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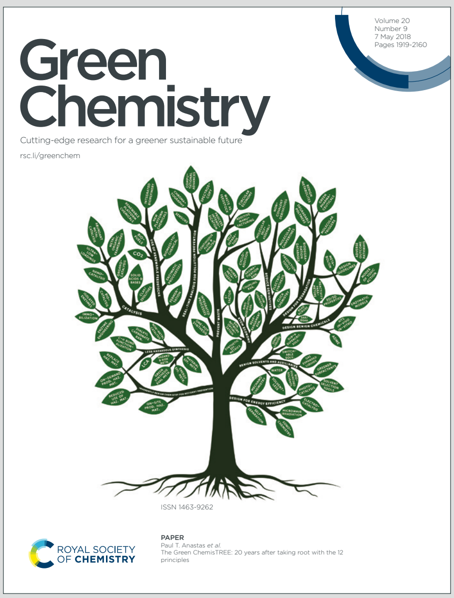

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Green foundation box

1. Upcycling of agricultural waste contributes to a circular bioeconomy. This work evaluates the environmental impact of zein, ethyl lactate, and their membrane derivatives. The membranes are solely derived from corn biomass (an agricultural waste).
2. The hotspots of zein extraction are identified. We show that solvent selection and management are essential for reducing the environmental impact of zein extraction. We also show the impact of systems thinking by applying different extraction methods with different environmental impacts for membrane production.
3. The optimal conditions of zein extraction, experimental comparisons under the same conditions, and life cycle assessments of bio-based and conventional membranes must be further researched.

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Life cycle assessment of zein, ethyl lactate, and derivative nanofiltration membranes

Claudia Oviedo,[1,2] Akos Cseke,[3] Gyorgy Szekely[1,2,4*]

[1] Advanced Membranes and Porous Materials Center; Physical Science and Engineering Division (PSE), King Abdullah University of Science and Technology, (KAUST), Thuwal, 23955-6900, Saudi Arabia

[2] Environmental Science and Engineering Program; Biological, Environmental Sciences and Engineering Division (BESE); King Abdullah University of Science and Technology (KAUST), Thuwal, 23955-6900, Saudi Arabia

[3] Department of Mechanical, Materials and Manufacturing Engineering; University of Nottingham, Nottingham, NG7 2RD, United Kingdom

[4] Chemical Engineering Program; Physical Science and Engineering Division (PSE), King Abdullah University of Science and Technology (KAUST), Thuwal, 23955-6900, Saudi Arabia

*email: gyorgy.szekely@kaust.edu.sa, tel: +966128082769, web: www.SzekelyGroup.com

Abstract

Agricultural biomass from corn can be upcycled into valuable biodegradable materials such as zein, ethyl lactate, and membranes, reducing depleting resource consumption, minimizing waste, and contributing to a circular bioeconomy. Zein, a protein sourced from corn, is an alternative to fossil-based polymers. Proteins are more abundant, biodegradable, nontoxic, complex, and versatile than other biopolymers. In ethyl lactate, an alternative green solvent emerging in various applications, zein can form nanofiltration membranes. Herein, we report the life cycle assessment of zein, ethyl lactate, and membranes derived from these compounds. The assessment compares different feedstock, solvents, purification techniques, and solvent management methods to identify the environmental hotspots, particularly during the extraction stage, as an aid for decision-making. Notably, although environmental impacts are not necessarily outweighed by utilizing agricultural waste, they can be reduced through high-protein-content feedstock and proper solvent management. We also show the impact of systems thinking through the production of membrane materials and close the life cycle loop via biodegradation.

Keywords: circular economy, waste utilization, upcycling, protein, green solvent, membrane

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1. Introduction

Production of fossil-based materials generates pollutants that threaten the environment and public health [1]. To overcome these challenges, we must systemically understand the entire supply chain from production to disposal. Biomass-derived bioplastics can meet the functional demands of fossil-based polymers while reducing the environmental burden. Meanwhile, concerns related to agricultural residues, food loss, and waste are increasing. Agriculture consumes vast quantities of natural resources; for instance, irrigation contributes approximately 70% to the global water consumption [2]. Additionally, agriculture predominantly contributes to climate change, emitting between one-quarter and -third of the global anthropogenic greenhouse gases per year [3]. For every kilogram of wasted protein, up to 750 kg of CO_2 is released into the atmosphere [4], equivalent to the emissions of a sports car driven for 3,200 km [5]. Therefore, understanding and optimizing the life cycles of these resource-intensive systems are essential (Figure 1). One promising strategy is the upcycling of biomass into high-value building blocks and complex materials, providing sustainable alternatives to primary production [6–10]. Such strategies promise to reduce waste, lower emissions, and support resilient economies [11–13].

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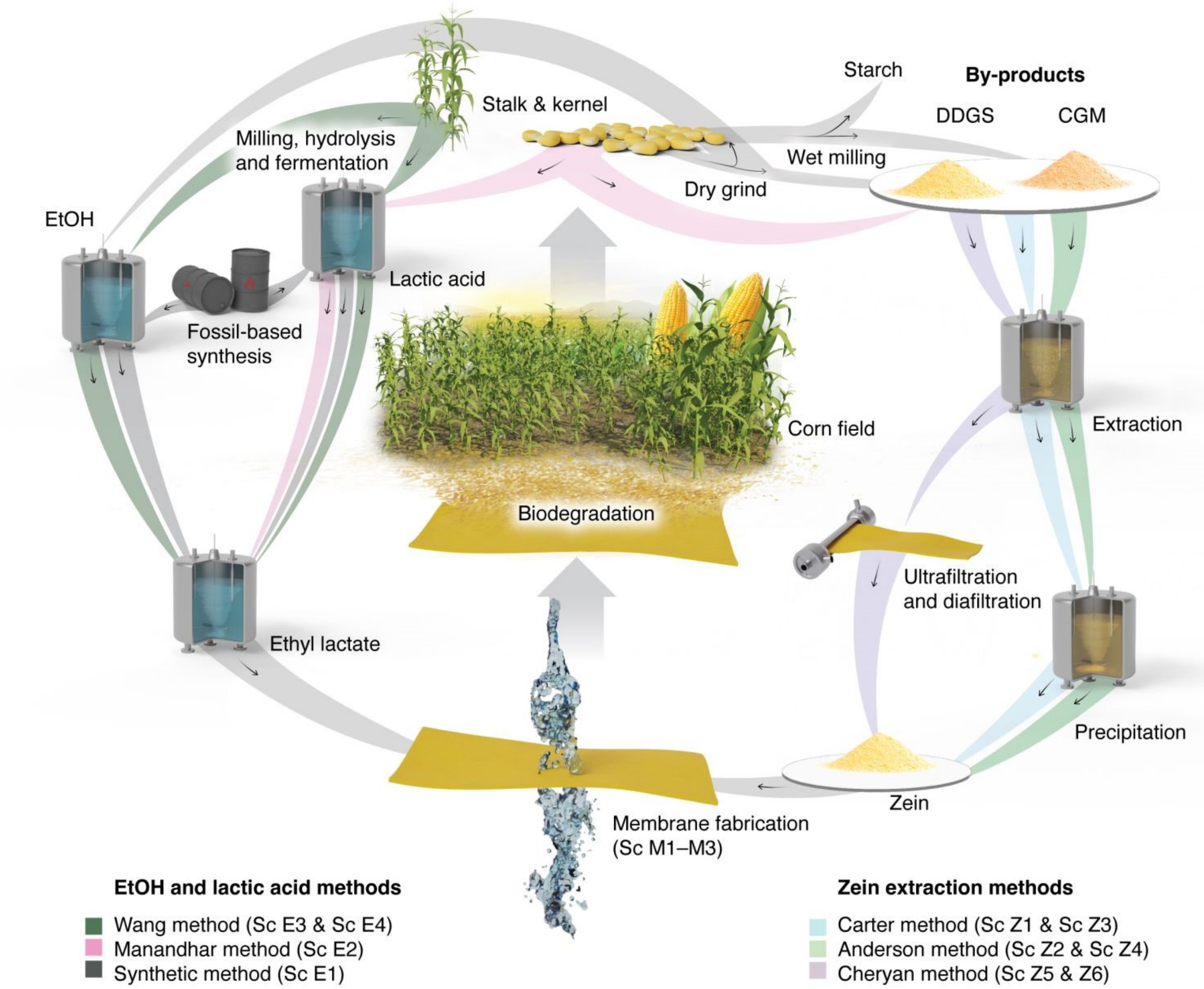

Figure 1. Closed-loop production routes of zein, ethyl lactate, and their derived membranes from the stalks and kernels of corn. Biodegradation is a viable, environmentally friendly option for disposing zein-based materials.

Corn is the most cultivated cereal crop worldwide and is deeply rooted in global food cultures. Besides being an important food source, corn is utilized in both agricultural products (such as animal feed) and industrial products (such as cornstarch, adhesives, sweeteners, and alcohol) [14–16]. The United States and China are the leading producers of corn, accounting for approximately half (~620 million metric tons) of the global annual production [17]. By maximizing the value of corn byproducts, we could enhance the overall efficiency of the corn industry, support environmental sustainability, and generate economic opportunities for rural communities [14].

At present, strategies for converting food waste including proteins into sustainable materials and technologies are rapidly emerging [4,18–20]. Zein, a protein obtained from byproducts of cornstarch and corn-

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syrup manufacturing [21], has been widely exploited for its distinctive hydrophobicity, high elasticity, and film-forming ability [22–26]. Among the biopolymers, zein offers outstanding scalability potential and is supported by the well-established corn biorefinery infrastructure [21]. Meanwhile, ethyl lactate is an emerging green solvent that promises to enhance the health, safety, and environmental friendliness of dissolution, reaction, and transportation processes. As a nontoxic, biodegradable, bio-based solvent with excellent solvency and a high boiling point, ethyl lactate has been variously applied in biomass fractionation [27], coatings [28], extraction [29], and membrane production [22,30,31].Membrane technology is widely recognized for its energy efficiency, nonthermal operation, and scalability. Membranes consume less energy, and therefore emit less CO_2, than conventional separation methods [32]. However, the sustainability of membrane-material manufacturing is compromised by reliance on fossil-based polymers and toxic solvents, along with waste generation and end-of-life incineration. Green solvents such as ethyl lactate can improve the sustainability of membrane manufacturing, which frequently utilizes *N*,*N*-dimethylformamide and *N*-methyl-2-pyrrolidone as the solvents and polyacrylonitrile, polyimide, and polybenzimidazole as the polymers. Approximately 80% of these conventional polymers are eventually deposited in landfills [33].

Several strategies have proposed greener solvents and chemicals, simplified production steps, prioritization of renewable raw materials, and biodegradable or recyclable membrane designs with low environmental persistence [34]. Biodegradable membranes naturally degrade at the end of their life cycle, minimizing landfill accumulation [33]. Although biopolymers have been well-explored for membrane production [35–38], proteins are promising sources of as-yet scarcely undeveloped materials [22]. The complexity and tenability of proteins renders them versatile and functionally adaptable [19], offering unique advantages for membrane design.

Although membrane fabrication has raised environmental concerns, life cycle assessments (LCAs) of membrane manufacturing are under-reported. LCA articles on bio-based materials [39–41] and on membranes produced through interfacial polymerization [42], electrospinning [43], and phase inversion [44–46] have been reported. The chosen solvents and polymers have been identified as the major environmental impact indicators of membrane fabrication [45].

A recent study has demonstrated the development and utilization of zein-based membranes in pharmaceutical purification [22], supporting the relevance of understanding and improving the sustainability of the zein production process. Despite growing interest in zein as a potential large-scale biopolymer, emissions related to zein production are lacking even in major databases such as Ecoinvent and Agri-footprint. To fill this knowledge gap, we quantified the emissions and evaluated the environmental impact of zein. By comparing various zein extraction methods, we identify key hotspots in production, support the scaling of zein production for commercial applications, and provide improved decision-making guidance for zein

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manufacturers. Our insights are potentially relevant to other solvent-based extraction systems, which often involve similar trade-offs between process efficiency, material quality and environmental performance. We also report the LCAs of ethyl lactate as an emerging green solvent and of membranes manufactured from zein and ethyl lactate.

2. Methodology

2.1 Goal and scope

The environmental impacts of manufacturing zein, ethyl lactate, and their derived membranes were evaluated through a cradle-to-gate LCA (Figure 1). In this analysis, the functional units of zein, ethyl lactate, and membranes were set at 1 kg, 1 kg, and 1 m^2, respectively. This selection assumes that the rejection performance and lifetime of the membranes are equivalent [22]. Accordingly, the analysis ignores the uncertainty in separation performance and focuses solely on the selected materials and processing methods. The influences of different parameters on the process and environmental impact of zein, ethyl lactate, and membrane production are evaluated in various scenarios. The zein extraction and ethyl-lactate production scenarios, both required for membrane manufacturing, are presented in Figure 1. The study was performed using SimaPro 9.6.0.1 [47] with the Ecoinvent 3.10 [48] and Agri-footprint 6.3 [49] databases.

2.2 Impact assessment

The impact assessment was performed midpoint using the ReCiPe 2016 Midpoint (H) V1.08/World (2010) H methodology [50], and the global warming potential (GWP) was selected as the main impact category. Other midpoint categories were included to assess the broader environmental impacts (water and land use) based on the recommendations in [51].

2.3 Inventory analysis

A zein-based membrane is produced by dissolving zein in ethyl lactate followed by casting and phase inversion. Both raw materials (zein and ethyl lactate) are extractable from corn. The data of zein membrane production [22], zein extraction [52–54], lactic acid [55,56], and ethanol (EtOH) [56] required for ethyl lactate synthesis [57] were obtained from the literature. The United States, the largest corn producer with approximately 377 million metric tons per year [17], was selected as the reference location of the energy-related inputs. Corn is primarily processed by dry milling, alkaline processing, wet milling, and dry-grind processing [21]. The first two methods produce corn for human consumption, wet milling yields starch and oil, along with corn gluten meal (CGM) and corn gluten feed as byproducts, and dry grinding obtains EtOH, along with distillers dried grains (DDG) or DDG with solubles (DDGS) as byproducts. CGM and DDGS byproducts are

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rich in protein and suitable sources of zein extraction. DDGS has received special attention as a potential zein source [58] because dry-grind EtOH plants produce approximately 40–50% of the EtOH in the United States.

We compared the environmental impacts of zein extraction scenarios with different feedstocks and solvents (Sc Z1–Z4), purification methods (Sc Z5 and Z6), and integrated solvent management methods (Sc Z7 and Z8). We also investigate the environmental impacts of ethyl lactate pathways with different feedstocks—synthetic (Sc E1), corn kernel (Sc E2), corn stalk (Sc E3 & E4), and different zein and membrane production pathways. The used solvent was modeled as a hazardous waste to be incinerated.

2.2.1 Zein extraction

We focus on the Carter, Anderson, and Cheryan methods to identify the hotspots of zein extraction (Figure 2). Zein extraction was evaluated and compared in different scenarios with different feedstocks, solvents, purification, and solvent management methods (Table 1).

In the commercial zein extraction method of Carter et al., CGM with 88 vol% 2-propanol is heated at 60 °C with agitation followed by precipitation at −18 °C and drying in a vacuum oven [59]. The product purity can be increased by redissolving and reprecipitating the zein. Anderson et al. optimized this method to increase the yield by adding a selective precipitation step and evaluating other solvents (e.g., 70 vol% EtOH) [52]. They also evaluated zein extraction from DDGS [53]. Cheryan et al. modified the Carter zein extraction method by integrating ultrafiltration membranes for concentration and purification via diafiltration [54].

The yield and extraction efficiency zein were calculated as

$$Zein\ yield\ [\%] = \frac{mass_{zein\ extracted}}{mass_{feedstock}} \times 100, \quad (1)$$

$$Zein\ extraction\ efficiency\ [\%] = \frac{(mass_{zein\ extracted})(protein\ purity, \%)}{mass_{zein\ available\ in\ feedstock}} \times 100, \quad (2)$$

where the available zein refers to the zein content in the feedstock (DDGS or CGM), and the zein extraction efficiency is the zein yield corrected for the zein content in the feedstock based on a well-stablished definition [52,53].

Zein is extracted by heating and cooling the solution. The energy associated with this process is calculated using the specific heat capacity (C_p, J kg^{-1} K^{-1}) as follows, where m_{heated} is the mass involved in the process, T_{final} is the target temperature, $T_{initial}$ is the starting temperature of the solution, and η is the efficiency of the process, here assumed as 0.9:

$$E_{heating}[J] = \frac{m_{heated} \cdot C_p \cdot (T_{final} - T_{initial})}{\eta}\ . \quad (3)$$

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Solvent management scenarios (Z7–9) are concerned with solvent reuse and recovery. Dickey et al. [60] demonstrated that a solvent can be reused up to three times without compromising the success of zein extraction. The Cheryan process also implements a solvent recovery step, usually through distillation, adsorption or membrane separations, with minimal waste generation. Membrane-based solvent recovery can reportedly reduce the CO_2 intensity of the process by 95% [61]. Herein, we assume 90% solvent recovery, the typical value in nanofiltration systems.

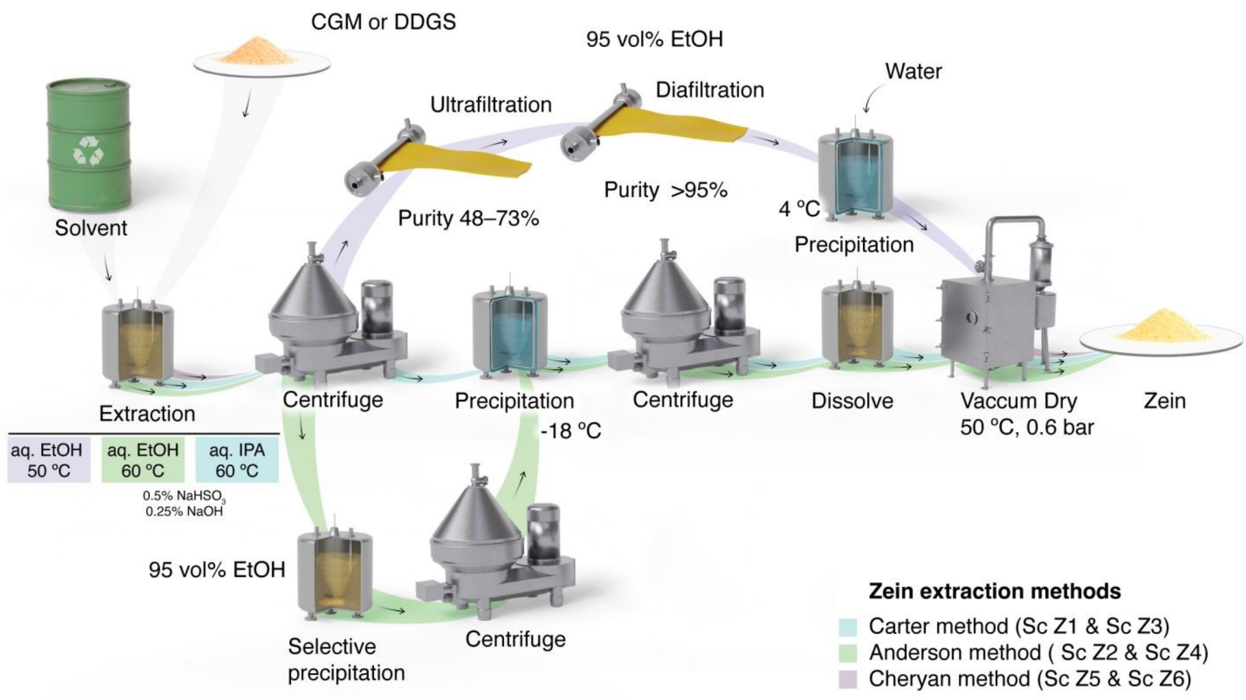

Figure 2. Extraction of zein through the Carter, Anderson, and Cheryan methods. The Anderson method modifies the original Carter method with a precipitation/centrifugation step after crude zein extraction and the Cheryan method adds ultrafiltration and diafiltration for zein concentration and purification.

Table 1. Different scenarios of zein extraction used in the analysis. Scenarios Z1–4 compare the effect of zein source and extraction solvent, scenarios Z5–6 compare zein purification methods, and scenarios Z7–9 compare solvent management methods.

Scenario	Method	Feedstock	Solvent	Solvent consumption (kg kg^{-1})	Solvent treatment	Purification	Yield (%)	Purity (%)	Extraction efficiency (%)
Z1	Carter	CGM	IPA	44.4	No	Precipitation	23.4	83.2	51.2
Z2	Anderson	CGM	EtOH	30.03	No	Precipitation	34.3	91.1	82.1

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Z3	Carter	DDGS	IPA	1030	No	Precipitation	1	58.6	10.8
Z4	Anderson	DDGS	EtOH	219	No	Precipitation	4.7	78.5	67.5
Z5	Cheryan	DDGS	EtOH	184	No	Precipitation	10.32	44	20.16
Z6	Cheryan	DDGS	EtOH	367	No	Ultrafiltration, Diafiltration	6.72	97	28.97
Z7	Sc Z3			343	Reuse				
Z8	Sc Z3			103	Recovery				
Z9	Sc Z6			37	Recovery				

2.2.2 Ethyl lactate production

Ethyl lactate is produced via the esterification of lactic acid and EtOH with water as a byproduct (Figure 3). Lactic acid and EtOH can be obtained synthetically or from natural sources. Herein, lactic acid and EtOH were obtained synthetically (E1), from corn grain (E2), from corn grain and corn stover (E3), or from corn stover (E4) (Table 2). The production methods of lactic acid and EtOH were based on data from Ecoinvent V3.10, the Manandhar method using corn as the feedstock for lactic acid production [55], and the Wang method, which cogenerates EtOH and lactic acid from corn stalks [56].

Table 2. Different scenarios of ethyl lactate production

Scenario	Source of lactic acid	Method	Source of EtOH	Method
E1	Synthetic	Ecoinvent	Synthetic	Ecoinvent
E2	Corn grain	Manandhar	Corn grain	Ecoinvent
E3	Corn stover	Wang	Corn grain	Ecoinvent
E4	Corn stover	Wang	Corn stover	Wang

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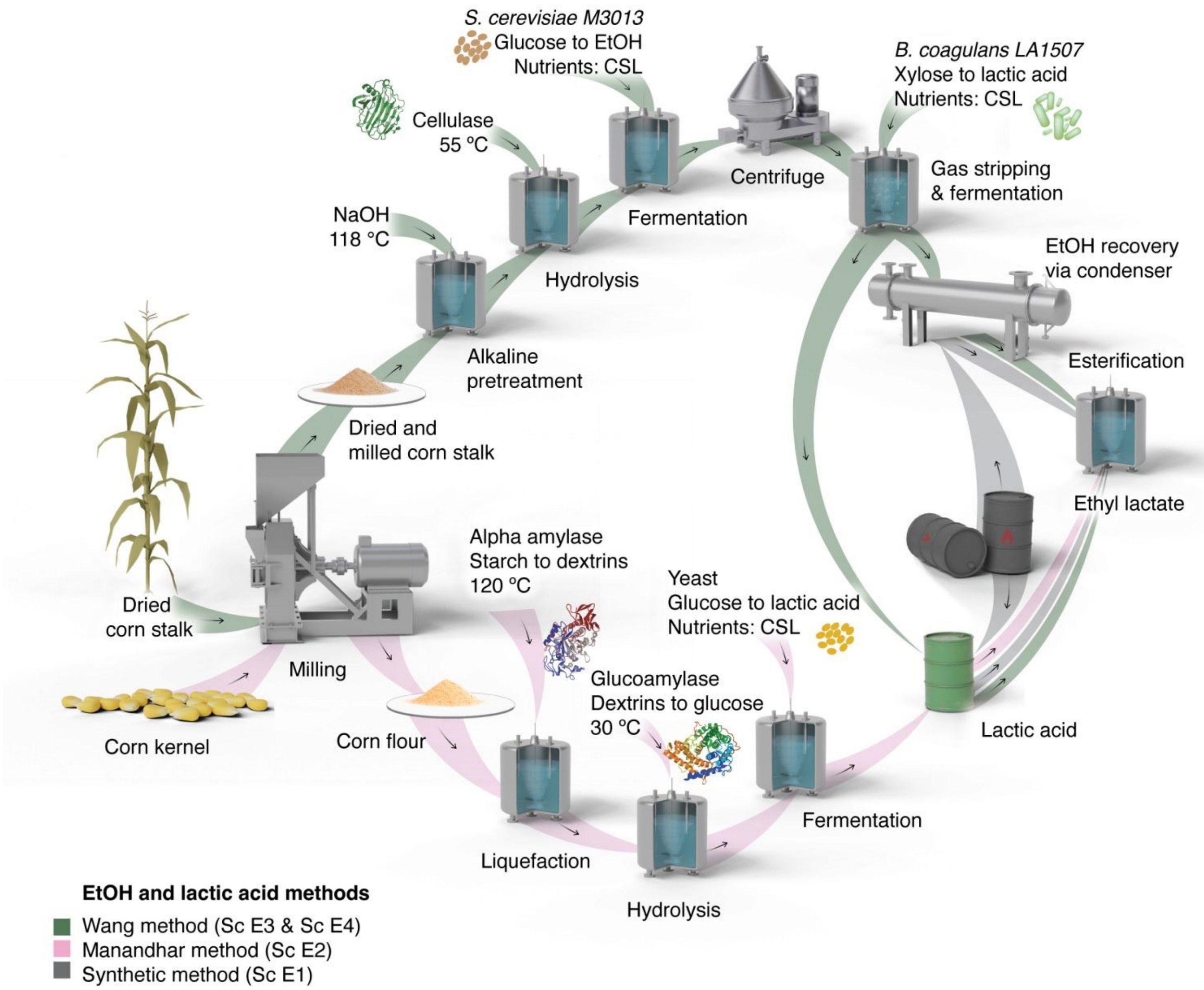

Figure 3. Ethyl lactate production from corn via esterification of lactic acid and EtOH. Lactic acid was obtained from corn grain using the Manandhar method, whereas EtOH and lactic acid were cogenerated from corn stalk using the Wang method. The synthetic methods were obtained from the Ecoinvent database. CSL: Corn-steep-liquor.

Manandhar et al. [46] produced lactic acid from corn grain to complement corn-based EtOH production. Comparing the fermentation pathways involving different microorganisms (bacteria, fungi and yeast), they found that the bacterial production pathway requires maintenance of the optimal conditions: pH = 5–7, temperature = 40–45 °C, nutrient-richness, and sterility. The pH is maintained through continuous neutralization of lactic acid. Fungi can ferment at lower pH than bacteria but produce lower yields. Therefore, the present work utilizes genetically engineered yeast because it produces lactic acid at low pH, reducing the need for neutralization agents. First, the corn grain was milled into flour, exposing the starch to the enzymes

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that efficiently degrade starch into its constituent sugars. Next, the mixture was cooked at high temperature (110–135 °C) and then gradually cooled to 30 °C. During this process, α-amylase converts starch to dextrins and glucoamylase degrades dextrins to glucose. Fermentation to lactic acid is performed by yeast, utilizing corn-steep-liquor glucose and diammonium phosphate as the nutrient and nitrogen sources, respectively. Subsequently, the broth was filtered and lactic acid was purified through esterification with methanol. Finally, the product was distilled, hydrolyzed, and dried to obtain a high-purity product. The electricity and steam-energy consumptions of this process are given in the original report [46].

The Wang method [47] begins with corn-stalk harvesting, chopping and drying at 80 °C, followed by milling to reduce the particle size and obtain dry corn stalk. Adding water and NaOH, the solid content was maintained at 10%, heated to 118 °C, and washed to neutral pH, obtaining an alkali-treated corn starch. The enzymatic hydrolysis was performed using cellulase at 55 °C, followed by vacuum filtration. The supernatant was subjected to EtOH fermentation in the presence of *Saccharomyces cerevisiae* and corn-steep-liquor powder in aqueous NaOH. The cells were removed by centrifugation and the supernatant was retained for lactic acid production. Air was bubbled through the fermentor, providing oxygen for the aerobic growth of *Bacillus coagulans*. Oxygen also carries EtOH through single-pass gas stripping, reducing the energy need for EtOH separation.

We compared four scenarios of ethyl lactate production (Table 2): Sc E1 synthetic, in which EtOH and lactic acid were synthetically produced, Sc E2, in which EtOH and lactic acid were obtained from corn grain, and Sc E3, in which EtOH and lactic acid were sourced from corn grain and corn stover, respectively. In scenario E4, ethanol and lactic acid were both produced from corn stover.

2.2.3 Zein membrane production

The membranes are produced through phase inversion. First, 23 wt% zein was dissolved in ethyl lactate to prepare the dope solution (Figure 4), which was stirred to obtain a viscous homogeneous solution [22]. Thin films were cast using a film applicator with a casting knife and then immediately immersed in a water-containing nonsolvent bath. The effects of different raw-material sources on the environmental footprint of this process were investigated in three scenarios: optimistic (M1), pessimistic (M2), and realistic (M3) (Table 3). These scenarios reflect the variations of the environmental burdens associated with the selection of different zein production pathways.

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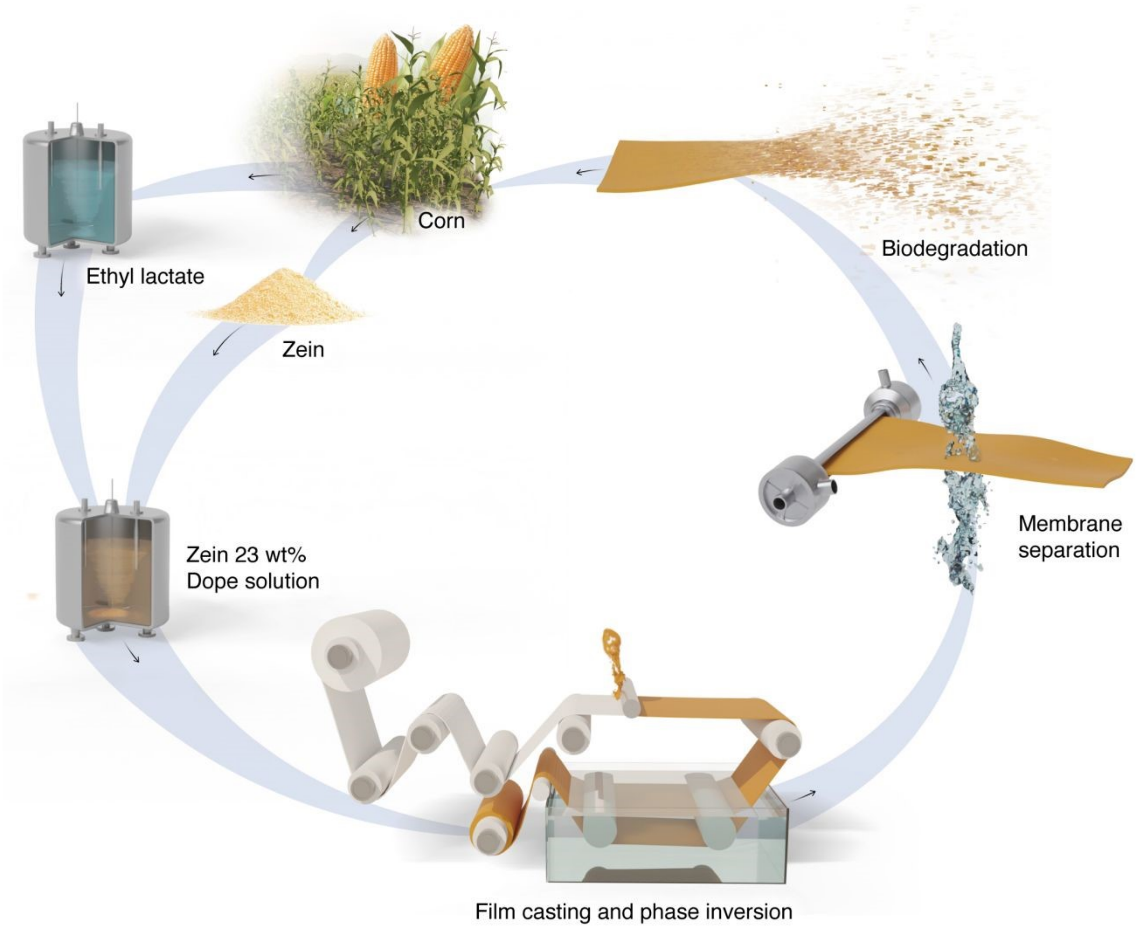

Figure 4. Production of a zein membrane from zein and ethyl lactate as the raw materials in the dope solution.

Table 3. Different scenarios of zein-based membrane production. The zein (Z2, Z4, Z9) and ethyl lactate (E3) scenarios are detailed in Tables 1 and 2, respectively.

Scenario	Zein source	Ethyl lactate source
M1	Sc Z2	Sc E3
M2	Sc Z4	Sc E3
M3	Sc Z9	Sc E3

3. Results and discussion

3.1 Hotspot analysis of the zein extraction

Figure 5 shows the environmental impacts of producing 1 kg of zein under different scenarios (Table 1). Scenarios Z3 and Z4 using the DDGS feedstock exhibited the highest GWPs (5,300 and 760 kg CO_2 eq, respectively). Z3 required approximately 5 times more solvent than Z4 to compensate the approximately 6-

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times lower zein extraction efficiency in this scenario than in Z4. Conversely, scenarios Z1 and Z2 using the CGM feedstock emitted only 232 and 107 kg CO_2 eq, respectively.

The land uses were 72 and 460 m^2a crop eq per kg zein, respectively, in scenarios Z3 and Z4, and 7 and 66 m^2a crop eq per kg zein, respectively, in scenarios Z1 and Z2. The land use impact in scenario Z4 was 66 times that of Z1. The land use requirement of Z4 is raised using corn-derived EtOH. Similarly, scenarios Z3 and Z4 consumed up to 20 times more water (20 and 11 m^3 per kg zein, respectively) than Sc Z1 and Sc Z2 (1.0 and 1.7 m^3 per kg zein, respectively). These results in scenarios Z1 and Z2 can be attributed to the higher availability of zein and higher zein yield from the CGM scenarios (23.4–34.3%) than from the Z3 and Z4 DDGS scenarios (1.0–4.7%; Table 1). Overall, CGM is a more sustainable zein source than DDGS.

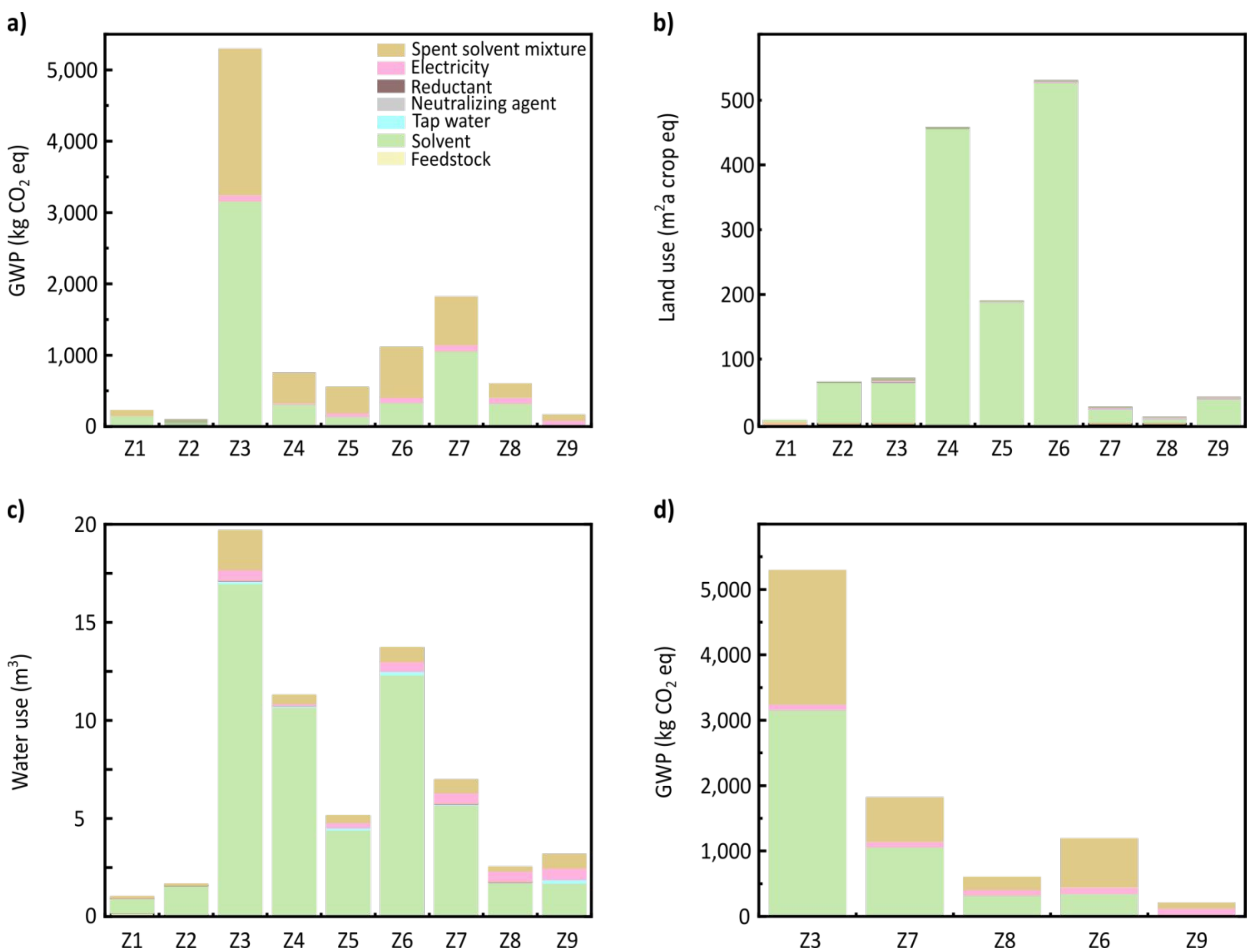

Figure 5. Environmental impacts of zein extraction through different scenarios presented in Table 2: (a) global warming potential, (b) land use, (c) water use, and (d) solvent management evaluation. Integration of solvent reuse or recovery reduces the GWP of zein extraction. The functional unit is 1 kg of zein. GWP: global warming potential.

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3.1.2. Solvent selection

Solvent exerts a major environmental impact across the different categories, contributing >95% to the GWP of zein extraction in Scenarios Z1–Z6. Specifically, the solvent accounts for approximately 42% ± 15%; while treatment and disposal of the spent solvent contribute an additional 54% ± 13%. The contributions of fresh solvent alone are elevated in the other categories. For instance, fresh solvent contributes 86% ± 25% of the water use and 85% ± 6% of the land use of zein extraction. The contribution to spent solvent is modest (typically <9%). The particularly high contribution of solvents to the GWP is driven by their incineration after use, which releases additional CO_2.

Scenarios Z2 and Z4 using EtOH exhibited lower GWPs (106 and 760 kg CO_2 eq, respectively; Figure 5a) than Z1 and Z2 using IPA (232 and 5,300 kg CO_2 eq, respectively). This reduction may be attributed to the higher polarity of EtOH, which enhances the solubilization and extraction efficiency of zein. Importantly, these reductions are attributable to the solvent and slight variations in the Anderson method (specifically, the addition of a selective precipitation step). The Anderson method using CGM and DDGS achieved higher zein recoveries (82% in Z2 and 67% in Z4) than the Carter method (51% in Z1 and 10% in Z3). This difference is primarily attributable to the solvent selection and modified extraction conditions of the Anderson method, suggesting that the extraction method can be improved and small variations can largely influence the extraction efficiency. Anderson modified the Carter process by adding a selective zein precipitation step before cold precipitation, which improves the yield and quality of extraction.

This trend was inverted for land use (Figure 5b): the EtOH-based (Sc Z2 and Sc Z4) scenarios utilized more land resources (66 and 460 m^2a crop eq per kg of zein, respectively) than the IPA-based scenarios Sc Z1 and Sc Z3 (8 and 72 m^2a crop eq per kg of zein, respectively). This result is primarily attributable to the biomass origin of EtOH, which (unlike fossil-based IPA) requires land use. Water use only moderately differed among the solvent scenarios (Figure 5c): Sc Z2 and Sc Z4 consumed 2 and 11.3 m^3 of water per kg zein, respectively, whereas Sc Z1 and Sc Z3 consumed 1.0 and 20 m^3 of water per kg zein, respectively. These variations can be attributed to the solvent production pathways.

The solvent management results (Figure 5d) present scenarios that improve the efficiency of solvent use. To explore potential improvements in process sustainability, we considered solvent reuse (Z7) and solvent recovery (Z8 and Z9) in scenarios Z3 and Z6, with high solvent intensity and large CO_2 emissions. Solvent reuse (scenario Z7) reduced the overall environmental footprint of Z3 by 65.57%, whereas solvent recovery (scenarios Z8 and Z9) reduced the footprints of Z3 and Z6 by 88.5% and 83.7%, respectively. Therefore, integrating solvent reuse or recovery can considerably reduce the environmental impact of zein production.

The sustainability of selected processes (Z3, Z7 and Z8) was compared through the Path2Green metric [62]. It is based on the 12 principles of green extraction and considers simultaneously environmental,

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economic and social aspects. It ranges from not environmentally friendly (–1) to highly environmentally friendly (+1). The scores obtained are illustrated in Figure 6, all of them positive showing a moderately sustainable profile. This reflects favorable aspects of zein extraction including its scalability, the use of recommended solvents, the ready to use extracts with multiple potential applications, the exhaustive extraction from corn biomass and its feasibility to be transformed in the area where it is produced. Socially and economically the process has benefits as well because large-scale biorefineries can support local communities by generating jobs, while using solvents that protect human and environmental health. However, the process also has its limitations as it relies on a monoculture crop, and requires high solvent inputs that also generate waste.

Scenario Z3 achieved a score of 0.152 indicating a slightly positive yet limited sustainability performance due to the highest virgin solvent consumption and high mass going to waste (90%). The incorporation of solvent reuse (Scenario Z7) and recovery (Scenario Z8) improved the score to 0.495 and 0.545, respectively, due to the considerable reduction of virgin solvent use and the waste generated to 33% and 9%, respectively. This value is comparable to the results reported from macauba kernel byproduct valorization through Soxhlet conventional extraction approach (0.5) and a high-pressure supercritical fluid extraction (0.6) obtaining 4 different ingredients including a protein-rich fraction [63].

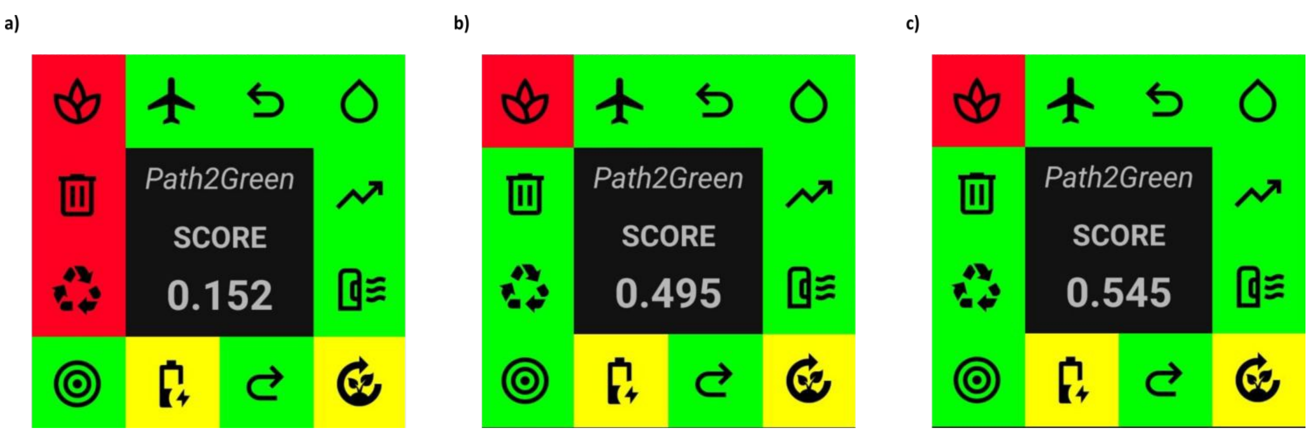

Figure 6. Pictograms showing the score obtained from Path2Green based on the 12 principles of green extraction for the extraction of zein in different scenarios: (a) Z3, the initial process; (b) Z7, involving solvent reuse; and (c) Z8, involving solvent recovery. The values are from –1 to +1, the highest representing a more environmentally friendly process. Each square around the score represents one principle, and a color is attributed based on its compliance from high (green) to low (red).

3.1.3. Purification

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Scenario Z5, in which zein was purified through precipitation alone, showed lower values of the three midpoint categories (GWP, land use, and water use) than the ultrafiltration and diafiltration scenarios. However, it also showed a lower zein purity. Specifically, the GWP, land use and water use in Z5 were 537 kg CO_2 eq per kg of zein, 190 m^2 a crop eq per kg of zein, and 5 m^3 per kg zein, and in Z6 were 1,160 kg CO_2 eq per kg, 528 m^2 a crop eq per kg of zein, and 14 m^3 per kg zein, respectively. Z5 achieved lower values in all categories than Z6, due to the lower amount of solvent used.

Scenario Z9 with the use of membranes for ultrafiltration/diafiltration is preferred to achieve a higher zein purity, and overall lower environmental impact. Notably, solvent use contributes more to the environmental impact of the overall zein production than the additional energy demand of membrane separations. Moreover, integrating membrane separation facilitates solvent recovery, further reducing the environmental footprint of zein extraction. Membrane separation is a promising pathway toward sustainability [64], particularly in industries such as protein purification where high product purity is necessary. By reducing resource consumption and emissions, membrane-based purification can improve the efficiency and sustainability of production systems.

3.2 Environmental impact of ethyl lactate production

Figure 7 depicts the environmental impacts of ethyl lactate production under the scenarios detailed in Table 2. The GWP, land use, and water use only modestly differed in scenarios E1–E4 with different sources of ethyl alcohol (synthetic, corn grain, and corn stover). The average GWP was 7.9 ± 1.6 kg CO_2 eq per kg ethyl lactate. However, some trends are observed: fully synthetic E1 shows the lowest emissions (5.91 kg CO_2 eq per kg ethyl lactate), followed by E3, E2, and E4 with emissions of 7.52, 8.32, and 9.69 CO_2 eq per kg ethyl lactate, respectively. The minimal GWP of the completely synthetic E1 scenario is possibly attributable to the optimized chemical process with high yield and low waste generation. Scenario E4 with the highest GWP requires rigorous control of the fermentation conditions and a large number of purification steps.

Reportedly, the GWP of ethyl lactate production from corn-based biomass is approximately 4.0–5.7 kg CO_2 eq per kg of ethyl lactate [65]. Another study on ethyl lactate production via reactive distillation with structured catalytic packing reported a GWP of 4.59-kg CO_2 eq per kg ethyl lactate [66]. The differences among the obtained GWPs can be attributed to the different databases used, along with disparate models, assumptions, and methodologies. The observed differences demonstrate the need for developing standardized protocols for LCA.

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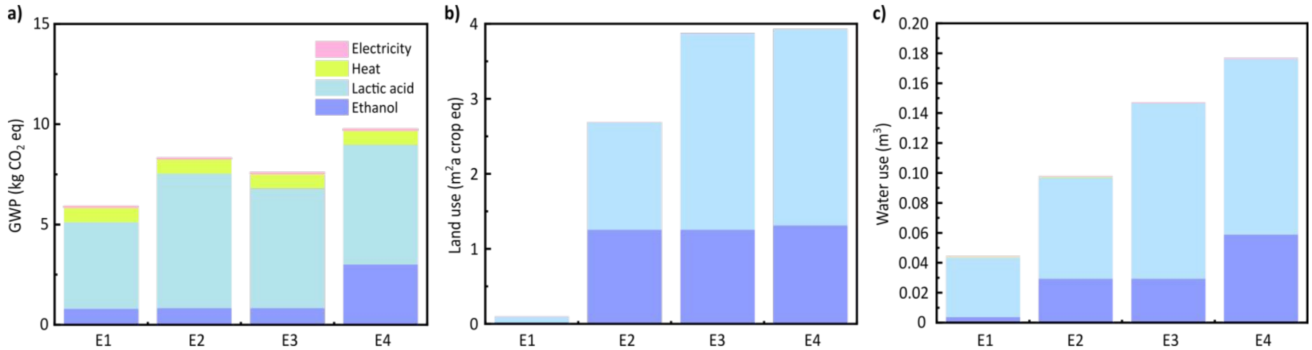

Figure 7. Environmental impacts of ethyl lactate production in the scenarios presented in Table 2: (a) global warming potential, (b) land use, and (c) water use.

Scenarios E3 and E4 utilize two different components of corn for lactic acid production: corn grain and corn stover, respectively. Despite using the same crop, the GWP was 7.51 CO_2 eq per kg ethyl lactate in scenario E3 and 9.69 kg CO_2 eq per kg ethyl lactate in scenario E4. Corn stover is often assumed to impose a minimal environmental burden because it comes from corn residues. Its highest impact on GWP herein is likely attributable to the lower yield and higher energy demand of pretreating and fermenting the lignocellulosic biomass of corn stover.

In addition, E4 contributes a higher proportion of EtOH to the GWP (3 kg CO_2 eq per kg ethyl lactate) than the synthetic E1 scenario (0.80 kg CO_2 eq per kg ethyl lactate) and the corn grain-based E2 and E3 scenarios (both 0.83 kg CO_2 eq per kg ethyl lactate). As highlighted in this comparison, a bio-based approach is not necessarily environmentally preferable. Although corn grain and corn stover are renewable biomasses, their emissions depend on the efficiency and scalability of processing.

Lactic acid was the main source of emissions and resource consumption, with a GWP of 73.6% ± 8.6%, a water use exceeding 76%, and a land use exceeding 65%. The land use was minimized in the fully synthetic scenario E1 (0.10 m^2a crop per kg ethyl lactate) because the ethyl lactate was derived from fossil fuels. In scenarios E2–E4, the average land uses were 3.5 ± 0.7 m^2a crop per kg ethyl lactate. The water use was also much lower in E1 (0.044 m^2 per kg of ethyl lactate) than in E2–E4 (0.14 ± 0.04 m^2 per kg of ethyl lactate). Scenarios E2–E4 require a comparatively large amount of water for biomass pretreatment and hydrolysis during lactic acid production.

3.3 Environmental impact of zein membrane production

Figure 8 shows the environmental impacts of membrane production using zein produced through different processes and ethyl lactate from E3 obtained from corn. Scenario M2 exerted the highest impact on all three categories: GWP, land use, and water use. In scenario M1 and M3, extracting zein through scenario

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Z2 and Z9, the GWP (4.98 and 7.96 kg CO_2 eq per m^2 of membrane) was 27% compared to that of M2, which extracts zein through scenario Z4 (29.87 CO_2 eq per m^2 of membrane). The land use was approximately 10 times higher in scenario M2 (17.62 m^2 a crop per m^2 of membrane) than in scenarios M1 and M3 (2.94 and 2.06 m^2 a crop per m^2 of membrane, respectively). The water use trended similarly to the land use. Scenario M2 required 0.44 m^3 of water per m^2 of membrane, exceeding the water uses of M3 and M1 (0.11 and 0.08 m^3 of water per m^2 of membrane, respectively) by approximately 2 and 6 times, respectively. The lower impact of M1 is attributed to the higher zein availability and more efficient extraction process in this scenario than in the other scenarios, leading to lower resource consumption per unit of membrane. Conversely, the high impacts of scenario M2 can be primarily explained by the inefficient extraction and lower availability of zein from DDGS. Scenario M3 balances purity, and solvent use through the optimized solvent recovery, offsetting some of the emissions and resource use associated with its solvent-intensive production that can benefit the use of DDGS obtained from ethanol industry for zein extraction.

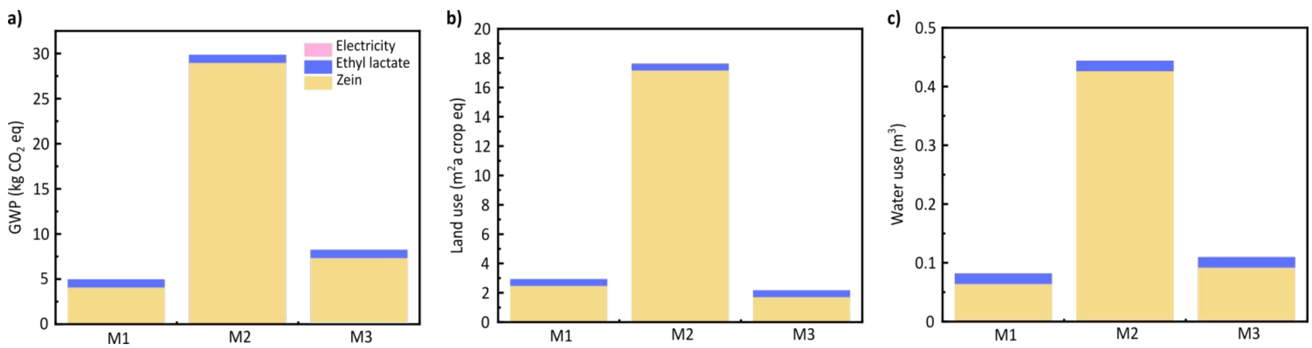

Figure 8. Environmental impacts of zein membrane fabrication in the different scenarios presented in Table 3: (a) global warming potential, (b) land use, and (c) water use.

Overall, these results highlight the importance of feedstock selection, process efficiency, and solvent management strategies in minimizing the environmental footprint of bio-based membrane production. Future work should focus on increasing the efficiency of the zein extraction method and implementing and enhancing solvent recovery to further reduce the emissions and resource use. Aligning with previous findings on polyvinylidene fluoride and polytetrafluoroethylene membranes, the environmental effect (positive or negative) of replacing fossil-based polymers with a biopolymer depends on production [45]. The 12 principles of green membrane materials and processes highlight the need for comprehensive LCAs to evaluate the environmental impact of membrane production and identify opportunities for improvement [34].

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Conclusions

The environmental impacts of zein protein, ethyl lactate (a green solvent), and nanofiltration membranes derived from these compounds were assessed through cradle-to-gate LCAs. The zein extraction process was evaluated through a complete analysis of CGM and DDGS as the main feedstocks, EtOH and IPA as solvents, and precipitation and ultrafiltration purification methods.

Our results demonstrate that the environmental sustainability of protein-based membrane manufacturing is predominantly determined by upstream extraction processes rather than downstream steps. The extraction solvent was identified as an environmental hotspot, exerting at least 95% of the environmental impact of zein production. Besides burdening the environment, solvents pose economic challenges because they are consumed in large quantities during low-yield extractions.

The zein production largely depends on the selected feedstock, which implicitly influences the environmental impact of the process. Lower yield processes require more raw material and solvents to produce the same amount of zein as a high yield process. High-protein raw materials such as CGM reduce the biomass quantity, solvent amount, and energy consumption of zein production. Nevertheless, the zein yields were modest and could be improved through process optimization.

Integrating membrane technologies and solvent management strategies can balance the product quality and environmental impact of the processes. Reuse and recovery of the solvent can reduce the environmental impact of zein extraction by up to 68% and 89%, respectively.

The GWP was little affected by the feedstock of ethyl-lactate production (synthetic, corn grain, or corn stover) but the bio-based feedstocks consumed substantially more water and land resources than the fossil-based synthetic routes. These resources were required for corn cultivation.

Notably, the selected material's (zein in the present case) production process determines the environmental impact of the resulting membrane. Valorizing agricultural byproducts does not automatically translate to sustainable production. These findings highlight the limitations of evaluating sustainability solely based on material origin or solvent properties, and emphasize the need for system level performance and resource efficiency.

An LCA during the early design phases of membrane development can evaluate the sustainability potential of the membrane and identify environmental hotspots that facilitate process optimization. All materials required for membrane production are derivable from corn byproducts and can be integrated into the current corn biorefineries, thus closing the sustainability loop. Our LCA approach is also applicable to other protein-extraction and waste-valorization processes.

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Data availability

The data supporting the findings of this study are provided in the Supporting Information.

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Contributions

CO: Writing – original draft, Validation, Methodology, Investigation, Visualization, Formal analysis, Data curation; AC: Validation, Methodology; GS: Writing – review & editing, Visualization, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this article.

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Data availability statement

The data supporting the findings of this study are provided in the Supporting Information.