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Unveiling the environmental costs of lignocellulosic film production with ionic liquids: the case of 1-ethyl-3-methylimidazolium acetate†

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Sustainable plastic film alternatives require more than simply adopting "green" feedstocks. This study presents a comprehensive life cycle assessment (LCA) of lignocellulosic films produced using the ionic liquid 1-ethyl-3-methylimidazolium acetate ($[C_2C_1im][OAc]$), including a single recycling cycle. Employing the ReCiPe 2016 framework, the environmental performance was evaluated across key impact categories: global warming potential (GWP), human health (HH), ecosystem quality (EQ), and resource scarcity (RS). The results reveal unexpectedly high environmental burdens, primarily driven by energy-intensive stages such as ionic liquid recovery, particularly freeze crystallisation and solvent evaporation. Electricity consumption and the production of the ionic liquid itself were consistently the dominant contributors across all categories, overshadowing the comparatively negligible impacts of lignin and cellulose. When benchmarked against commercial cellophane, the lignocellulosic films demonstrated substantially higher environmental impacts in every category assessed. These findings challenge the assumption that bio-based inherently equates to sustainable and highlight the urgent need for process optimisation, energy efficiency improvements, and integration of low-carbon power sources to unlock the true environmental potential of lignocellulosic film technologies.

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Sustainability spotlight

Ionic liquids are often regarded as a green solvent for processing biomass feedstock to create sustainable lignocellulosic films that could address the plastic waste crisis. However, very few studies have examined the overall environmental impact of ionic liquids for lignocellulose processing. We present a comprehensive LCA of lignocellulosic films produced using 1-ethyl-3-methylimidazolium acetate ($[C_2C_1im][OAc]$). The results reveal unexpectedly high environmental burdens, primarily driven by energy-intensive stages such as ionic liquid recovery, particularly freeze crystallisation and solvent evaporation. Our work challenges the notion that bio-based materials inherently equate to sustainable options and underscores the urgent need for process optimisation, energy efficiency improvements. It also aligns with several UN Sustainability Development goals: (i) Responsible Consumption and Production, and (ii) Climate action.

Introduction

Recent studies in sustainable materials research underscore cellulosic films as viable and eco-friendly alternatives to conventional plastic films derived from fossil resources.^{1–3} A particularly promising avenue within this domain is the integration of lignin into cellulosic matrices, a strategy that significantly enhances these bioplastics' mechanical robustness and environmental resilience. Lignin, with its intrinsic capacity for nanoscale entanglement and hydrogen bonding, effectively

reinforces cellulose micro- and nanofibrils, yielding materials with superior thermal stability and mechanical strength.^{4,5} Furthermore, the incorporation of lignin into cellulose films has demonstrated marked improvements in critical functional properties, including UV-shielding, biodegradability, antioxidant and antibacterial activity, and water resistance.^{6–8} Economically, lignin presents a significant advantage as an abundant, low-cost byproduct of the pulp and paper industry, which reduces the need for extensive biomass fractionation and cellulose purification, thereby lowering production costs.^{5,9} Additionally, lignin's biodegradable nature and compatibility with recycling processes further elevate the environmental credentials of lignocellulosic bioplastics, making them a highly attractive option for sustainable production in alignment with circular economy goals.^{4,10}

Ionic liquids (ILs), due to their unique ability to dissolve cellulose and lignocellulose under relatively mild conditions,

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provide an efficient route to produce these high-performance films while avoiding the use of hazardous chemicals required in traditional plastic manufacturing.^{11,12} Imidazolium-based ILs, such as 1-butyl-3-methylimidazolium chloride ($[C_4C_1im]Cl$) and 1-ethyl-3-methylimidazolium acetate ($[C_2C_1im][OAc]$) or EmimAc, have been extensively investigated for their ability to dissolve cellulose and lignocellulose effectively.^{13–17} Films produced using $[C_2C_1im][OAc]$ not only demonstrate excellent barrier properties but are also fully biodegradable, positioning them as a viable alternative to conventional plastics.¹⁸

Despite the proven advantages of lignocellulosic films over conventional flexible plastics, the complete range of environmental impacts linked to their production—such as effects on human health, ecosystem quality, and resource scarcity—remains insufficiently explored. This is particularly true for the production and recyclability of ILs, which are integral to the manufacturing process. The synthesis of ILs is often energy-intensive, and concerns about their potential ecotoxicity and environmental persistence are heightened when solvent recycling is not fully optimised.^{19,20} Consequently, while IL-based lignocellulosic films present a promising sustainable alternative, a comprehensive assessment of their environmental impact is essential to gauge their long-term sustainability accurately.

To address these uncertainties and identify key environmental hotspots in the production of lignocellulosic films using ILs, we present a comprehensive life cycle assessment (LCA) of films produced with $[C_2C_1im][OAc]$, considering a scenario with a single recycling cycle of this ionic liquid. Using the hierarchist perspective²¹ of the ReCiPe 2016 framework,²² the analysis focuses on Global Warming Potential (GWP) and three key endpoint indicators: Human Health (HH), evaluating potential disease burdens; Ecosystem Quality (EQ), assessing impacts on biodiversity; and Resource Scarcity (RS), measuring natural resource depletion and associated economic costs.

Methodology

The following section outlines the methodology used to evaluate the environmental performance of the lignocellulosic films, beginning with a description of the synthesis of the ionic liquid and the subsequent film fabrication process. This forms the basis for the life cycle assessment, which integrates both foreground and background system data to capture the complete environmental profile.

Synthesis of ionic liquid and fabrication of lignocellulosic films

In order to select the most suitable candidate for LCA, a series of lignocellulosic films with varying cellulose-to-lignin ratios were prepared and characterised under controlled laboratory conditions.²³ Some relevant experimental results, summarised in Table 1, include polymer composition, film thickness, maximum tensile strength (σ_{max}), and Young's modulus (E). The mechanical performance of these films— σ_{max} ranging from 65.44 MPa to 93.15 MPa and E values between 5.83 GPa and 8.54

Table 1 Tensile properties of the lignocellulosic films²³

Sample	Ratio of polymers (%)		Thickness (μm)	σ_{max} (MPa)	E (GPa)
	Cellulose	Lignin			
F-0	100	0	19.4 ± 3.9	80.0 ± 9.9	7.1 ± 1.2
F-1	96	4	24.4 ± 3.3	65.4 ± 8.4	6.3 ± 1.0
F-2	92	8	24.4 ± 4.7	93.2 ± 6.6	8.5 ± 0.6
F-3	80	20	25.6 ± 3.4	76.8 ± 5.5	6.6 ± 0.5
F-4	71	29	28.5 ± 3.1	72.3 ± 4.3	5.8 ± 0.3

GPa—falls well within or above the typical range reported for commercial cellophane ($\sigma_{max} = 40$ –125 MPa; $E = 0.9$ –3 GPa^{24,25}). These results align with previous research, which suggests that lignin contents between 5% and 10% can effectively improve the mechanical properties of cellulose-based films without compromising their functionality.^{26,27} Sample F-2, composed of 92% cellulose and 8% lignin, demonstrated the highest tensile strength (93.15 MPa) and was thus selected as the reference material for further sustainability analysis through LCA.

Fig. 1 illustrates the production process studied. Due to the lack of process data regarding the industrial production of

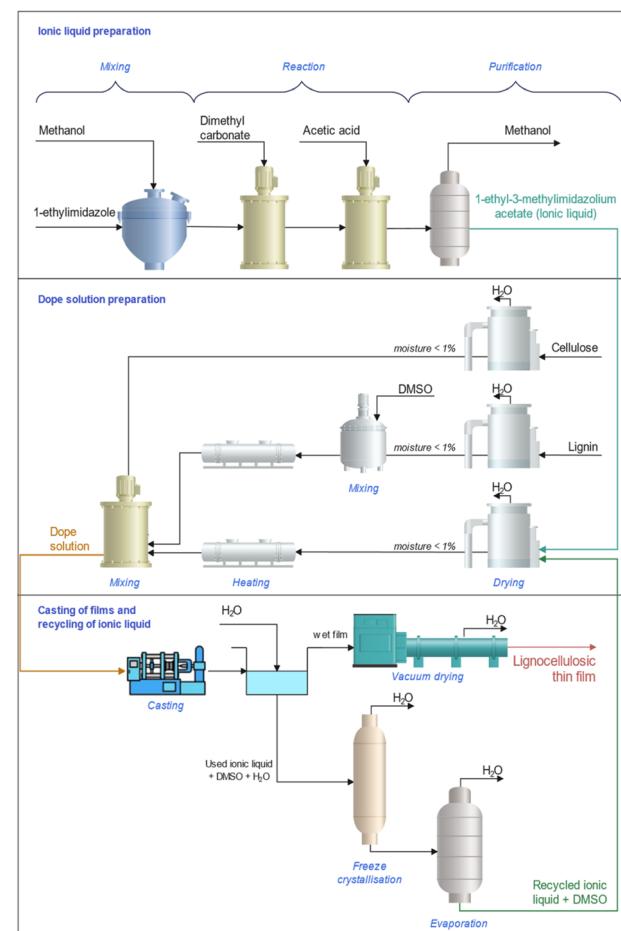


Fig. 1 Lignocellulosic film production: process flowsheet.



[C₂C₁im][OAc] and lignocellulosic films, our own lab-based results and process simulations were used to approximate mass and energy consumption at scale, guided by established process design heuristics. The synthesis starts with the dissolution of 1-ethylimidazole in methanol, reacting it with dimethyl carbonate under pressure at 140 °C, adding acetic acid, and then purifying the product through filtration and methanol evaporation. This IL was subsequently employed in the dope solution preparation, where lignin was first dissolved in DMSO and then combined with cellulose and the IL at 70 °C. The resulting dope solution was cast into films and immersed in water baths to regenerate the cellulose and remove solvents.

To recover and recycle the ionic liquid, the films were first subjected to vacuum drying to remove residual moisture and facilitate subsequent recovery steps. The remaining aqueous mixture, containing water, DMSO, and the ionic liquid, was then processed through a combination of freeze crystallisation and evaporation. This hybrid separation route was chosen based on its enhanced suitability for managing highly diluted systems characteristic of cellulose regeneration processes, where large volumes of water must be separated from relatively small quantities of solute.

Conventional separation methods, such as vacuum distillation, while effective in less dilute systems, become both energetically and economically impractical at this scale due to the high latent heat of vaporisation of water and the low volatility of the IL and DMSO. Moreover, elevated temperatures required in distillation may lead to thermal degradation of the IL or undesirable chemical changes in DMSO, compromising the integrity and recyclability of both solvents. Freeze crystallisation offers an alternative by operating at low temperatures to selectively crystallise out water, thereby reducing the thermal stress on the remaining components and preserving solvent quality.^{28,29}

Life-cycle assessment (LCA)

To evaluate the environmental impacts of producing [C₂C₁im][OAc] in our process, the methodology proposed by Cuéllar-Franca *et al.*³⁰ was applied. Then, Aspen Plus v14 (ref. 31) was used to model and replicate the production processes of the dope solution, film casting, and IL recycling (foreground system). After estimating the mass and energy needs, the guidelines provided by Bruijn *et al.*³² were followed to apply LCA. The LCA models were executed using SimaPro v9,³³ connected to Ecoinvent 3.8 (ref. 34) for data input, based on the approach and assumptions explained below.

Goal and scope definition. We adopted a cradle-to-gate methodology, encompassing all emissions throughout the whole life cycle, starting from the production of the ionic liquid and ending with the production of lignocellulose film. The functional units for the analyses are defined as follows: 1 kg of [C₂C₁im][OAc] for the preparation of the IL, 1 kg of dope solution for the preparation of the dope solution, and 1 kg of lignocellulose film for the casting of films and the recycling of the IL.

Life-cycle inventory (LCI). The LCI integrates data from both the foreground system, which includes mass and energy inputs

from the production processes, and the background system, which encompasses the surrounding processes that supply inputs to those main processes. The information on the foreground system was obtained from the process model in Aspen Plus. Conversely, the background system data were retrieved from Ecoinvent v3.8. The application of Allocation at the Point of Substitution (APoS) was justified due to its ability to encompass the effects of both production and treatment processes.³⁵

Water recovered from the separation steps was assumed to undergo treatment in wastewater facilities. Energy consumption for recycling processes was derived from Liu *et al.*,²⁸ while electricity use for film casting was obtained from existing literature.³⁶ Comprehensive process inventories are available in the ESI S1.†

Environmental impact assessment (EIA). Following the hierarchist cultural perspective,²¹ we employed the ReCiPe 2016 framework²² as implemented in SimaPro. Our study primarily focused on evaluating global warming potential (GWP) along with three endpoint indicators: human health (HH), ecosystem quality (EQ), and resource scarcity (RS). These endpoint indicators were derived through the aggregation of various midpoint indicators, as listed below:

HH (DALY): global warming (HH), stratospheric ozone depletion, ionising radiation, ozone formation (HH), fine particulate matter formation, human carcinogenic toxicity, human non-carcinogenic toxicity, and water consumption (HH).

EQ (species.yr): global warming (terrestrial ecosystems), global warming (freshwater ecosystems), ozone formation (terrestrial ecosystems), terrestrial acidification, freshwater eutrophication, marine eutrophication, terrestrial ecotoxicity, freshwater ecotoxicity, marine ecotoxicity, land use, water consumption (terrestrial ecosystems), and water consumption (aquatic ecosystems).

RS (USD₂₀₁₃): mineral resource scarcity and fossil resource scarcity.

Results and discussion

Analysis at the endpoint level + GWP: ionic liquid production

The LCA results presenting the synthesis of [C₂C₁im][OAc] in our production process are shown in Fig. 2. Methanol-related emissions are deemed negligible, assuming all methanol is recirculated through an energy-efficient recycling process.³⁷ The total GWP for the production process is 4.6 kg CO₂ eq. per kg [C₂C₁im][OAc]. The most significant contributor is 1-ethylimidazole, accounting for approximately 60% of the total GWP. Dimethyl carbonate also contributes significantly (26%), followed by heating from natural gas (NG) (7%), acetic acid (3%), electricity (2%), and direct emissions (3%). The total impact on HH is 0.9×10^{-5} DALY per kg [C₂C₁im][OAc]. Again, 1-ethylimidazole is the dominant contributor (62%), followed by dimethyl carbonate (27%). Acetic acid, direct emissions, heating from NG, and electricity contribute less significantly, with respective impacts of 4%, 1%, 4%, and 2%. The total EQ impact is 2.3×10^{-8} species year per kg [C₂C₁im][OAc]. 1-



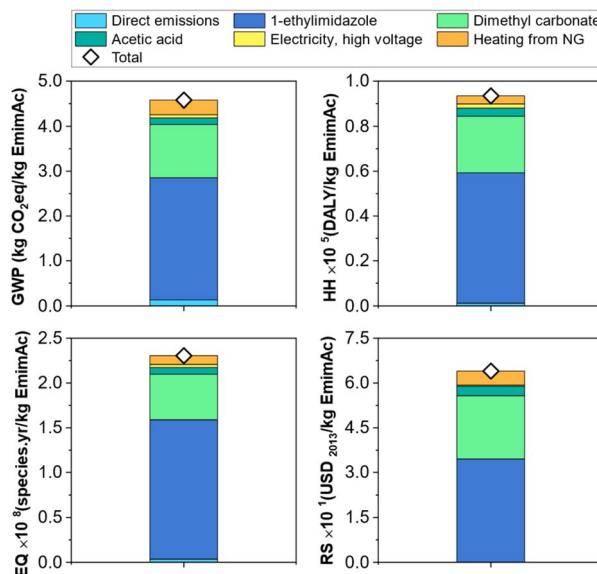


Fig. 2 Ionic liquid (IL) preparation: LCA at the endpoint level + GWP per kg of IL. GWP: global warming potential, HH: human health, EQ: ecosystem quality, RS: resource scarcity.

Ethylimidazole leads with an impact equivalent to 67% of the total EQ impact, followed by dimethyl carbonate (22%), heating from NG (4%), acetic acid (3%), direct emissions (2%), and electricity (2%). The total impact on RS is 0.64 USD₂₀₁₃ per kg [C₂C₁im][OAc]. The major contributor is 1-ethylimidazole, with 54% of the total RS impact, followed by dimethyl carbonate (33%). Acetic acid, heating from NG, and electricity have lesser contributions, with impacts equivalent to 5%, 7%, and 1%, respectively. In this analysis, direct emissions have no measurable impact on resources. Therefore, the production of [C₂C₁im][OAc] shows significant environmental impacts primarily driven by the use of 1-ethylimidazole and dimethyl carbonate across all categories.

Analysis at the endpoint level + GWP: dope solution

Fig. 3 shows the GWP and endpoint-level results of the environmental assessment of the production of the dope solution. The total GWP for producing 1 kg dope solution is 4.1 kg CO₂ eq. The largest contributor is [C₂C₁im][OAc], accounting for approximately 79% of the total GWP. Electricity usage significantly impacts GWP, contributing 12%. Dimethyl sulfoxide contributes 7%, while cellulose and lignin have minimal contributions, at <2%. Wastewater treatment's impact is negligible. The total impact in the HH impact category is 8.7 × 10⁻⁶ DALY per kg dope solution. Again, 1-ethyl-3-methylimidazolium acetate is the dominant factor, contributing 76%. Dimethyl sulfoxide and electricity follow, contributing 9% each. Cellulose and lignin have minor impacts, contributing 5% and <1%. For EQ impacts, the total impact is 2.1 × 10⁻⁸ species year per kg dope solution. The contributions from different components follow a similar trend, with 1-ethyl-3-methylimidazolium acetate being the highest at 77%. Electricity contributes 10%, and dimethyl sulfoxide 8%. Cellulose and lignin contribute 5%

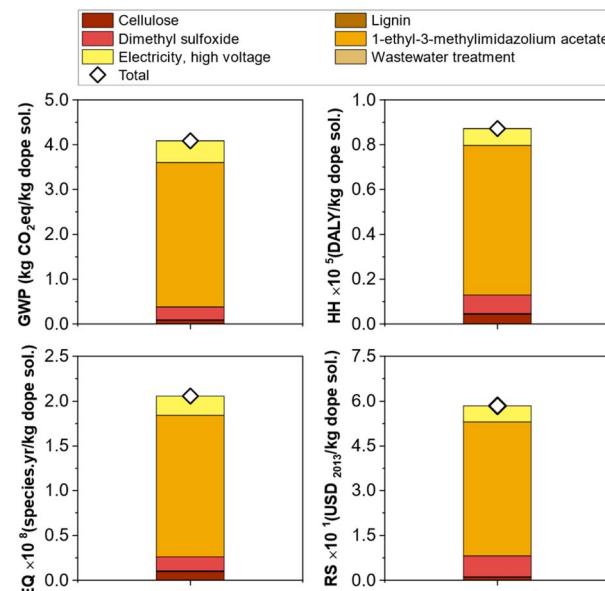


Fig. 3 Dope solution: LCA at the endpoint level + GWP per kg of dope solution. GWP: global warming potential, HH: human health, EQ: ecosystem quality, RS: resource scarcity.

and <1%. The total RS impact is 0.58 USD₂₀₁₃ per kg dope solution. Here, 1-ethyl-3-methylimidazolium acetate is the primary contributor at 77%. Dimethyl sulfoxide contributes 12%, and electricity adds 9%. Cellulose and lignin contribute 2% and <1%, respectively. This analysis shows that in all impact categories, [C₂C₁im][OAc] consistently emerges as the largest contributor, ranging from 76% to 77%. Electricity and dimethyl sulfoxide also have significant impacts, while cellulose, lignin, and wastewater treatment contribute minimally.

Analysis at the endpoint level + GWP: lignocellulosic thin film production

Fig. 4 presents the GWP and endpoint-level results from the environmental assessment of lignocellulosic thin film production, accounting for a single ionic liquid recycling cycle. The GWP is measured at 183 kg CO₂ eq. per kg film. Significant contributors include electricity for freeze crystallisation, which accounts for 102 kg CO₂ eq. per kg film (56% of the total GWP), followed by the dope solution, contributing 47 kg CO₂ eq. per kg film (26%). Electricity for evaporation adds 43 kg CO₂ eq. per kg film (24%), and electricity for vacuum drying contributes 24 kg CO₂ eq. per kg film (14%). The total impact on HH is 2.9 × 10⁻⁴ DALY per kg film. The primary contributors include electricity for freeze crystallisation, impacting 1.6 × 10⁻⁴ DALY per kg film (55% of the total human health impact), and the dope solution, contributing 1 × 10⁻⁴ DALY per kg film (34%). Electricity for evaporation and vacuum drying also significantly affect human health, with contributions of 6.8 × 10⁻⁵ DALY per kg film (23%) and 3.9 × 10⁻⁵ DALY per kg film (13%), respectively. The impact on EQ, measured at 8.1 × 10⁻⁷ species year per kg film, is predominantly influenced by electricity for freeze crystallisation, contributing 4.5 × 10⁻⁷ species year per kg film (56%),



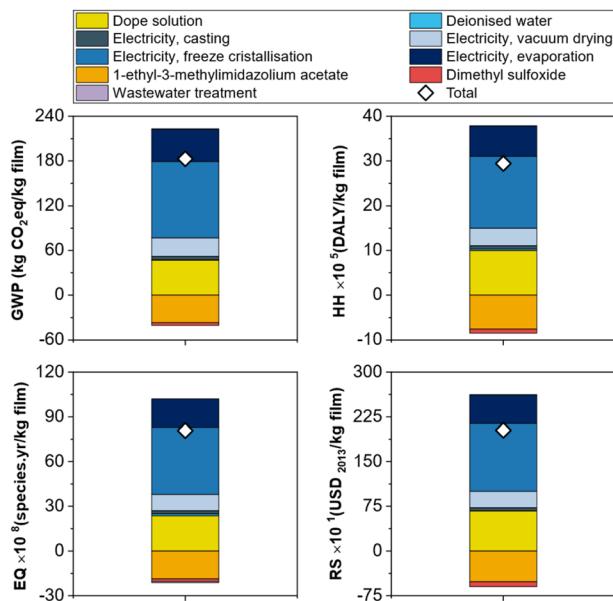


Fig. 4 Lignocellulosic thin film production: LCA at the endpoint level + GWP per kg of film. GWP: global warming potential, HH: human health, EQ: ecosystem quality, RS: resource scarcity. A single recycling cycle of the ionic liquid ($[C_2C_1im][OAc]$) is accounted for. Negative values in all impact categories reflect this. The emissions related to electricity for vacuum drying, freeze crystallisation, and evaporation are tied to the recycling cycle.

and the dope solution, adding 2.4×10^{-7} species year per kg film (29%). Electricity for evaporation contributes 1.9×10^{-7} species year per kg film (24%), while electricity for vacuum drying adds 1.1×10^{-7} species year per kg film (14%). Recycling $[C_2C_1im][OAc]$ results in a negative impact of -1.9×10^{-7} species year per kg film, indicating a reduction in overall ecosystem damage. The RS impact is 20.30 USD_{2013} per kg film. Electricity for freeze crystallisation is the most significant contributor, accounting for 11.40 USD_{2013} per kg film (56%), followed by the dope solution, contributing 6.71 USD_{2013} per kg film (33%). Electricity for evaporation adds 4.79 USD_{2013} per kg film (24%), while electricity for vacuum drying contributes 2.76 USD_{2013} per kg film (13.6%). Notably, the recycling of $[C_2C_1im][OAc]$ and DMSO results in negative contributions of -5.15 USD_{2013} per kg film and -0.81 USD_{2013} per kg film, respectively, demonstrating resource savings through recycling.

Comparison at the endpoint level + GWP: lignocellulosic thin film vs. cellophane production

A comparison of the LCA results for lignocellulose film and cellophane (see ESI S2†) reveals that the GWP of the lignocellulosic film is approximately 31 times higher than that of cellophane. For HH impacts, the lignocellulosic film is about 37 times greater. In terms of EQ, the lignocellulosic film impacts approximately 27 times more than cellophane. The difference is particularly pronounced in RS, with the lignocellulosic film being approximately five orders of magnitude more resource-intensive than cellophane. Notably, the cited source for cellophane indicates no impact on fossil resource scarcity, with only

minimal impact observed in the mineral resource scarcity category.³⁸ This comparison shows that cellophane has a markedly lower environmental impact across all four categories when compared to the lignocellulosic films studied, indicating that, for now, cellophane remains the more sustainable option.

It is important to note that the comparison between lignocellulosic films and commercial cellophane is intended as an indicative benchmark rather than a definitive equivalence. Cellophane represents a mature, industrial-scale technology that has undergone decades of optimisation, whereas the lignocellulosic films assessed here are based on early-stage laboratory processes that have yet to benefit from scale-up efficiencies or advanced process integration. The purpose of this comparison is not to draw conclusive judgments about the superiority of one material over the other but rather to provide context for understanding the current environmental performance of lignocellulosic films and to identify the key challenges that must be addressed for these materials to become competitive from a sustainability perspective.

Conclusions

This study presents a cradle-to-gate life cycle assessment (LCA) of lignocellulosic film production using 1-ethyl-3-methylimidazolium acetate ($[C_2C_1im][OAc]$) as the cellulose-dissolving ionic liquid, with environmental impacts assessed across global warming potential (GWP), human health (HH), ecosystem quality (EQ), and resource scarcity (RS). Based on experimental data and process simulation, the assessment identifies key environmental hotspots and provides insight into the process-level challenges of implementing bio-based films at scale.

The most significant environmental burdens stem from the ionic liquid recovery stage, particularly due to the energy demands of freeze crystallisation, evaporation, and vacuum drying. This separation strategy was selected for its enhanced technical suitability in managing highly diluted aqueous systems, which are typical of cellulose regeneration, where conventional methods like vacuum distillation are energetically inefficient and risk degrading the solvents. Freeze crystallisation allows for selective water removal under low-temperature conditions, helping to preserve the chemical integrity of $[C_2C_1im][OAc]$ and DMSO. However, this advantage comes at the cost of high electricity consumption, which dominates the environmental impacts across all categories. Therefore, while the recycling of ionic liquids is typically promoted as a strategy to enhance resource efficiency, our findings suggest that, under the specific conditions evaluated, its environmental costs may outweigh the benefits—particularly when dependent on carbon-intensive energy sources. By contrast, the environmental contributions from cellulose and lignin, as renewable biopolymer inputs, are comparatively minor. This underscores that the sustainability of bio-based materials depends not only on feedstock selection but equally on the efficiency of processing infrastructure and the carbon footprint of supporting energy systems.



To contextualise these findings, we benchmarked the assessed system against commercial cellophane, a fully optimised industrial product. Nonetheless, benchmarking the two systems reveals that lignocellulosic films currently exhibit significantly higher environmental impacts—by one to two orders of magnitude in most categories—pointing to the urgent need for innovation. The disparity is not a reflection of the intrinsic unsustainability of bio-based films, but rather of the challenges associated with early-stage processing technologies that have yet to benefit from optimisation, scale-up, and cleaner energy inputs.

Although the results of this study are specific to the selected ionic liquid, film formulation, and process configuration, the overall conclusions offer broader relevance. The production of cellulose-based films using ionic liquids generally follows similar process steps—polymer dissolution, film casting, coagulation, and solvent recovery—and is subject to comparable operational constraints. As such, the insights gained here serve as a valid reference point to inform future efforts aimed at improving both production methodologies and the strategic selection of raw materials and utilities. By identifying key environmental hotspots, particularly those related to solvent recovery and energy use, this study provides a foundational benchmark to guide future research in ionic liquid design, energy-efficient separation technologies, and the integration of low-carbon energy systems into the production of bio-based materials.

Ongoing optimisation of IL recovery processes, exploration of lower-impact solvent systems, and coupling with renewable energy will be essential for improving the sustainability of lignocellulosic films. As the field advances toward commercial maturity, embedding life cycle thinking into process development will be critical to realising the environmental potential of bio-based material technologies.

Data availability

Supporting data are provided in the ESI.†

Conflicts of interest

There are no conflicts to declare.

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