21. Alam MN, Islam MS, Christopher LP. Sustainable production of cellulose-based hydrogels with superb absorbing potential in physiological saline. J Phys Chem Lett. 2019;4(5):9419–26. https://doi.org/10.1021/acsomega.9b00651.
22. Kirchmajer DM, Watson CA, Ranson M, Panhuis MIH. Gelapin, a degradable genipin cross-linked gelatin hydrogel. RSC Adv. 2013;3(4):1073–81.
23. Li X, Li Q, Xu X, Su Y, Yue Q, Gao B. Characterization, swelling and slow-release properties of a new controlled release fertilizer based on wheat straw cellulose hydrogel. J Taiwan Inst Chem Eng. 2016;60:564–72. https://doi.org/10.1016/j.jtice.2015.10.027.
24. Liu X, Liao J, Song H, Yang Y, Guan C, Zhang Z. A biochar-based route for environmentally friendly controlled release of nitrogen: Urea-loaded biochar and bentonite composite. Sci Rep. 2019;9(9548):1–12.
25. Rossi AM, Villareal M, Juarez MD, Samman NC. Nitrogen contents in food: a comparison between the kjeldahl and hach methods. J Argent Chem Soc. 2004;94(4–6):99–108.
26. Hach CC, Bowden BK, Kopelove AB, Brayton SV. More powerful peroxide Kjeldahl Digestion Method. J Assoc Off Anal Chem. 1987;70(5):783–7.
27. Korsmeyer RW, Gurny R, Doelker E, Buri P, Peppas NA. Mechanism of soliute release from porous hydrophilic polymers. I J Pharm. 1983;15(1):25–35.
28. El-Sherbiny IM, Salama A, Sarhan AA. Grafting study and antifungal activity of a carboxymethyl cellulose derivative. Int J Polym Mater. 2009;58(9):453–67.
29. El-Sakhawy M, Tohamy HAS, Salama A, Kamel S. Thermal properties of carboxymethyl cellulose acetate butyrate. Cellul Chem Technol. 2019;53(7–8):667–75.
30. Chetouani A, Elkolli M, Bounekhel M, Benachour D. Physicochemical characterization of gelatin-cmc composite edibles films from polyion-complex hydrogels. J Chil Chem Soc. 2014;59(1):2279–83.
31. Voon LK, Cem S, Pang SC, Chin SF. Highly porous cellulose beads of controllable sizes derived from egenerated cellulose of printed paper wastes. Mater Lett. 2016;164:264–6. https://doi.org/10.1016/j.matlet.2015.10.161.
32. Jampi ALW, Chin SF, Wasli ME, Chia CH. Preparation of cellulose hydrogel from sago pith waste as a medium for seed germination. J Phys Sci. 2021;32(1):13–26.
33. Kareem SA, Dere I, Gungula DT, Andrew FP, Saddiq AM, Adebayo EF, Tame VT, Kefas HM, Joseph J, Patrick DO. Synthesis and characterization of slow-release fertilizer hydrogel based on hydroxy propyl methyl cellulose, polyvinyl alcohol, glycerol and blended paper. Gel. 2021;7(4):262–78.
34. Wei H, Wang H, Chu H, Li J. Preparation and characterization of slow-release and water-retention fertilizer based on starch and halloysite. Int J Biol Macromol. 2019;133:1210–8. https://doi.org/10.1016/j.ijbiomac.2019.04.183.
35. Quesada-Perez M, Maroto-Centeno JA, Forcada J, Hidalgo-Alvarez R. Gel swelling theories: the classical formalism and recent approaches. Soft Matter. 2011;7(22):10536–47.
36. Brandt MJ, Johnson KM, Elphinston AJ, Ratnayaka DD. Hydrology and surface supplies Twort's water supply. 7th ed. Butterworth Heinemann: IWA publishing; 2017.
37. Khoo SM, Moghaddam PN, Fareghi AR, Movagharnezhad N. Synthesis of a cellulose-based hydrogel network: characterization and study of urea fertilizer slow release. J Appl Polym Sci. 2016;133(5):42935–44.
38. Liang Y, Cao X, Zhao L, Xu X, Harris W. Phosphorus release from dairy manure, the manure-derived biochar, and their amended soil: effects of phosphorus nature and soil property. J Environ Qual. 2014;43(4):1504–9.
39. Ding H, Zhang YS, Li WH, Zhen XZ, Wang MK, Tang LN, Chen DL. Nutrients release from a novel gel-based slow/controlled release fertilizer. Appl Environ Soil Sci. 2016;2016:1–13. https://doi.org/10.1155/2016/2013463.
40. Trenkel ME. Slow-and controlled-release and stabilised fertilizers: an option for enhancing nutrient use efficiency in agriculture. 2nded. Paris: International Fertiliser Industry Association (IFA); 2010.
41. Hermida L, Agustian J. Slow release urea fertilizer synthesized through recrystallization of urea incorporating natural bentonite using various binders. Environ Technol Innov. 2019;13:113–21. https://doi.org/10.1016/j.eti.2018.11.005.
42. Bi S, Barinelli V, Sobkowicz MJ. Degradable controlled release fertilizer composite prepared via extrusion: fabrication, characterization and release mechanism. Polymers. 2020;12(2):301–15.

Publisher's Note