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Research Article

Green Metrics Evaluation of a Sustainable Ultrasonication-Assisted Routes to Cellulose Ethers from Banana (*Musa balbisiana* Colla) Blossom Cellulose

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Abstract

To promote sustainable chemical transformation and environmentally friendly approaches, balance economic feasibility and ecological protection are essential. In this study, cellulose ethers, such as methyl cellulose (MC), carboxymethyl cellulose (CMC), and hydroxypropyl cellulose (HPC), were successfully synthesized from banana (*Musa balbisiana* Colla) blossom cellulose (BBC) through the Williamson ether reaction using a green ultrasonication method at ambient temperature for 30–40 mins. A simple ultrasonic bath system was employed to facilitate efficient cavitation under mild and accessible operating conditions. The yields of MC, CMC, and HPC were 96.5, 98.3, and 97.5%, with corresponding degrees of substitution (DS) of 2.00, 0.71, and 0.86, respectively. An environmental assessment of each synthesis was conducted using Environmental Assessment Tool for Organic Syntheses (EATOS) software and Andraos worksheets, evaluating substrates, solvents, products, and by-products. The results demonstrated that this method offers significant advantages, including low environmental impact, minimal chemical consumption, and near-ideal green chemistry parameters, such as atom economy (AE), reaction yield, stoichiometric factor (1/SF), material recovery (MRP), and reaction mass efficiency (RME), outperforming conventional methods reported in the literature.

Keywords: carboxymethyl cellulose (CMC), cellulose ethers, green metrics, hydroxypropyl cellulose (HPC), methyl cellulose (MC), ultrasonication

1. INTRODUCTION

Cellulose, the most abundant natural polymer, are primarily found in plant cell walls [1]-[7]. Banana blossoms are among the plant sources with a particularly high cellulose content [8]. As a highly accessible and renewable raw material, cellulose is an excellent candidate for producing value-added materials for diverse industrial applications, including as a food additive [9]. Its structure is composed of β-1,4-linked glucopyranose chains stabilized by extensive intra- and inter-molecular hydrogen bonding, which contributes to its high crystallinity and insolubility in water and most organic solvents. To overcome these limitations and expand its applicability, cellulose can be chemically modified into water-soluble derivatives such as methyl cellulose (MC), carboxymethyl cellulose (CMC), and hydroxypropyl cellulose (HPC) [10]. These derivatives are predominantly synthesized through the Williamson ether synthesis, an S_N2 nucleophilic substitution reaction between a cellulose alkoxide and an alkyl halide (or epoxide for HPC). The alkoxide intermediate is generated by deprotonation of hydroxyl groups with a strong base, such as sodium hydroxide [11]-[12].

Some cellulose ether syntheses have traditionally employed conventional heating methods. While effective, these methods present significant drawbacks for industrial-scale applications, primarily due to their prolonged reaction times and high energy consumption [13-15]. An alternative is the ultrasonically assisted method, which offers a more environmentally friendly and sustainable approach [11-16]. Ultrasonication provides several advantages, including simplicity, lower cost, shorter reaction times, and higher efficiency compared to conventional heating, water bath shaking, or magnetic stirring [14][17][18]. This technique has been shown to improve synthesis outcomes, as demonstrated in the preparation of CMC from office paper waste, which required only 30 min and yielded a high degree of substitution (DS) [15][19]. Owing to these benefits, ultrasonication is consistent with the principles of green chemistry and represents a promising strategy for cellulose ether synthesis.

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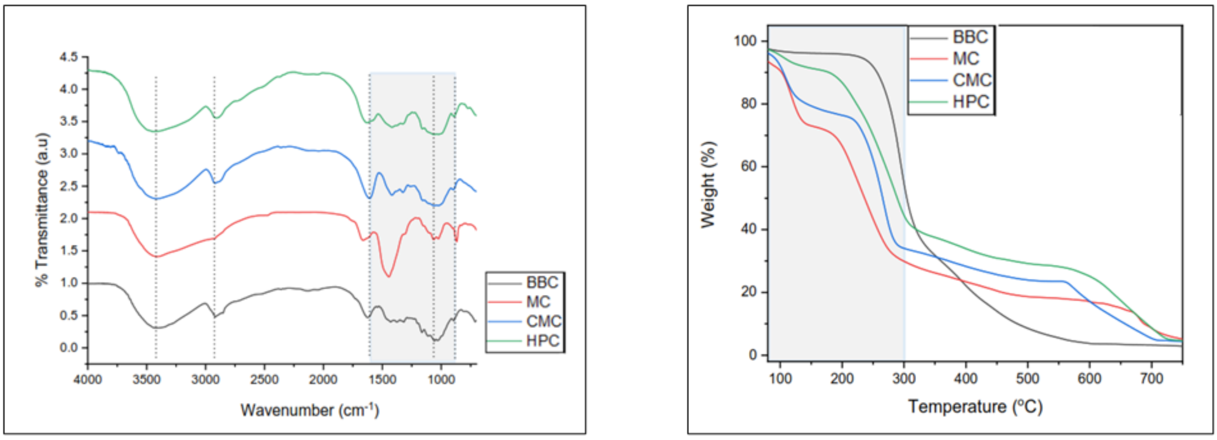


(a) (b)

Figure 1. (a) FTIR and (b) TGA spectra of the cellulose, MC, CMC and HPC.

Green chemistry is an approach designed to minimize or eliminate the use and generation of hazardous substances, thereby promoting safer and more environmentally friendly chemical processes [20][21]. This concept was developed to prevent pollution at its source and reduce reliance on harmful materials, supporting sustainable development in both laboratory and industrial settings [22][23]. The framework of green chemistry is defined by 12 guiding principles, such as waste prevention, atom economy, less hazardous chemical synthesis, designing safer chemicals, safer solvents and auxiliaries, energy efficiency, use of renewable feedstocks, reduction of derivatives, catalysis, design for degradation, real-time analysis, and inherently safer chemistry for accident prevention [20]-[24]. While not all principles can be applied simultaneously, their integration depends on the synthesis route and materials used. The degree to which a process adheres to these principles can be evaluated using green metrics, which provide a systematic assessment of its environmental performance.

Green metrics offer a quantitative framework for assessing chemical processes against the principles of green chemistry, enabling the evaluation of environmental performance and the identification of areas for improvement. Effective metrics should be objective, simple, clear, and quantifiable [25]. A process can only be considered “greener” if its environmental impact is demonstrably lower than that of conventional methods, as determined by green metric assessment [26]. Several tools have been developed for this purpose, including Environmental Assessment Tool for Organic Syntheses (EATOS) software [27] and the Andraos spreadsheet [19][22]. These approaches have been applied to evaluate eco-friendliness in the synthesis of various compounds [19][22][27]-[29]. However, their application in evaluating ultrasonication-assisted synthesis of cellulose ethers from agricultural waste biomass remains limited. Therefore, the novelty of this study lies in (*i*) the utilization of banana (*Musa balbisiana* Colla) blossom as an underexplored cellulose source for the simultaneous synthesis of cellulose ethers (MC, CMC, and HPC), (*ii*) the implementation of a mild ultrasonication-assisted method to enhance synthesis efficiency, and (*iii*) the integrated evaluation of environmental performance using complementary green metrics tools (EATOS and the Andraos spreadsheet). This combined approach provides a more comprehensive understanding of both the technical and environmental advantages of the process compared to previously reported methods.

2. MATERIALS AND METHODS

2.1. Materials

The reactions were conducted using an ultrasonic cleaner (DSA50-GLI) operating at 50 W power and 40 kHz frequency. This ultrasonic bath system was selected because it is widely available, cost-effective, and capable of generating adequate cavitation for efficient cellulose etherification under mild and energy-efficient conditions. The chemicals used are banana (*Musa balbisiana Colla*) blossom cellulose (BBC) (obtained through isolation), sodium hydroxide (Merck), dichloromethane (Merck), monochloroacetic acid (Sigma-Aldrich), propylene oxide (Sigma-Aldrich), glacial acetic acid (Merck), methanol (Merck), and ethanol (Merck). All chemicals were of analytical grade and used without further purification.

2.2. Methods

2.2.1. Synthesis of Methyl Cellulose (MC) by Ultrasonication Method

The synthesis of MC involved two steps: alkalization and methylation [11]. BBC (0.4 g) was dispersed in 50% (w/v) NaOH solution (10 mL) and stirred for 1 h at room temperature to form alkali cellulose. Acetone (8 mL) was then added as the solvent, followed by CH_2Cl_2 (6 mL) as the methylating agent. The mixture was subjected to ultrasonic irradiation for 30 min at room temperature. The resulting product was neutralized with 10% (v/v) acetic acid, washed with acetone to remove by-products, filtered, and dried at room temperature to obtain MC.

2.2.2. Synthesis of Carboxymethyl Cellulose (CMC) by Ultrasonication Method

BBC (0.4 g) was treated with 30% (w/v) NaOH aqueous solution (2 mL) and stirred for 1 h at room temperature. Subsequently, isopropanol (8 mL) was added as the solvent along with monochloroacetic acid (1 g) as the etherification agent. The reaction mixture was subjected to ultrasonication for 40 min at room temperature [15]. The product was neutralized with glacial acetic acid and washed three times with 80% ethanol/water and once with methanol to remove unreacted reagents and salts. The suspension was filtered and dried at room temperature to yield CMC [11].

2.2.3. Synthesis of Hydroxypropyl Cellulose (HPC) by Ultrasonication Method

HPC synthesis was performed in two steps: alkalization and etherification [11]. BBC (0.4 g) was suspended in 10% (w/v) NaOH solution (2 mL) and isopropanol (8 mL), followed by stirring for 1 h

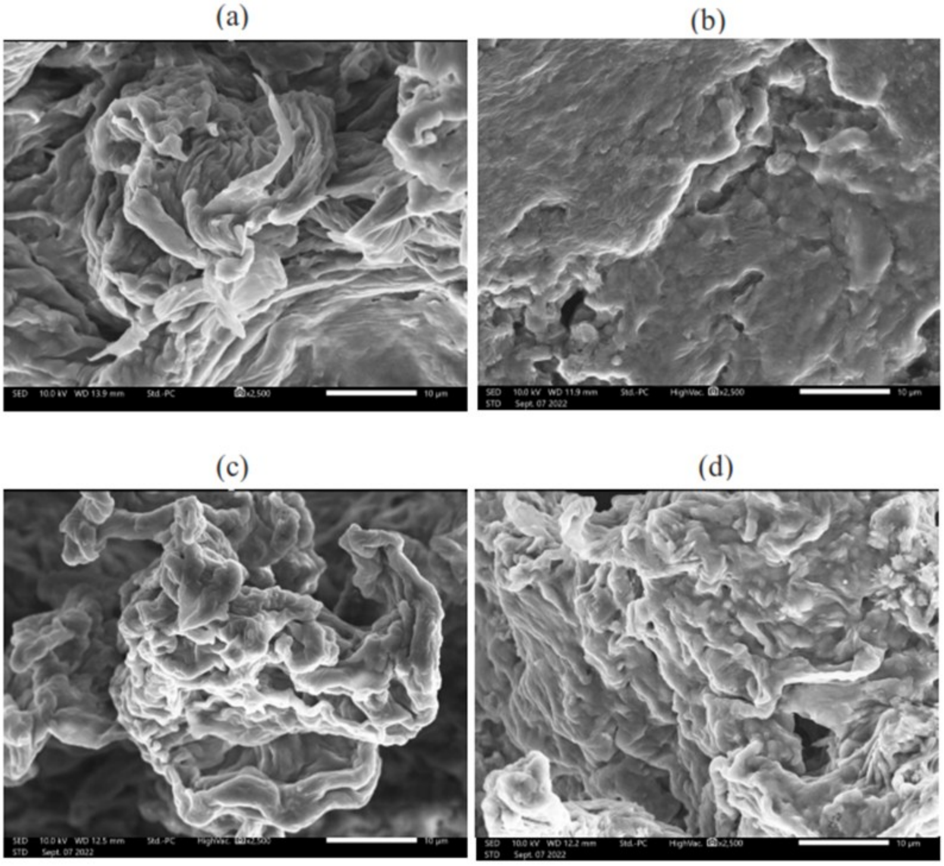
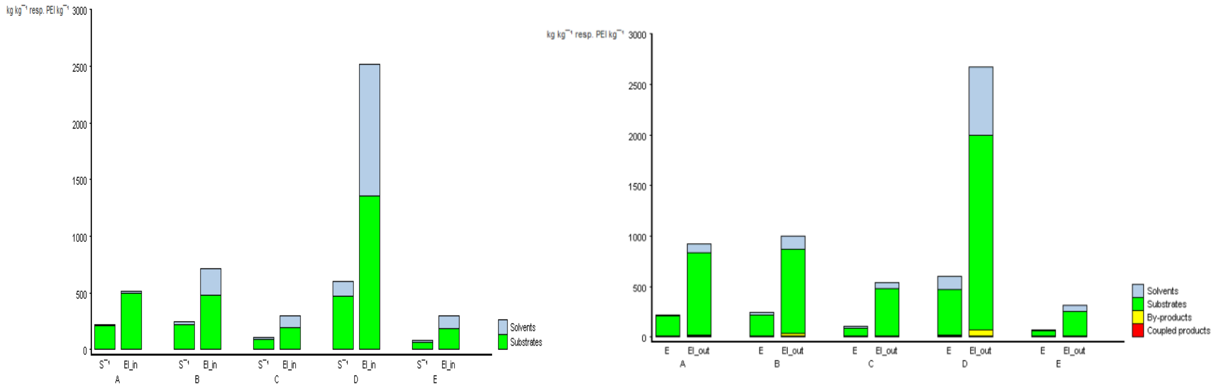


Figure 2. SEM images of the (a) cellulose, (b) MC, (c) CMC, and (d) HPC.

Table 1. Comparison of MC synthesis from various literature.

Code	Cellulose Sources	Method	Synthesis Condition		
			Temperature (°C)	Time (h)	Yield (%)
A	Sugarcane bagasse [41]	Heating	50	3	44.9
B	Corn cob [42]	Heating	50	3	20.7
C	Bacteria [43]	Heating	50	5	66.0
D	Pineapple stump [44]	Reflux	60	6	9.2
E	Banana Blossom	Ultrasonication	25	0.5	97.0



(a)

(b)

Figure 3. Comparison of (a) S^{-1} dan EI_{in} values from synthesis MC with various literatures using EATOS software, and E-Factor dan EI_{out} values from synthesis MC with various literatures using EATOS software.

at room temperature. Propylene oxide (1.5 mL) was then added as the hydroxypropylating agent, and the mixture was subjected to ultrasonic irradiation for 30 min. The reaction was terminated by neutralization with 10% (v/v) acetic acid, followed by washing with 80% ethanol to remove by-products. The final suspension was filtered and dried at room temperature to obtain HPC. The BBC, MC, CMC and HPC were analysed by Fourier Transform Infrared (FT-IR) spectroscopy, thermogravimetric analysis (TGA), and scanning electron microscope (SEM).

2.2.4. Environmental Assessment

The environmental assessment of each cellulose ether synthesis (MC, CMC, and HPC) was conducted using the freeware EATOS. Required input data were obtained from the Material Safety Data Sheets (MSDS) of Sigma-Aldrich, Merck, and PubChem, as well as relevant literature. The EATOS analysis quantified four indices: mass index (S^{-1}), environmental index input (EI_{in}), environmental factor (E-factor), and environmental index output (EI_{out}). These indices incorporate stoichiometry, material consumption, costs, risk, toxicity, ecotoxicity, and accumulation profiles. The S^{-1} value represents the quantity of chemicals used per unit product, while EI_{in} reflects the environmental impact cost of raw materials. The E-factor reflects the quantity of waste produced. The EI_{out} parameter, expressed as potential environmental impact (PEI), reflects the environmental burden of the generated waste. Additionally, an environmental evaluation was carried out using the Andraos spreadsheet, which evaluates all chemical inputs, including substrates, solvents, products, and by-products. Input data consisted of reaction stoichiometry, chemical consumption, and the formation of water (H_2O) and sodium chloride (NaCl) as by-products. The calculated green metrics are atom economy (AE), reaction yield (Rxn Yield), stoichiometric factor (1/SF), material recovery parameter (MRP), and reaction mass efficiency (RME). Each cellulose ether synthesis pathway (MC, CMC, and HPC) was compared with prior studies to evaluate its relative

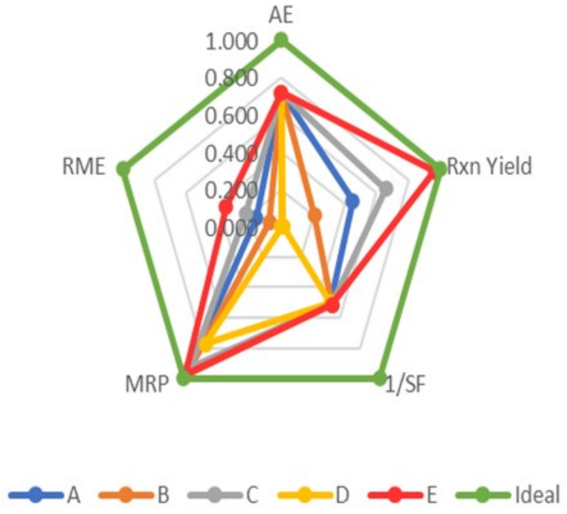


Figure 4. Measurement results of AE, RME, Rxn yield, 1/SF, and MRP values of MC synthesis using Andraos spreadsheet.

environmental sustainability. The previous studies were designated as codes A, B, C, and D, while the present reaction condition is referred to as code E.

3. RESULTS AND DISCUSSIONS

3.1. Synthesis of MC, CMC and HPC via Ultrasonication Method

The cellulose derivatives were successfully synthesized using an ultrasonication-assisted method in accordance with green chemistry principles. The high yields obtained (97–98%) demonstrate the efficiency of the process and reproducibility of the method. This approach relies on acoustic cavitation, where ultrasonic waves generate and collapse microbubbles in the liquid phase, producing localized high temperatures and pressures that accelerate chemical reactions [11][30]. Notably, the use of a simple ultrasonic bath system further highlights the practicality and scalability of the method, as it enables efficient reaction enhancement under mild conditions without the need for specialized high-intensity ultrasonic equipment.

MC was synthesised in 97% yield and DS value of 2.0. The CMC was obtained in 98% yield, with DS of 0.7. The HPC was achieved in 98%, with DS of. 0.9. These DS values fall within the typical range reported for cellulose derivatives and provide quantitative confirmation of successful substitution [31]-[34]. In terms of solubility, the derivatives demonstrated polarity-dependent behaviour. The MC dissolved in DMSO. The CMC was readily soluble in water due to hydrogen bonding but insoluble in non-polar solvents [35]-[38]. The HPC dissolved in polar solvents such as water and ethanol, while remaining insoluble in non-polar solvents like *n*-hexane and chloroform. This change in solubility behaviour compared to cellulose indicates disruption of the original hydrogen-bonding network. Such solubility transitions are widely accepted as indirect evidence of chemical modification in cellulose systems. Increased degrees of substitution enhanced solubility in polar media by strengthening intermolecular interactions and disrupting native hydrogen bonding within the cellulose structure [39]-[40].

The FTIR spectra confirmed successful functionalization in all derivatives (Figure 1). Cellulose (BBC) exhibited characteristic absorption bands at ~3340 cm^{-1} (O–H stretching vibration), ~900 cm^{-1} (C–H stretching), ~1640 cm^{-1} (OH bending), and in the region of 1160–1030 cm^{-1} corresponding to C–O–C and C–O stretching vibrations of the glucopyranose backbone. For MC, additional absorption bands were observed in the 1500–800 cm^{-1} region, particularly around ~1450 cm^{-1} and ~1375 cm^{-1}, which are attributed to $-CH_3$ asymmetric and symmetric bending vibrations, respectively. The appearance of these peaks, along with increased intensity in the C–H stretching region (~2900 cm^{-1}), indicates successful methyl substitution of hydroxyl groups. In the case of CMC, distinct bands appeared at ~1600–1650 cm^{-1}

and ~1410–1450 cm^{-1}, corresponding to the asymmetric and symmetric stretching vibrations of the carboxylate (–COO$^-$) group. The presence of these peaks, which are absent in cellulose, confirms the introduction of carboxymethyl functionalities. Additionally, the increased intensity in the 1000–1100 cm^{-1} region is associated with C–O stretching of ether linkages formed during substitution. For HPC, characteristic absorptions were observed in the range of 1000–1416 cm^{-1}, particularly around ~1370 cm^{-1} (C–H bending of –CH_3 groups) and ~1050–1150 cm^{-1} (C–O–C stretching), indicating the presence of hydroxypropyl ether groups. A slight broadening of the O–H stretching band (~3340 cm^{-1}) was also observed, reflecting changes in hydrogen bonding due to substitution. FTIR spectra showed characteristic absorption bands corresponding to the introduced functional groups. Although FTIR analysis is primarily qualitative, the assignment of specific functional groups to well-defined wavenumbers, together with the measured DS values and consistent physicochemical properties, provides strong evidence for successful modification.

The TGA spectra indicated reduced thermal stability in all cellulose derivatives compared to cellulose, confirming the successful chemical modification (Figure 1). Cellulose exhibits higher thermal resistance due to its highly ordered crystalline structure and strong intermolecular hydrogen bonding network. In contrast, all derivatives showed major degradation in the range of 200–300 °C, which is associated with the introduction of substituent groups that disrupt hydrogen bonding and decrease crystallinity. HPC exhibits the highest thermal stability among the derivatives, likely due to the presence of hydroxypropyl groups that provide some flexibility while still maintaining intermolecular interactions. CMC shows moderate stability due to the presence of carboxymethyl groups introduces ionic character, which weakens thermal resistance but still retains some structural integrity. In contrast, MC demonstrates the lowest thermal stability, as methyl substitution more significantly disrupts intermolecular hydrogen bonding without providing additional stabilizing interactions.

The SEM image of MC revealed a transition from smooth cellulose fibres to a rough surface. The SEM analysis of CMC exhibited smooth yet uneven fibers, attributed to alkaline-induced structural changes. The SEM images of HPC showed rough and uneven fibers that retained their fibrillar morphology (Figure 2). While SEM does

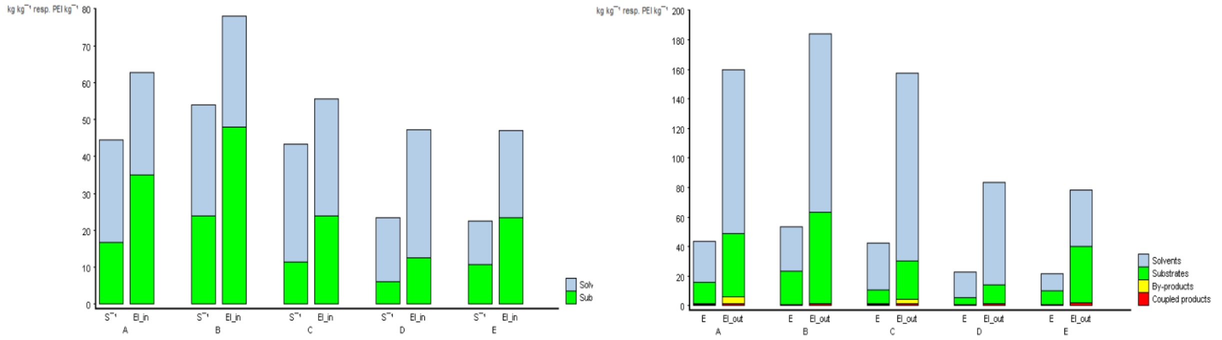
Table 2. Green metric measurement results of synthesis MC using Andraos spreadsheet.

Parameter	A	B	C	D	E	Ideal
AE	0.718	0.718	0.718	0.718	0.718	1
Rxn Yield	0.449	0.207	0.660	0.009	0.965	1
1/SF	0.504	0.514	0.514	0.503	0.514	1
MRP	0.972	0.974	0.927	0.774	0.977	1
RME	0.158	0.074	0.226	0.003	0.348	1

Table 3. Comparison of CMC synthesis from various literature.

Code	Cellulose Sources	Method	Synthesis Condition		
			Temperature (°C)	Time (h)	Yield (%)
A	Rice husk [45]	Heating	50	3	85.0
B	Rice straw [46]	Microwave	45	1	96.0
C	Office paper waste [47]	Ultrasonication	25	0.5	96.0
D	Corn cob [48]	Heating	55	3.5	98.0
E	Banana Blossom	Ultrasonication	25	0.67	98.0

Note: The power used in the ultrasonication method (code C, Ref. 15 was 18.75 W/mL. In our work (code E, ultrasonication was performed at a power of 50 W/mL and a frequency of 40 kHz.



(a)

(b)

Figure 5. Comparison of (a) S^{-1} dan EI_{in} values from synthesis CMC with various literatures using EATOS software, and (b) E-Factor dan EI_{out} values from synthesis CMC with various literatures using EATOS software.

not directly confirm chemical substitution, the observed morphological changes are consistent with structural disruption expected after derivatization. Overall, the combination of DS determination (quantitative evidence), FTIR (functional group identification), TGA (thermal behaviour), solubility analysis, and SEM (morphological changes) provides converging and internally consistent evidence of successful cellulose modification. This multi-technique approach minimizes the limitations of individual methods and is accepted for confirming cellulose derivatization.

3.2. Green Metrics Evaluation of MC

The greenness of the synthesis process of MC was assessed using EATOS software. The environmental performance of this study (synthesis E) was compared with four previously reported MC syntheses (Table 1), differing in cellulose sources, methods, and reaction conditions. Table 1 shows that MC yield is strongly influenced by both cellulose source and synthesis method. Conventional heating (codes A, B and C) produced moderate yields (20.7–66.0%), with highest value observed for bacterial cellulose. The reflux method (code D) gave the lowest yield (9.2%) despite harsher conditions, indicating limited efficiency. In contrast, ultrasonication (code E) using banana blossom achieved the highest yield (97%) under mild conditions (25 °C, 30 min). This superior performance is attributed to cavitation effects that enhance cellulose disruption, mass transfer, and reaction efficiency, consistent with trends reported in other ultrasonication-based studies**.** The EATOS analysis in Figure 3 further supports these findings by correlating synthesis conditions and yield with environmental impact metrics.

Among the evaluated routes, synthesis D (reflux method) performed the poorest, exhibiting the highest S^{-1} (592.99 kg/kg), E-factor (591.99 kg/kg), EI_{in} (2513.18 PEI/kg), and EI_{out} (2665.85 PEI/kg). This unfavorable performed is attributed to its high substrate loading, excessive acetone consumption, and low product yield, which collectively led to substantial resource use and waste generation. Syntheses A, B and C, employing conventional heating, showed intermediate performance. Their S^{-1} (101.42–237.02 kg/kg) and E-factor (100.42-236.02 kg/kg) values, together with EI_{in} (294.70–708.64 PEI/kg) and EI_{out} (530.03–995.96 PEI/kg) results, were markedly lower than those of the reflux method but still indicate considerable scope for improving process efficiency and waste minimization. In contrast, synthesis E (this study, ultrasonication-assisted) demonstrated the most favourable metrics, achieving the lowest S^{-1} (69.96 kg/kg) and E-factor (68.91 kg/kg), along with minimal EI_{in} (287.05 PEI/kg) and EI_{out} (309.03 PEI/kg). In agreement with Table 1, the superior yield obtained in synthesis E directly contributes to improved material efficiency and lower environmental burdens.

The E-factor and EI_{out} values of five MC syntheses were evaluated to assess waste generation and its associated environmental impact. Among the studied processes, synthesis D, which employed the reflux method using pineapple stump cellulose, exhibited the highest E-factor (591.99 kg input/kg product) and EI_{out} (2665.85 PEI/kg) (Table 1, Figure 3). These values resulted from the excessive use of acetone solvent and substrate, leading to a substantial amount of waste and elevated environmental impact. In contrast, syntheses A, B, and C, conducted via heating, generated lower E-factors (213.99, 236.02, and 100.42 kg input/kg product, respectively) and correspondingly reduced EI_{out} values (912.35, 995.96, and 530.03 PEI/kg). The most favorable outcome was observed in synthesis E (this study), where ultrasonication produced the lowest E-factor (68.91 kg input/kg product) and EI_{out} (309.03 PEI/kg). This improvement is attributed to the optimized balance between substrate and acetone solvent, minimizing waste and reducing environmental impact.

Further assessment was conducted using the Andraos spreadsheet (Table 2, Figure 4). The AE values remained constant across all syntheses due to the assumption of identical etherification reactions. However, synthesis D displayed the lowest values for all other parameters, primarily due to its high substrate and solvent requirements coupled with a low yield of 9.2%. In contrast, synthesis E (this study) achieved the highest values of reaction yield, 1/SF, MRP, and RME, demonstrating that ultrasonication, under the conditions of cellulose:NaOH ratio (1:25 w/v), etherification agent (6 mL), and acetone solvent (8 mL), offers the most sustainable and efficient environmental profile. Overall, the results show that ultrasonication provides a more efficient and environmentally friendly approach for MC synthesis, combining high yield, lower resource consumption, and reduced waste generation compared to conventional heating and reflux methods.

3.3. Green Metrics Evaluation of CMC

The CMC synthesis generated water (H_2O) and sodium chloride (NaCl) as by-products. To assess environmental performance, the CMC synthesis in this study was compared with four previous reports employing different cellulose sources, methods, and reaction conditions (Table 3, Figure 5). Table 3 demonstrates that both the synthesis method and

Table 4. Green metric measurement results of synthesis CMC using Andraos spreadsheet.

Parameter	A	B	C	D	E	Ideal
AE	0.785	0.785	0.785	0.785	0.785	1
Rxn Yield	0.850	0.960	0.960	0.980	0.983	1
1/SF	0.453	0.326	0.454	0.454	0.454	1
MRP	0.922	0.641	0.899	0.703	0.978	1
RME	0.279	0.158	0.308	0.246	0.342	1

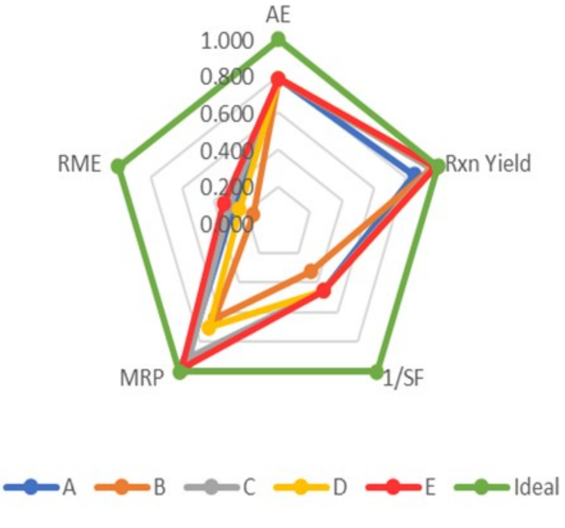
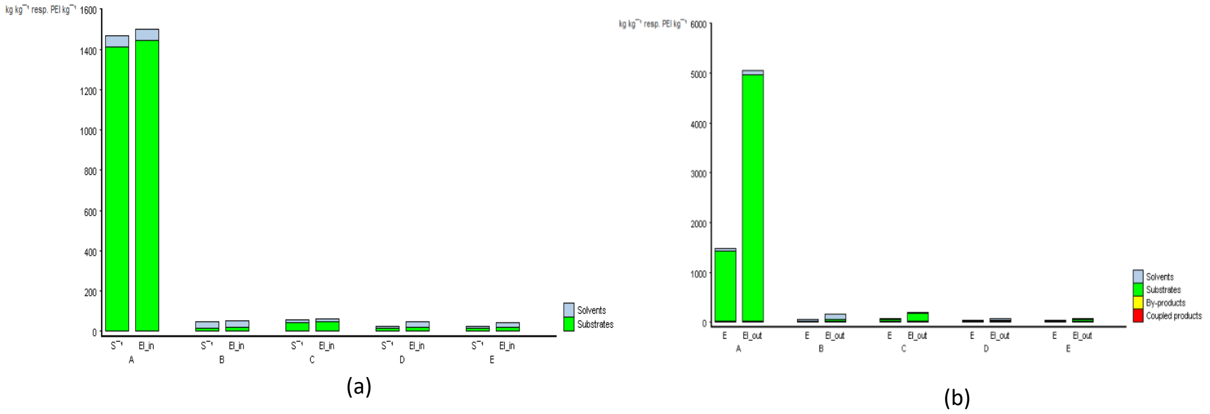


Figure 6. Measurement results of AE, RME, Rxn yield, ½ SF, and MRP values of CMC synthesis using Andraos spreadsheet.

Table 5. Comparison of HPC synthesis from various literature.

Code	Cellulose Sources	Method	Synthesis Condition		
			Temperature (°C)	Time (h)	Yield (%)
A	Sugarcane bagasse [49]	Reflux	60	6	48.0
B	Bacterial cellulose [50]	Heating	70	6	76.5
C	Paper waste [51]	Heating	55	3.5	78.3
D	Palm empty fruit bunches [52]	Heating	55	3	76.9
E	Banana Blossom	Ultrasonication	25	0.5	98.0



(a)

(b)

Figure 7. Comparison of (a) S-1 dan EI_{in} values from synthesis HPC with various literatures using EATOS software, and (b) E-Factor dan EI_{out} values from synthesis HPC with various literatures using EATOS software.

cellulose source strongly affect yield and efficiency. Conventional heating requires higher temperatures (50–55 °C) and longer times (3.0–3.5 h) but does not consistently produce high yields. In contrast, microwave and ultrasonication methods achieve higher yields (96–98%) under milder conditions and shorter times. Ultrasonication is particularly effective, operating at 25 °C within 30–40 min while maintaining high yield. Additionally, cellulose source influences performance, with agricultural residues like corn cob and banana blossom yielding up to 98%, whereas rice husk shows lower efficiency due to structural complexity. Overall, advanced methods combined with suitable raw materials enhance CMC synthesis efficiency and reduce energy consumption.

Among the five syntheses, the microwave-assisted route (synthesis B) produced the highest S^{-1} (53.76 kg input/kg product) and EI_{in} (77.81 PEI/kg), largely due to excessive solvent and substrate consumption. Syntheses A and D, both employing conventional heating, displayed lower S^{-1} (44.27 and 23.24 kg input/kg product, respectively) and EI_{in} (43.02 and 47.07 PEI/kg). In contrast, ultrasonication routes (syntheses C and E) showed improved environmental performance, with synthesis E (this study) yielding the lowest S^{-1} (22.39 kg input/kg product) and EI_{in} (46.83 PEI/kg). These results highlight ultrasonication as a greener alternative, attributed to the reduced amount, cost, and risk of chemicals used in the process.

The EATOS analysis quantified the waste produced during CMC synthesis by calculating the E-factor and the waste impact (EI_{out}), as summarized in Figure 5. Among the five syntheses, synthesis B exhibited the highest E-factor (52.76 kg input/kg product), primarily due to extensive use of isopropanol solvent, which accounted for 57% of the waste generated. Syntheses A and D, both employing conventional heating, produced lower E-factors of 43.27 and 42.02 kg input/kg product, respectively. In contrast, the ultrasonication routes (syntheses C and E) generated considerably less waste, with E-factor values of 22.24 and 21.39 kg input/kg product, respectively. Notably, synthesis E (this study) produced the least waste while also achieving the highest yield, demonstrating the superior efficiency of this method.

The EI_{out} analysis further highlighted differences in environmental impact. Synthesis B yielded the highest EIout (183.44 PEI/kg), again reflecting its heavy reliance on isopropanol. Syntheses A and D showed lower EI_{out} values (159.48 and 83.00 PEI/kg, respectively). Although syntheses C and E both used ultrasonication, synthesis C exhibited a higher EI_{out} (157.06 PEI/kg) and lower yield compared to synthesis E. The present study (synthesis E), which synthesized CMC from banana blossom cellulose, achieved the lowest EI_{out} (77.81 PEI/kg). This outcome can be attributed to the balanced use of isopropanol relative to substrate, minimizing waste and environmental impact while maintaining high efficiency (yield 98.3%).

To complement the EATOS assessment, the five syntheses were also evaluated using the Andraos spreadsheet (Table 4 and Figure 6). AE values were identical across all syntheses, consistent with the same underlying carboxymethylation reaction. However, synthesis B showed the lowest values for 1/SF, MRP, and RME, owing to the high consumption of substrate and isopropanol solvent, despite exhibiting a relatively higher Rxn yield than synthesis A. By contrast, synthesis E (this study) demonstrated the highest values for all parameters (Rxn yield, 1/SF, MRP, and RME). These results confirm that the ultrasonication method, employing a cellulose:NaOH ratio of 1:5 (w/v), monochloroacetic acid as etherifying agent (1 g), and 8 mL isopropanol solvent, provides the most favourable.

3.4. Green Metrics Evaluation of HPC

Green metrics assessment was conducted using EATOS software to evaluate all chemicals involved in HPC synthesis. Environmental performance of the present synthesis (synthesis E) was compared with four previous studies (syntheses A, B, C and D) employing different cellulose sources, methods, and reaction conditions (Table 5 and Figure 7). Table 5 shows that conventional methods, including

Table 6. Green metric measurement results of synthesis HPC using Andraos spreadsheet.

Parameter	A	B	C	D	E	Ideal
AE	0.992	0.992	0.992	0.992	0.992	1
Rxn Yield	0.480	0.765	0.783	0.769	0.975	1
1/SF	0.344	0.413	0.413	0.413	0.413	1
MRP	0.861	0.837	0.838	0.940	0.975	1
RME	0.141	0.262	0.269	0.296	0.389	1

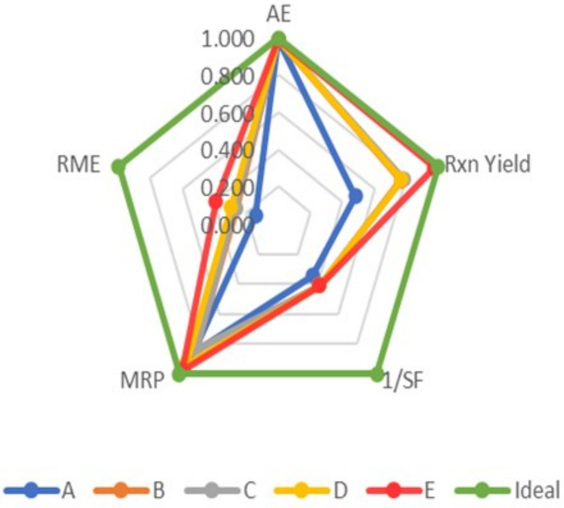


Figure 8. Measurement results of AE, RME, Rxn yield, 1/SF, and MRP values of HPC synthesis using Andraos spreadsheet.

reflux and heating (A, B, C and D), generally provide moderate yields (48.0–78.3%) under relatively harsh conditions (55–70 °C, 3–6 h). In contrast, ultrasonication (E, this study) achieved the highest yield (98%) under much milder conditions (25 °C, 30 min), highlighting the efficiency of cavitation in enhancing mass transfer and reaction kinetics.

The EATOS results in Figure 7 further highlight clear differences in environmental performance and allow direct comparison of input-related and output-related environmental impacts across the studied routes. Synthesis A, which employed the reflux method, exhibited the highest S^{-1} (1463.93 kg input/kg product) and EI_{in} (1494.61 PEI/kg), primarily due to extensive consumption of isopropanol solvent and substrate. Syntheses B, C, and D, carried out through conventional heating, produced substantially lower S^{-1} values (44.02, 54.24, and 22.98 kg input/kg product, respectively) and EI_{in} values (47.71, 55.95, and 44.08 PEI/kg, respectively). By contrast, synthesis E (this study) showed the lowest S^{-1} (21.36 kg input/kg product) and EI_{in} (39.47 PEI/kg), reflecting the reduced amount, cost, and hazard level of the chemicals used. These results highlight the superior environmental performance of ultrasonication for HPC synthesis compared to conventional methods. This trend is consistent with its high yield and improved reaction efficiency under ultrasonication conditions.

Synthesis A exhibited the highest E-Factor (1462.93 kg input/kg product), primarily due to the extensive use of substrate and isopropanol solvent. In contrast, syntheses B, C, D, and E generated substantially lower amounts of waste, with E-Factors of 43.02, 53.24, 21.98, and 20.26 kg input/kg product, respectively. Notably, synthesis E (this study) produced the lowest waste, as no significant excess of solvent or substrate was required, while simultaneously achieving the highest yield (98%). Synthesis A showed the highest EI_{out} value (5040.97 PEI/kg), confirming that the large consumption of isopropanol solvent results in the most significant environmental impact. By comparison, syntheses B, C, D, and E showed markedly lower EI_{out} values of 148.12, 186.86, 57.02, and 39.45 PEI/kg, respectively. The ultrasonication-assisted synthesis (synthesis E) demonstrated the most favourable environmental profile, combining high yield and DS values with minimal waste generation and the lowest environmental impact. the comparison clearly demonstrates a strong correlation between ultrasonication-assisted synthesis, higher product yield, and improved green metrics performance. These findings confirm ultrasonication as a superior green method compared to conventional heating and reflux-based syntheses.

The green metrics parameters of AE, RME, Rxn yield, 1/SF, and MRP for HPC synthesis were calculated using the Andraos spreadsheet and compared with four previous studies (Table 6, Figure 8). As expected, the AE values were identical across all syntheses (A, B, C, D and E) due to the assumption of the same etherification reaction pathway. This high AE value, approaching the ideal, indicates minimal waste generation and is consistent with the principles of green chemistry. Among the five syntheses, synthesis A showed the lowest performance in all parameters, which can be attributed to the high substrate-to-solvent (isopropanol) ratio used and its relatively low yield (48%). In contrast, synthesis E (this study) demonstrated the highest values for Rxn yield, 1/SF, MRP, and RME, reflecting its superior efficiency. Specifically, HPC synthesis via the ultrasonication method, employing a cellulose:NaOH ratio of 1:5 (w/v), 1.5 mL of etherification agent, and 8 mL of isopropanol solvent, yielded a high product output (97.5%) and a favorable environmental profile. Overall, the Andraos analysis confirms that ultrasonication is a greener and more efficient method compared to conventional heating and reflux-based syntheses, owing to shorter reaction time, higher yield, and significantly improved green metrics performance.

4. CONCLUSIONS

This study demonstrates a rapid and sustainable ultrasonication-assisted strategy for the synthesis of cellulose ethers (MC, CMC, and HPC) directly from banana blossom cellulose under ambient conditions. Beyond reproducing established etherification chemistry, the key advancement of this work is the combination of a low-energy, solvent-efficient ultrasonication platform with a rigorous, quantitative green chemistry benchmarking framework applied to multiple cellulose ether derivatives derived from an underutilized biomass resource. The ultrasonication process enables significantly accelerated etherification with high yields and favorable substitution levels while minimizing energy input and process complexity compared with conventional heating and other reported methodologies. Green metrics evaluation using both EATOS and the Andraos spreadsheet consistently confirms near-ideal environmental performance, with substantial improvements across all key sustainability indicators relative to conventional and literature-reported processes. Overall, this work establishes ultrasonication as a robust enabling technology for high-efficiency biomass valorization and positions it as a genuinely competitive green alternative for scalable cellulose ether production. It further provides a standardized quantitative framework for benchmarking the nvironmental performance of etherification processes, extending its relevance beyond the specific system studied.

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Conflicts of Interest

The authors declare no conflict of interest.

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