Pedro Patrique Ferreira da Silva

# Environmental performance of leather and leather-like materials applied to footwear

Master's thesis in Industrial Ecology Supervisor: Johan Berg Pettersen Co-supervisor: Kamila Krych July 2023

Master's thesis

Norwegian University of Science and Technology Faculty of Engineering Department of Energy and Process Engineering



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#### Abstract

The growing concerns surrounding the environmental impacts of leather production have spurred the emergence of alternative materials in the market. Synthetic alternatives made of polyurethane and polyvinylchloride do not solve the problem, as they also pose burdens to the environment. This prompts the rise of biobased leather-like materials such as Piñatex® and Desserto®, that are claimed to be more sustainable than the industry staples. However, as many of these materials are still in the initial stages of technological development, they often lack comprehensive environmental assessments. In this study, we compared the environmental performance of eight footwear materials using a hybrid methodology that combines qualitative and quantitative assessments. We applied the Lifecycle Screening of Emerging Technologies (LISET) framework and comparative streamlined Life Cycle Assessment (LCA). Our findings reveal that, among the set footwear materials, bovine leather exhibits the highest environmental impacts across most impact categories, but it excels in terms of durability. Furthermore, we observed that the environmental impacts of biobased materials are primarily influenced by the scale of production, particularly in relation to non-optimized energy consumption. We also identified limitations in data availability and quality, as well as resulting uncertainties stemming from modeling assumptions.

#### Keywords

Life Cycle Assessment; Sustainable footwear; Vegan leather; Leather alternatives; Leather-like materials; Environmental performance.

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### Abbreviations

- BCS = Basic chromium sulfates
- CC = Climate change
- CED = Cumulative energy demand
- DMF = Dimethylformamide
- FE= Freshwater eutrophication
- FEP = Freshwater eutrophication potential
- FU = Functional units
- GWP100 = Global warming potential over 100 years
- HHV = Higher heating value

- LCA = Life Cycle Assessment
- LCI = Life Cycle Inventory
- LiSET = Lifecycle Screening of Emerging Technologies
- LLM = Leather-like material
- MSDS = Material safety data sheet
- PALF = Pineapple leaf fibers
- PEFCR = Product Environmental Footprint Category Rules
- PLA = Polylactic acid
- PMF = Particulate matter formation
- PMFP = Particulate matter formation potential
- PPE = Personal protective equipment
- PU = Polyurethane
- PVC = Polyvinyl chloride
- TA = Terrestrial acidification
- TAP = Terrestrial acidification potential
- WC = Water consumption
- WCP = Water consumption potential

**DISCLAIMER**: I authored this thesis as a manuscript for the Journal of Cleaner Production, published by Elsevier. The findings described here are new to the field, as the literature research showed last semester. The manuscript builds on knowledge acquired in the Industrial Ecology fall project and expands it with more elaborate methodology.

#### 1. Introduction

The sustainability of bovine leather has caused many discussions over the years (Kumar & Joshiba, 2020; Nithyaprakash et al., 2020; Ramchandani & Coste-Maniere, 2020). This multipurpose material is an integral part of industries like fashion, furniture, and automotive. However, the process of transforming animal hides into finished leather, commonly known as tanning, has significant environmental impacts. Besides the direct methane emissions and deforestation caused by cattle ranching and feed production (Rudel et al., 2009), the leather industry produces one of the highest concentrations of toxic effluent per unit output by using about 130 different chemicals during tanning (Kumar & Joshiba, 2020). As Buljan et al. (2000) show, 1000 kg of pickled hides requires approximately 450 kg of added chemicals, of which 90% is discarded as waste.

Among these chemicals are basic chromium sulfates (BCS). Chromium-based tanning represents 80% of the industry (UNIDO, 2017), offering more resistance and color retention than vegetable-based alternatives (Kokkinos & Zouboulis, 2020). In exchange, the health of workers is sacrificed given that chromium is recognized as a potent carcinogenic agent (Guertin et al., 2004; Wang et al., 2017). On top of that, Shakir et al. (2012) state that the tanneries' effluent is frequently released into the environment without proper treatment, potentially harming local communities and ecosystems.

New alternatives to chromium-tanned leather are gaining space in footwear production. They encompass both different tanning processes (e.g., vegetable tanning and chromium-free tanning), or different leatherlike materials (LLMs). Polyurethane (PU) and polyvinyl chloride (PVC) are two fossil based LLMs (Rathinamoorthy & Kiruba, 2020) that were developed in the 1950s and 1960s as an inexpensive yet resistant and visually appealing replacement for the animal product (Fernandes, 2021). However, they entail new challenges, as they are reported as microplastics releasers (Bai et al., 2017) and known for the dioxins emissions when incinerated (Wool, 2013). Moreover, their fossil origin is not desirable as society moves towards a circular economy in which oil-based materials are phased out (Shogren et al., 2019).

The biobased LLMs denote a new generation of alternatives to leather in fashion. They come from sources like pineapple leaves fibers (PALF), apple skin, mycelium, bacterial cellulose, cork, vine, cactus, among others (Fernandes, 2021; Hildebrandt et al., 2021; Ramchandani & Coste-Maniere, 2020; Waltz, 2022). Biobased LLMs explore a wide range of agricultural waste and subproducts as they trend in high-end apparel, footwear, and accessories (Harris, 2022; Nithyaprakash et al., 2020; Ramchandani & Coste-

Maniere, 2020). Additionally, they represent a viable option for adepts of a vegan lifestyle (Choi & Lee, 2021; Jung et al., 2016; Minh & Ngan, 2021; Walcher & Ihl, 2020).

Many claims are made regarding biobased LLMs, from associating the word 'vegan' to socially and environmentally responsible (Ramchandani & Coste-Maniere, 2020), to coupling the terms 'biobased' and 'eco-friendly'. However, apart from few scientific studies that cover the environmental assessment of biobased materials, e.g., Bründl (2022), Hildebrandt et al. (2021), and Williams et al. (2022), the comparison of such alternatives to industry staple like bovine leather and fossil-based synthetic materials is demure and not comprehensive.

The standard way to verify these statements is through a Life Cycle Assessment (LCA), which is a widely recognized tool for assessing the environmental impact of product systems (Guinée, 2002). Many LCA studies have been conducted to measure the environmental impact of bovine leather, namely Baquero et al. (2021), Laurenti et al. (2017), Navarro et al. (2020), Notarnicola et al. (2011), Tasca & Puccini (2019) UNIDO (2017), Yu et al. (2021), among others. However, the data availability of emerging technologies such as biobased LLMs can limit a conventional LCA, which highlights the need for alternative approaches, such as hybrid methodologies that combine qualitative and quantitative data (Hung et al., 2020).

Therefore, the aim of this research was to unveil the environmental performance of biobased leather-like materials when compared to current footwear staple materials, namely bovine leather, and fossil-based synthetics. We achieved that by adopting a hybrid methodology, in which impacts such as carbon emissions, energy demand, water use, are alongside qualitative aspects like durability and biodegradability. The overarching objective is to contribute to the sustainable fashion literature by providing a peer-reviewed source about the environmental sustainability of footwear materials.

#### 2. Methods

The methodology of this analysis is composed of two main parts: the Lifecycle Screening of Emerging Technologies (LISET) framework which applies qualitative and semi-quantitate data, and a streamlined Life Cycle Assessment (LCA), which is a fully quantitative framework. **Figure 1** shows the hybrid methodology and a brief explanation of both follows.



*Figure 1* – *Flowchart of hybrid methodology for environmental performance analysis of footwear materials. There are two parts in the flowchart, one green (LiSET) and one pink (LCA).* 

#### 2.1. Lifecycle Screening of Emerging Technologies (LiSET)

The Life Cycle Screening of Emerging Technologies (LiSET) framework seems promising in comparing the environmental performance of materials at a lab or pilot scale, which is the case for most biobased LLMs. Developed by Hung et al. (2020), this framework is specifically designed to evaluate technologies in their preliminary stages of development and can effectively identify data gaps through a combination of qualitative and semi-quantitative data analysis. A pre-LiSET step selects technologies and sets the scope of comparison, followed by the five main phases described in the framework.

#### 2.1.1. Pre-LiSET: Selected candidates and scope

The LiSET framework requires the selection of a reference technology and alternatives, all referred to as candidates (Hung et al., 2020). Here, the reference candidate is chromium-tanned bovine leather, and the alternatives are vegetable-tanned bovine leather, fossil synthetic leather (PU and PVC), and biobased alternatives, from sources like pineapple leaf fibers (PALF), cactus, kombucha bacterial cellulose, and mycelium. We selected the biobased LLMs due to sufficient data availability.

The unit of comparison is 1 m<sup>2</sup> of finished material, with 1.2-to-1.4-millimeter thickness. This unit complies with the Product Environmental Footprint Category Rules (PEFCR) for leather, described by Rosa-Giglio et al. (2018), and it agrees with the functional unit in other studies about leather, like Brugnoli (2012), Laurenti et al. (2017) and UNIDO (2017). The system boundaries for leather cover animal farming up to material finishing, following recommendations by the PEFCR (Rosa-Giglio et al., 2018). Similarly, the alternative product systems also have a cradle-to-gate boundary. We chose footwear as the focal application based on the availability of literature when compared to other items like accessories.

#### 2.1.2. Step 1: Decomposition terms

The first step decomposes the life cycle of a technology in a finite number of contributors to environmental impacts. These terms cover both the direct and embodied impacts of a product in more general terms. We selected 'Environmental Intensity of Materials', 'Material performance', and 'Energy efficiency' as decomposition terms, following the work of Ellingsen et al. (2016).

#### 2.1.3. Step 2: Lifecycle aspects

The second step represents the conversion of the decomposition terms in valuable metrics, also known as lifecycle aspects. The aspects translate into physical properties of each decomposition term. Intrinsic aspects entail immutable properties of the technology, while the extrinsic aspects cover characteristics that depend on the location of the supply chain. We nominated nine aspects: six intrinsic (exposure to chemical hazards, water consumption, tear resistance, non-plastic content, value creation from by-products and waste, and energy demand) and three extrinsic (carbon emissions, terrestrial acidification, and freshwater eutrophication).

#### Environmental intensity of materials

The environmental intensity of materials encompasses two intrinsic aspects: 'Exposure to chemical hazards' and 'water consumption'. We selected the first aspect based on the direct contact of workers with toxic chemicals, such as formaldehyde, chromium-based sulfates, and acids. In addition to the substantial

volumes of these harmful substances being used, tanning chemicals are often handled without the use of protective gear (Garai, 2014), which further emphasizes the importance of this criterion.

We included water consumption as a second intrinsic aspect because it is an important aspect of multiple of the evaluated materials; high volumes of water are used for leather tanning (Baquero et al., 2021; Yu et al., 2021), PU synthesis (Wool, 2013), bacterial cellulose fermentation (da Silva Junior et al., 2022; Fernandes, 2021), and farming biobased materials (Hernández-Becerra et al., 2022; Hildebrandt et al., 2021).

Three extrinsic aspects were selected: 'carbon emissions', 'terrestrial acidification', and 'freshwater eutrophication', as endorsed by Rosa-Giglio et al. (2018). 'Carbon emissions' are the main driver for global warming and, hence, are a crucial point on a product's environmental performance. 'Terrestrial acidification' and 'freshwater eutrophication' are two other major indicators of the environmental impacts of leather, given that both feed production and cattle ranching emit massive quantities of sulfur and phosphorous (Hinckley et al., 2020). These are also two major resource flows highlighted in the planetary boundaries scope (O'Neill et al., 2018).

#### Material performance

In material performance, 'tear resistance', 'non-plastic content', and 'value creation form waste byproducts and waste', are three intrinsic points under analysis. Tear resistance is one of the most important physical characteristics for leather shoes (IULTCS, 2011; Meyer et al., 2021) because it gives a direct idea on the product's durability. Physical properties like durability are important to be accounted for because they are perceived drawbacks for LLMs (Nithyaprakash et al., 2020; Ramchandani & Coste-Maniere, 2020). The experiment to measure tear resistance consists of creating a slit in a rectangular sample and pulling each side in opposite directions until the material is completely torn. The applied force required to separate the two halves is recorded as the tear strength (IULTCS, 2011).

Most vegan synthetic alternatives contain high plastics content (Williams et al., 2022). The percentage of 'non-plastic content' praises materials with a lower likelihood of generating microplastics, while using fewer fossil resources. Finally, 'value creation from by-products and waste' corresponds to the use of co-products, by-products, and waste as feedstock in the manufacturing processes, which gives credit to materials that add value to agricultural waste, like the PALF alternative.

#### Energy intensity

'Energy demand' is the only intrinsic aspect that accounts for both electricity and heat used in the manufacturing processes of the evaluated materials. It is crucial to account for the energy use in leather production as it is inherently connected to the overall environmental performance of the material, especially regarding carbon emission (Laurenti et al., 2017; Rosa-Giglio et al., 2018; UNIDO, 2017).

#### 2.1.4. Step 3: Grading system

Qualitative grades were defined for 'exposure to chemical hazards', 'non-plastic content', and 'value creation from by-products and waste'), based on information provided in patents like Cázarez Duarte & Lopez Velarde (2021), sustainability reports, e.g., Ananas Anam Ltd. (2021), and companies websites. For 'exposure to chemical hazards', we determined the grades by correlating the material safety data sheets (MSDS) of chemical inputs and the categorization made by (EC) N° 1272/2008, in which chemicals are classified from 1 (worst effect) to 4 (milder effects) for hazard classes like acute toxicity. We considered health risks associated with eyes, skin, oral ingestion, and inhalation as preventable with the correct use of personal protective equipment (PPE), while risk like reproductive toxicity and carcinogenicity are not preventable with the use of PPEs alone. As for the remaining categories, three grade intervals (green, yellow, and red) were generated by Jenks natural break optimization, as recommended by Hung et al. (2020). Jenks breaks optimization is based on an algorithm that divides a dataset into homogenous classes (North, 2009), in this case into three coarse grades. As Khan (2012) shows, Jenks optimization prevents misinterpretations caused by grouping samples based on a number or equal intervals. **Table S1** in the supplementary material presents the starting and ending points of each category for this set of candidates.

#### 2.1.5. Step 4: Iterations

The fourth step involves iterating through the first three steps, refining the results through data collection, and addressing any incompleteness. As we added new data sources, we fine-tuned the qualitative scoring, leading to smoother grade intervals. This refinement process, as discussed by Hung et al. (2020), enhances the reliability of the LiSET matrix. **Tables S4-S11** in the supplementary material exhibit all the values used in the LiSET matrix and respective sources.

#### 2.1.6. Step 5: Result matrix

The resolution increases as the screening progresses to a point where each point of data in the matrix comes close to a product flow, elementary flow, or key parameter. The comprehensive breakdown of material requirements and direct emissions was fed into a quantitative LCA, which was the second part of the methodology.

#### 2.1.7. Sensitivity

We evaluated the robustness of the LiSET matrix by performing a local one-factor-at-a-time sensitivity analysis. New matrices were plotted after varying each quantitative data point in +10% and -10%, which is the standard in uncertainty calculation (Saltelli et al., 2019). When a quantitative data point changes, the Jenks optimization algorithm may identify a new natural break that was not present in the dataset before, resulting in a different grading system and, consequently, a distinct matrix. **Figures S1-S4** in the supplementary material display the sensitivity results.

#### 2.2. Streamlined Life Cycle Assessment

An attributional streamlined comparative LCA is performed, following the guidelines in the PEFCR (Rosa-Giglio et al., 2018) for leather. Streamlined LCA refers to a set of methods and tools that aim to reduce the data requirements, simplify the analysis, and accelerate the results of LCA, while maintaining sufficient accuracy and rigor to support decision-making (Alcaraz et al., 2018). This is achieved by adopting streamlining strategies like eliminating the upstream and/or downstream processes, limiting the number of environmental impacts evaluated, mixing qualitative and quantitative data depending on availability, among others (Hung et al., 2020).

LCA complies with the four phases framework described in the ISO 14040:2006 (European Committee for Standardization, 2006a) and ISO 14044:2006 (European Committee for Standardization, 2006b): goal and scope definition, construction of a life cycle inventory, impact assessment, and interpretation of results.

#### 2.2.1. Goal and Scope

The aim of this streamlined LCA is to quantify the environmental performance of the seven materials in **Table 1** to provide scientific support for decision-making when comparing the manufacturing of different footwear materials, while also assessing the limitations. We exclude the mycelium-based alternative due to insufficient information. Potential target audiences include businesses in the footwear industry and researchers investigating the environmental impacts of leather and its alternatives.

Table 1 – Types of bovine leather and leather-like alternatives to be applied in footwear.					

Material	Description
Chromium-tanned bovine leather	Chromium-tanned leather is the reference material in the bovine leather market, with 80% of the market share (UNIDO, 2017). Chromium (III) covalently bonds to collagen molecules in hides, assuring resistance and stability (Zhang et al., 2019).
Vegetable-tanned bovine leather	Plant-based tannins represent an alternative to chromium-based tanning (China et al., 2020). Vegetable-tanned leather has a 15% market share and is more stable over time than the chromium counterpart (UNIDO, 2017).
Polyurethane (PU) leather-like material	This synthetic leather comes from a solution containing fossil plasticizer, i.e., PU, which polymerizes around knitted or nonwoven fabric when submerged in dimethylformamide (Xia et al., 2007). Coating treatments are common.
Polyvinyl chloride (PVC) leather-like material	A solution containing 55% of a fossil polymer (PVC), 40% plasticizer, and 5% filler yields a foamy layer that is glued to a polyester fabric (Gurera & Bhushan, 2018). Texturization and coating treatments follow.
Pineapple leaf fibers (PALF) leather- like material	The content of this nonwoven material includes 70% fibers, 30% bioplastic, i.e., polylactic acid, and 10% fossil plasticizer, namely polyurethane (Hijosa et al., 2013). Due to the natural resistance of PALF, the alternative does not require extra reinforcement. Piñatex <sup>®</sup> by Ananas Anam <sup>™</sup> is the main commercial version of PALF material, with manufacturing in the Philippines and Spain.
Cactus leather-like material	The content of this nonwoven material includes up to 80% of fossil plasticizer, i.e., polyurethane, and 20% cactus leaf powder. It uses a backing fabric to guarantee resistance and durability (Cázarez Duarte & Lopez Velarde, 2021). Desserto® by Adriano Di Marti™ is the main product in the market, with production plants in Mexico.
Kombucha bacterial cellulose leather-like material	This material is a by-product of kombucha production. After the cellulose purification, coloring, and reconstitution, the material dries in a thin layer (da Silva Junior et al., 2022; Fernandes, 2021). The material is commonly reinforced with a backing fabric and coated with natural oils and waxes. Commercial actors include: Polybion™ with Celium™, and BUCHA BIO™ with Hikari™ and Shorai™.

The functional unit (FU) is one square meter of finished material with a thickness between 1.2 and 1.4 millimeters, which is the typical thickness of upper in shoes (Bruno et al., 2007; Laurenti et al., 2017; Notarnicola et al., 2011).

We adopted a cradle-to-gate system boundary for the comparative LCA, which accounts for the upstream and core processes of a product system. For bovine leather, this covers cattle farming and tanning, as per recommendation of the PEFCR (Rosa-Giglio et al., 2018). In the case of LLMs, the boundary encompasses material extraction, farming (for plant based LLMs) and manufacturing. We presented the schematics of each system and respective manufacturing processes in the supplementary material (from **Figure S6** to **Figure S12**). This assessment did not include the downstream processes like wastewater treatment, in line with the PEFCR and the streamlined LCA framework (Arzoumanidis et al., 2017; Moberg et al., 2014).

#### 2.2.2. Life Cycle Inventories (LCI)

The inventories for chromium tanning and vegetable tanning are compiled from Yu et al. (2021) and Baquero et al. (2021), respectively, complemented by data from Canals et al. (2002), Laurenti et al. (2017), Notarnicola et al. (2011), Ömür et al. (2016), Schäfer et al. (2021), Tasca & Puccini (2019), and UNIDO (2017). The *ecoinvent* activity titled 'market for cattle for slaughtering, live weight' comprises the upstream processes associated with cattle ranching. The inputs for slaughtering, preservation, and finishing stages are gathered from Buljan & Král (2019) and Notarnicola et al. (2011).

The inventory for PU LLM is based on the wet method patented by Xia et al. (2007). The authors elaborate on a plethora of approaches to produce PU synthetic leather, but we selected the simplest one known as 'Example 1'. The inventory for manufacturing of PVC LLM follows the four steps presented in Gurera & Bhushan (2018), known as the dry method to produce fake leather. The inventory for pineapple based LLM is based on the patent by Hijosa et al. (2013), that describes the manufacturing process for Piñatex<sup>®</sup>. Although modelling an inventory based on patent literature has its limitations, e.g., the lack of precision regarding the values, it is a frequent practice in LCA studies like Reichmanis & Sabahi (2017).

The inventory for cactus LLM comes from Hernández-Becerra et al. (2022) and the patent by Cázarez Duarte & Lopez Velarde (2021), who illustrate the production of Desserto<sup>®</sup>. The inventory for kombucha LLM is an adaptation from da Silva Junior et al. (2022), which communicate a method to produce a naturally died leather-like material from bacterial cellulose on a laboratory scale. Large scale studies have been done before, e.g., Bründl (2022), but the inventories have not been disclosed. We estimated the water and electricity inputs based on the experience of a biotechnologist involved in the production of similar materials (Claudio J. G. da Silva Junior, personal communication, February 2, 2023), who authored many sources about kombucha LLM production. The full inventories for all materials can be found in **Tables S12-S27** of the supplementary material.

#### 2.2.2.1. Allocation

The production of leather and some leather-like alternatives results in multiple outputs, creating the need for allocation. For bovine leather, 12% of the impacts from cattle raising should be allocated (by mass) to cattle to be slaughtered, out of which 3.5% should be allocated (economically) to hides and skins. During tanning, 60% of the impacts are allocated to finished leather, while the remainder goes to hair, flesh splits, and other by-products (Rosa-Giglio et al., 2018). We allocated 3.4% of the impacts for kombucha production to the cellulose biofilm, following Mohammadshirazi & Bagheri Kalhor (2016). All the impacts

of pineapple production get allocated to the fruit and not the leaves, as Hildebrandt et al. (2021) discuss. The production of the remaining materials, i.e., PU, PVC, and cactus, does not have co-products, which implies that 100% of the impacts in the respective product systems are allocated to the materials themselves.

#### 2.2.3. Life cycle impact assessment (LCIA)

**Table 2Table 2** shows the midpoint impact categories, indicators, and characterization factors that comply with the instructions provided by the PEFCR for leather. All categories are the hierarchic perspective of ReCiPe 2016 v1.03 method, originally described by Goedkoop et al. (2008), except for cumulative energy demand (CED), which was assumed based on Hildebrandt et al. (2021) and Laurenti et al. (2017).

**Table 2** - Midpoint impact categories under study, with their impact category names and abbreviations (Ab.), indicator names and units, and characterization factor names, abbreviations, and units.

Impact category		Indicator		Characterization factor				
Name	Ab.	Name	Unit	Name	Ab.	Unit		
Climate change	CC	Infra-red radiative forcing	W.yr/m²	Global warming potential	GWP100	kg (CO <sub>2-eq</sub> )		
Cumulative energy demand	CED	Energy demand	MJ	Higher heating value	HHV	MJ <sub>-eq</sub>		
Freshwater eutrophication	FE	Phosphorus concentration	yr.kg/m³	Freshwater eutrophication potential	FEP	kg (P <sub>-eq</sub> )		
Particulate matter formation	PMF	PM2.5 intake	kg	Particulate matter formation potential	PMFP	kg (PM <sub>2.5-eq</sub> )		
Terrestrial acidification	ТА	Base saturation	ppt.yr <sup>a</sup>	Terrestrial acidification potential	ТАР	kg (SO <sub>2-eq</sub> )		
Water consumption	WC	Amount of water	m³	Water consumption potential	WCP	m³ (water)		

<sup>a</sup>: The unit *ppt* refers to units of equivalent chlorine.

#### 2.2.3.1. The Activity Browser and database

The streamlined LCA was modelled in the Activity Browser (AB), an open-source, user-friendly software that builds upon Brightway2 (Steubing et al., 2020). The models used data from the *ecoinvent* 3.9.1 database (Wernet et al., 2016).

#### 2.2.3.2. Uncertainties

We calculated the uncertainty of the LCA results through a Monte Carlo simulation. This method transfers the uncertainty of inputs to the outputs by incorporating random sampling error iteratively, which yields a distribution of potential model results (McCandless & Gustafson, 2017; Saltelli et al., 2019).

#### 3. Results

#### 3.1. LISET

**Figure 2** displays the LiSET matrix for bovine leather, fossil-based materials, and biobased alternatives. The grades for each lifecycle aspect range from green (high performance) to red (low performance). When data is incomplete, unclear, or not available, a blank score is assigned, which is the case for most of the candidates in the criteria 'terrestrial acidification' and 'freshwater eutrophication'. Out of the 72 data points in the matrix, 22 of them (30.5%) are red, 22 (30.5%) are yellow, and 12 (16.7%) are green. Additionally, 16 cells (22.3%) score blank, implying no data available in the literature.

To establish a coarse grading system, it is necessary to have a minimum of three data points, with one corresponding to each grade (Hung et al., 2020). However, 'terrestrial acidification' and 'freshwater eutrophication' do not meet this requirement, making it impossible to create three intervals for scoring. Nonetheless, the final matrix includes the values available in the literature with an asterisk. This approach highlights significant data gaps that future studies could address.

The LiSET matrix shows that carbon emissions and energy demand correlate for four out of the eight alternatives: chromium-tanned leather and vegetable-tanned leather score red in both categories, cactus LLM scores green, and mycelium LLM scores yellow. The potential correlation is not as clear for the remaining four alternatives. Despite PU, PVC, and PALF LLMs consuming medium-to-high amounts of energy, their carbon emissions received a green score in the LiSET matrix. For PU synthetic, the energy is used for water heating, particularly during the washing process at 55°C (Xia et al., 2007). This heat can come from many diverse sources, including low-carbon heating systems. As for PALF LLM, the material consumes a high amount of energy per square meter (red score) but has low emissions (green score). This can be attributed to the content of polylactic acid (PLA) from maize (Hildebrandt et al., 2021), which has an energy intensive processing (Chen & Patel, 2012; Wellenreuther et al., 2022). The bacterial cellulosebased material also consumes a significant amount of energy (red score), but it exhibits a yellow score in emissions. As demonstrated by Hildebrandt et al. (2021), most of this energy is derived from the remaining sucrose used for fermentation, which is also referred to as non-oxidized biogenic energy or reducing sugars. This form of energy has a minimal impact on the carbon footprint of a product compared to electricity and heat, primarily due to the small quantity of sugar that is not converted into cellulose (Malbaša et al., 2008) and the inherently low carbon intensity of sucrose (García et al., 2016; Rein, 2010).

		Gen leat	Genuine leather		Fossil-based materials		Bio-based materials		
<ul> <li>High performance</li> <li>Medium performance</li> <li>Low performance</li> <li>Missing data</li> </ul>		Bovine leather (chromium tanning)	Bovine leather (Vegetable tanning)	Polyurethane (PU)	Polyvinylchloride (PVC)	Pineapple leaf fibers (PALF)	Cactus	Kombucha bacterial cellulose (KBC)	Mycelium
	Exposure to chemical hazards (MSDS)								
onment nsity of terials	Water consumption (in L/m²)								
Enviro inte ma	Carbon emissions (in kgCO2-eq/m²)								
al ance	Tear resistance (in N/mm)								
Materi	Non-plastic content								
be	Value creation from by-products and waste								
Energy intensity	Energy demand (in MJ/m²)								
ıer	Terrestrial acidification (in kgSO2-eq/m²)	*						*	
Oth	Freshwater eutrophication (in kgP-eq/m²)	*						*	

*Figure 2* - Lifecycle Screening of Emerging Technologies for leather and leather-like alternatives. An asterisk (\*) denotes data found in the literature that was insufficient for establishing grading ranges for a particular aspect.

Both alternatives for bovine leather received a red grade in terms of water consumption. As discussed by UNIDO (2017), a sizable portion of water usage occurs before the slaughterhouse in cattle farming, in feed irrigation. On the other side of the spectrum, PVC, cactus, and mycelium leather-like materials got a green score. These materials' low water intensity clearly connects to feedstock and manufacturing. For instance, cacti are adapted for desert climate, which implies little to no water inputs in the form of irrigation.

The LiSET matrix reveals that one of the alternatives matched the tear resistance of chromium-tanned leather, pointing out an important trade-off of the materials. To improve resistance of biobased materials, manufacturers incorporate synthetic binders like PU. Cactus LLM, for example, is reported to contain

around 80% of its weight in plasticizer (Alternative Leather Co., 2023), while PALF LLM consists of 30% of total weight in biodegradable plastic (PLA) and 10% in PU (Hijosa et al., 2013). Counterintuitively, kombucha LLM receives a green score in this category because the production applies natural coating products without fossil polymers. As per mycelium and bovine leather, the presence of plasticizers and their concentration depend on the finishing process under consideration, granting these alternatives a yellow score in this aspect (Jones et al., 2021; Rosa-Giglio et al., 2018; Williams et al., 2022).

Certain materials derive value from agricultural waste, such as PALF and mycelium alternatives (green score). While bacterial cellulose can be produced from various waste sources (Nguyen et al., 2021), this assessment assumes its origin from kombucha production. Some producers view the cellulosic biofilm as waste, others consider it a by-product (Rathinamoorthy & Kiruba, 2020), hence the yellow score in the LiSET matrix. The same discussion seems relevant for bovine leather, as the material is commonly depicted as a co-product, a by-product, or as waste from the milk and meat industry (Ramchandani & Coste-Maniere, 2020). However, the most correct classification according to the PEFCR is as a co-product (Rosa-Giglio et al., 2018), hence the yellow score.

**Figure 2** illustrates that all alternatives receive either a red or yellow score in terms of exposure to chemical hazards, as they contain at least one chemical listed as category 1 in the Regulation (EC) No 1272/2008. However, it is important to note that the scores are highly dependent on the manufacturing system, meaning that they could vary if we adopted a different production method.

The sensitivity analysis (**Figure S1** to **Figure S4**, in the supplementary material) shows that the LiSET matrix results are robust. The water consumption grades are more susceptible to changes induced by the variation of value, when compared to carbon emissions or tear resistance that have very few alterations in the original matrix. In most cases, the changes improve the initial score for some alternatives. For example, if the tear resistance of cactus LLM is 10% higher, the material gets a yellow grade in this aspect, instead of the original red.

#### 3.2. Streamlined LCA

Figure 3 to Figure 8 show the results of the streamlined comparative LCA.



= Cattle for slaughtering = Electricity = Heat (natural gas) = Hard coal = Ethylene = Xylene = Heat (not natural gas) = Other

**Figure 3** - Global warming potential of  $1 m^2$  of footwear material. Processes with below 5% contribution were aggregated into "Other".



Figure 4 – Cumulative energy demand of  $1 m^2$  of footwear material. Processes with below 10% contribution were aggregated into "Other"



Figure 5 – Water consumption potential of  $1 m^2$  of footwear material. Processes with below 10% contribution were aggregated into "Other".



*Figure 6* – Terrestrial acidification potential of  $1 m^2$  of footwear material. Processes with below 5% contribution were aggregated into "Other".



*Figure 7* – Particulate matter formation (< 2.5  $\mu$ m) potential of 1 m<sup>2</sup> of footwear material. Processes with below 5% contribution were aggregated into "Other".



Figure 8 – Freshwater eutrophication potential of  $1 m^2$  of footwear material. Processes with below 5% contribution were aggregated into "Other".

The carbon emission results obtained through LCA (**Figure 3**)confirm the carbon emissions scores in the LiSET matrix. The two animal-based materials exhibit the highest carbon emissions per square meter, while PU and cactus LLMs are comparable and fall within the lowest range, along with PVC LLM. The bacterial cellulose material falls within the medium range, supporting the yellow score in the LiSET. Only the pineapple leaf fibers alternative scores higher in LCA than in LiSET, which could be explained by potential differences between the LiSET and LCA assumptions for**Error! Reference source not found.** the degumming p rocess.

Cattle farming is the process that contributes the most to the carbon footprint of leather, both through direct methane emissions and other emissions associated with feed production. This results in a footprint of  $35.19 \text{ kgCO}_{2-\text{eq}}/\text{m}^2$  for chromium-tanned leather, with  $8.2 \text{ kgCO}_{2-\text{eq}}/\text{m}^2$  emitted after the slaughterhouse stage. On the other hand, the vegetable-tanned alternative has the highest footprint in the group at  $36.04 \text{ kgCO}_{2-\text{eq}}/\text{m}^2$ , with 9 kgCO<sub>2-eq</sub>/m<sup>2</sup> emitted after the slaughterhouse. The lowest carbon emissions result belongs to the PVC option, with  $2.98 \text{ kgCO}_{2-\text{eq}}/\text{m}^2$ .

The cumulative energy demand (**Figure 4**) plot follows the same pattern as the global warming potential one, pointing to a potential correlation between the two impacts. Both bovine leather alternatives are in the lead position with 255.3 MJ/m<sup>2</sup> for vegetable-tanned, and 212.5 MJ/m<sup>2</sup> for the chromium-tanned one. In the yellow zone comes kombucha and PALF LLM, with 170.9 MJ/m<sup>2</sup> and 168.4 MJ/m<sup>2</sup>, respectively. In the lower range, one sees alternatives made of cactus (86.4 MJ/m<sup>2</sup>), PU (82.5 MJ/m<sup>2</sup>) and PVC (54.1 MJ/m<sup>2</sup>).

**Figure 5** confirms that almost half of the water consumption potential for bovine leather comes from cattle ranching. The two animal-based alternatives, i.e., chromium- and vegetable-tanned leather, present the second and third highest water footprint among alternatives, around 300 L/m<sup>2</sup>. The LCA results also confirm that PU, PVC, and cactus alternatives place at the lower end of the comparison, with 42.6 L/m<sup>2</sup>, 31.6 L/m<sup>2</sup>, and 43.3 L/m<sup>2</sup>, respectively. The alternative made of pineapple leaf fibers have a medium water consumption both in the LiSET (yellow score) and in the LCA (170 L/m<sup>2</sup>). Although Hildebrandt et al. (2021) explore the influence of maize cultivation (for PLA) to the WCP of PALF material, we found sodium hydroxide used in degumming to be more influential in this category.

The water consumption for bacterial cellulose is different in the LiSET and in the LCA. While the material receives a yellow grade in the qualitative matrix, the LCA results indicate that kombucha LLM has a higher water footprint than genuine leather, amounting to 332 L/m<sup>2</sup>. The allocation of impacts explains the disparity, particularly regarding tea and sucrose, which is a critical aspect of the assessment (Hildebrandt

et al., 2021). Additionally, differences in post-processing methods also play a significant role. The processes involved in cellulose purification and reconstitution, production of natural dyes, and coating with coconut oil increase the water consumption potential, as depicted in **Figure 5**.

We identified a pattern in the plots for terrestrial acidification (**Figure 6**), particulate matter formation (**Figure 7**), and freshwater eutrophication (**Figure 8**). In the three cases, vegetable-tanned leather has the highest impact potential, followed by the chromium-tanned counterpart. The tanning agents have a minor influence in eutrophication and acidification when compared to electricity for tanning and the inputs for cattle farming. In the medium performance cluster, one spots the options made of bacterial cellulose and pineapple leaf fibers. The lower level of readiness of these alternatives plays a key role in determining their TAP, PMFP, and FEP. This is due to the non-optimized energy consumption for manufacturing pilot scale material, which results in electricity driving the impact in the three categories. The PALF option even surpass the yellow performance boundary in FEP, which is a direct result of the majorly fossil-based electricity mix in the Philippines. The materials made of cactus, PU, and PVC place in the green zone for the three categories.

The LCA sensitivity (supplementary **Figure S13**) illustrates that the overall result can change. Although some categories, like CED and FEP, have a wider range of results, i.e., higher uncertainty, other categories oppose that. One clear example is GWP (**Figure S13A**), in which there are three distinctive clusters of results. Materials in the same cluster have overlapping distribution, meaning it is not possible to determine the difference among their results. However, it possible to know that the cluster with chromium-tanned and vegetable-tanned leather has a much higher carbon footprint than the cluster with the PU, PVC, and cactus alternatives due to the distance of the two clusters in the Monte Carlo simulation plots. This agrees with what we have discussed here, although we acknowledge the need for a more refined confidence test in the future.

#### 4. Discussion

#### 4.1. Comparison with literature values

The carbon emissions results (**Figure 3**) of bovine leather align with values previously described (Brugnoli, 2012; Bründl, 2022; Laurenti et al., 2017; Schäfer et al., 2021). The emissions for PU and PVC LLM are 4.36 and 2.98 kgCO<sub>2-eq</sub>/m<sup>2</sup>, respectively, which strongly agree with Schäfer et al. (2021) and Wool (2013). As for kombucha LLM, the footprint is 10.67 kgCO<sub>2-eq</sub>/m<sup>2</sup>, in line with Bründl (2022).

The results for the two plant-based materials, namely cactus and PALF, differ from the values reported in the literature. While Adriano di Marti (2022) describes emissions of 1.39 kgCO<sub>2-eq</sub>/m<sup>2</sup> for cactus LLM, we obtained 4.6 kgCO<sub>2-eq</sub>/m<sup>2</sup>. The main source of this variance is the higher energy demand in our model, which is 86.4 MJ/m<sup>2</sup>, compared to the reported value of 34.33 MJ/m<sup>2</sup>. This energy is used in polymer homogenization and coating processes, which are parts not fully described in the patent by Cázarez Duarte & Lopez Velarde (2021), introducing additional uncertainty to the original results. Additionally, assumptions regarding carbon capture by cacti are quite ambiguous in the original model, which exposes the inexact nature of the results previously reported. As per PALF material, we obtained a footprint of 11.12 kgCO<sub>2-eq</sub>/m<sup>2</sup>, which disagrees with the value reported by Ananas Anam Ltd. (2021) of 2.69 kgCO<sub>2-eq</sub>/m<sup>2</sup>. One explanation for this difference could be the inclusion of credits for prevented biomass burning, which we did not assume in our model. However, it is not possible to confirm if this was the approach adopted by the company due to data disclosure issues.

Water consumption potential is another point of contrast between the model and values presented in the literature. Although the results for chromium- and vegetable-tanned leather are on the high end of the comparison (289.24 L/m<sup>2</sup> and 314 L/m<sup>2</sup>, respectively), they are still relatively low when compared to the estimated 16,500 L/m<sup>2</sup> reported by UNIDO (2017). However, the same source mentions that 95% of this water usage derives from rainwater for feed production (green water). It is unclear whether the authors consider the allocation factors described by the PEFCR for leather, which would mean that a massive portion of impacts, including water consumption potential, are allocated to milk and meat production.

Among the leather-like materials, the only water footprint confirmed by the literature is the PVC one (31.6  $L/m^2$ , which is in the range that Wool (2013) describes). The cactus LLM demands double the water volume than reported by Adriano di Marti (2022) (43.3  $L/m^2$  versus 20  $L/m^2$ ), and we found PU synthetic to have a much lower water usage than reported by Wool (2013) (42.6  $L/m^2$  in contrast to 268.05  $L/m^2$ ). The cotton content in the backing material could be responsible for the difference. As mentioned in the supplementary

material, we assumed a 100% polyester backing for materials made of PU, PVC, cactus, and bacterial cellulose. However, the application of a poly-cotton blend fabric could significantly increase the water footprint, considering the high use of this resource in cotton production. Pineapple fibers also present a slightly higher footprint than the one reported by Ananas Anam Ltd. (2021), i.e., 170.1 L/m<sup>2</sup> against 144 L/m<sup>2</sup>. Since this alternative does not use reinforcement material, we attribute the difference to the PLA from maize. Biopolymers coming from rainfed crops usually have a lower water intensity than the ones from irrigated farms (Hildebrandt et al., 2021), which could be the case in this model.

The LCA results for TAP and FEP for bovine leather and kombucha LLM closely relate to previously reported values (Bründl, 2022; Bruno et al., 2007; Notarnicola et al., 2011; Yu et al., 2021).

#### 4.2. Areas for improvement of current practices

Alternative water heating methods could be explored on an industrial scale. As discussed, energy drives the carbon emissions of most leather-like materials, which highlights the need for improvement in this aspect. Nonetheless, Laurenti et al. (2017) discuss how a tannery in Brazil decoupled their energy usage from carbon emissions by adopting biomass as an energy source, which could be applicable in this case. Similarly, UNIDO (2017) reports a 5 to 8.3 kgCO<sub>2-eq</sub>/m<sup>2</sup> reduction in carbon emissions per unit output with a solar water heating system in a tannery in Bangladesh. Another solution is the combined power and heat cogeneration, which yields hot water as a co-product (Maghanki et al., 2013).

What vegetable-tanning saves in chemical-related emissions due to the absence of chromium sulfates, it makes up for in electricity-related emissions, as additional agitation and drying processes are required (Baquero et al., 2021). Furthermore, the biobased leather-like materials exhibit comparable or higher carbon emissions than the fossil-based alternatives. This is understandable, considering that the former ones are still in the lab or pilot scale, while PU and PVC are industry staples with highly optimized processes. The main contrast between these two groups lies in electricity consumption, which becomes one of the primary areas of improvement when scaling up an emerging technology (Williams et al., 2022).

The degumming of pineapple leaf fibers is the main issue for PALF leather-like material production. In our model, we assumed an alkali degumming process based on the patent by Hijosa et al. (2013). However, this process is highly variable in the literature due to four crucial variables: the concentration of NaOH, the liquor ratio (i.e., the amount of NaOH solution per kilogram of pineapple leaves), the incubation time, and the incubation temperature. The concentration of NaOH can vary from 1% (Hazarika et al., 2017) to 4% (Chollakup et al., 2017), while the literature describes liquor ratios ranging from 10:1 (Hazarika et al., 2017)

to 100:1 (Munawar et al., 2008). The incubation time can range from 30 minutes (Hijosa et al., 2013) to 12 hours (Chiliveri et al., 2016), and the temperature can vary from 80°C (Li et al., 2011) to 100°C (Chollakup et al., 2017). These four variables determine the heat used in degumming, which is the main driver of emissions in this process.

The diminished carbon emissions of leather-like materials can be outweighed by their lower physical resistance. Properties like tear resistance directly impact the durability and lifespan of leather-like materials (Meyer et al., 2021). A longer lifespan like the one bovine leather presents could favor the material over the alternatives in circular design strategies like designing for physical endurance (den Hollander et al., 2017). However, a further study should encompass additional characteristics such as tensile strength and flex resistance, which are incredibly relevant for footwear (Maina et al., 2019). We should also account for peeling and crackling behavior, which are common concerns regarding synthetic materials.

The high plastic content is another problem to be tackled because it affects the post-life treatment of these materials, as most plasticizers cannot be recycled when contaminated with fabric or leather (Kemona & Piotrowska, 2020). Consequently, these materials are typically directed toward incineration, producing NOx, dioxins, and isocyanates (Fernandes, 2021). The low plastic content in materials like kombucha LLM can be a direct result of the production scale, suggesting that PU coating could become a reality in a mass-production scenario. In that case, the bacterial cellulose alternative would receive a yellow or red score in the LiSET matrix, depending on the concentration of the plasticizer.

Regarding the exposure to the chemical hazards, alternative modes of production could be proposed. For instance, dimethylformamide (DMF) is an integral part of polyurethane polymerization, despite its association with reproductive toxicity. However, the production DMF-free PU synthetic leather is already happening (Wang & Jin, 2018), which has the potential to change the categorization of this material in the LiSET matrix. We considered certain types of accident risks as preventable through the proper use of personal protective equipment (PPE), as disclosed in **Step 3: Grading system** and in the supplementary material. However, the effective prevention of these chemical hazards is not guaranteed, especially considering that many of the compared materials are produced in countries with lenient regulations concerning working conditions and chemical disposal (Garai, 2014). Additionally, risks not included in the MSDS should be studied, such as the potential for endocrine disruption caused by plasticizers present in PU and PVC materials (Li & Ko, 2012; Mathieu-Denoncourt et al., 2015).

#### 4.3. Limitations

Although streamlining the scope of an LCA has its advantages, such as reducing data requirements, it has shortcomings in other areas. One of the major drawbacks is the omission of downstream processes, which results in a less comprehensive outcome compared to the full version.

Data quality and availability are also hindrances. The LiSET matrix builds on sources that are not necessarily peer-reviewed, often including patents, and to a lesser extent, companies' reports. The LCA also suffers from this limitation, as we retrieved the life cycle inventories from secondary data, which has its own share of uncertainties. Although different assumptions could change the results of this study, the main conclusions remain unchanged, such as the trade-off between environmental impacts and durability, or the increased use of plastics in biobased materials.

The heat source used for water heating is one of the most influential assumptions here. Depending on the material, either industrial heat from natural gas or electricity is utilized to boil water. The supplementary material offers sufficient justification for the selection of each heat source, considering the scale of production. Another influential assumption is the backing material in PU synthetic leather, as previously discussed.

#### 4.4. Further work

Other qualitative and semi-quantitative lifecycle aspects could be incorporated into subsequent iterations of the LiSET matrix, e.g., 'the cost per square meter of material', given that biobased leather alternatives target high-end products, resulting in higher material costs; and 'the potential for microplastic generation', which is an impact that current LCA methodologies often overlook.

The streamlined comparative LCA could be improved by extending the system boundaries to shoe assembly and end-of-life stages, which would include impacts due to e.g., incineration (Gottfridsson & Zhang, 2015), implying high emissions of dioxins, NO, NH<sub>3</sub>, and HCN for shoes made of PU and PVC (Garrido et al., 2017). Different applications and leather sources could be explored in further studies; for example, as Hansen et al. (2021) show, there is a difference between chemical consumption for tanning leather for shoes, furniture, or clothes. A new LCA could explore the impacts of regionality and transport-related emissions, as well as the trade-offs related to land use.

#### 5. Conclusions

Selecting the best material for footwear is a complex task, as most options present trade-offs. The comparative LCA depicts bovine leather to have the worst environmental impact in all categories, except for cumulative energy demand. However, according to the LiSET matrix, no alternative material can match the tear resistance of genuine leather, which directly correlates with its durability. Therefore, the lower environmental footprint of alternatives to bovine leather comes at the expense of their inferior durability. As a result, more production would be required to achieve the same lifespan as genuine leather, thus negating the environmental benefits. Another point to consider is that most biobased LLMs contain high concentrations of plastics, which serves as a trade-off between lower carbon emissions and the potential to create value from waste. This raises the question of which impact holds more significance and which aspect we can focus on improving in the future.

The production of emerging materials, such as biobased alternatives, is currently suboptimal, resulting in high energy demands and significant environmental impacts. While comparing technologies at distinct levels of readiness may seem unfair to options still in laboratory or pilot scales, it is necessary to identify potential areas for improvement. For instance, in the case of the PALF-based material, it is crucial for researchers and manufacturers to focus on improving the degumming process, as it contributes the most to carbon emissions within the material's lifecycle. Furthermore, the LiSET matrix clearly highlights data gaps in the literature, particularly regarding acidification and eutrophication potential. These areas should be prioritized for future research endeavors. Even though biobased materials could be considered as worse than fossil synthetics in terms of emissions and energy consumption, they represent a significant step towards a generation of footwear materials that add value to waste while reducing the reliance on fossil polymers.

The hybrid assessment represents a straightforward approach to comparing technologies across a plethora of qualitative and quantitative criteria. While comparative LCA provides scientific rigor through standardized environmental assessment, the LiSET matrix embraces options with limited data availability and explores aspects not typically covered by a conventional LCA. We highly recommend this holistic assessment to compare the physical, environmental, and economic performance of competitive technologies. It enables the identification of hotspots and potential substitutes for industry staples.

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NTNU Department of Energy and Process Engineering

# Environmental performance of leather and leatherlike materials applied to footwear

SUPPLEMENTARY MATERIALS



Author: Pedro Patrique Ferreira da Silva Supervisor: Johan Berg Pettersen, Associate Professor, NTNU Industrial Ecology Program Co-supervisor: Kamila Krych, PhD Candidate, NTNU Industrial Ecology Program

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# Abbreviations

BCS = Basic chromium sulfates

- DMF = Dimethylformamide
- GLO = Global
- KBC = Kombucha bacterial cellulose
- LCA = Life Cycle Assessment
- LiSET = Lifecycle Screening of Emerging Technologies
- LLM = Leather-like material
- MSDS = Material safety data sheet
- PALF = Pineapple leaf fibers
- PEFCR = Product Environmental Footprint Category Rules
- PLA = Polylactic acid
- PPE = Personal protective equipment
- PU = Polyurethane
- PVC = Polyvinyl chloride
- RoW = Rest of the world
- SCOBY = Symbiotic community of bacteria and yeast

# 1. Lifecycle Screening of Emerging Technologies (LiSET)

**Table S1** - Coarse grading system for the LiSET of bovine leather and respective alternatives applied to footwear.

Exposure to chemical hazards (qualitative)			
The material uses chemicals from categories 1,2, 3 or 4 in the (EC) No 1272/2008, which the effects cannot			
be prevented by using PPEs alone.			
The material uses chemicals from categories 1, 2, 3 or 4 in the (EC) No 1272/2008, which the effects can be			
prevented by using PPEs like gloves, glasses, and masks.			
The material only uses chemicals that are classified as non-hazardous in the (EC) No 1272/2008.			
Water consumption (quantitative – optimized)			
The material consumes more than 268.05 L/m <sup>2</sup> of water.			
The material consumes more than 68.5 L/m <sup>2</sup> of water and less or equal to 268.05 L/m <sup>2</sup> of water.			
The material consumes 68.5 L/m <sup>2</sup> of water or less.			
Carbon emissions (quantitative – optimized)			
The material emits more than 15.25 kgCO <sub>2-eq</sub> /m <sup>2</sup> .			
The material emits more than 3.9 kgCO <sub>2-eq</sub> /m <sup>2</sup> and less or equal to 15.25 kgCO <sub>2-eq</sub> /m <sup>2</sup> .			
The material emits 3.9 kgCO <sub>2-eq</sub> /m <sup>2</sup> or less.			
Tear resistance (quantitative – optimized)			
The material has a tear resistance of 37.2 N/mm or lower.			
The material has a tear resistance is lower or equal to 52.6 N/mm and higher than 37.2 N/mm.			
The material has a tear resistance higher than 52.6 N/mm.			
Non-plastic content (qualitative)			
The material contains polymers like polyurethane and, therefore, it does not biodegrade.			
The material can biodegrade, with proper technology.			
The material can easily biodegrade.			
Value creation from by-products and waste (qualitative)			
The material is not known for creating value from agricultural by-products or waste.			
The material creates value to agricultural co-product or by-product.			
The material creates value to agricultural waste.			
Energy demand (quantitative – optimized)			
The material consumes more than 97.93 MJ/m <sup>2</sup> .			
The material consumes more than 36.2 MJ/m <sup>2</sup> and less or equal to 97.93 MJ/m <sup>2</sup> .			
The material consumes 36.2 MJ/m <sup>2</sup> or less.			

<b>Table S2</b> – Transfer coefficient	between live cattle weight to	finished leather	(in kilograms)
, , , , , , , , , , , , , , , , , , , ,	<u> </u>	2	

	Converts to	Efficiency (w/w)	Reference
Live weight	Raw hides	8%	Limeneh et al. (2022); Pearson & Dutson, (1992)
Wet salted hides	Finished leather	20%	Jiang et al. (2016); Cabeza et al. (1998); Notarnicola et al. (2011)
Raw hides	Wet-salted hides	100%	Buljan & Král (2019) ª
Wet-salted hides	Crust leather	Between 33 and 48%	Notarnicola et al. (2011); Yu et al. (2021)
Crust leather	Finished leather	between 42 and 61%	Author's calculations based on Notarnicola et al. (2011) and Yu et al. (2021)

<sup>a</sup>: As Buljan & Král (2019) explain, the difference in weight between raw hides and wet-salted hides is negligible. For (dry) salted hides, the weight lost due to dehydration of the hide is compensated by the weight of salt, which results in the same weight as raw hides.

Table S3 – Conversion factor between area and weight for bovine leather and different leather-like materials

Material	Area (in m²)	Weight (in kg)	Reference
Finished bovine leather	1	1.04	Based on the PEFCR (Rosa-Giglio et al., 2018)
Polyurethane/Polyvinylchloride	1	0.326	Sphera Solutions GmbH (2020)
leather-like material			
Pineapple leaf fibers leather-like	1	0.404	Hildebrandt et al. (2021)
material			
Cactus leather-like material	1	0.595	Based on Alternative Leather Co. (2023) <sup>b</sup>
Kombucha bacterial cellulose leather-	1	0.250	Author's calculation
like material			
Mycelium leather-like material	1	0.620	Based on Kaplan & Bei (2018)

<sup>a</sup>: As per the PEFCR, the production of 1 m<sup>2</sup> of bovine finished leather for footwear requires 7.41 kg of raw hides. However, this research uses 5.74 kg of raw hide per 1 m<sup>2</sup> of finished leather, which is specifically meant for calves' skin applied to footwear. Due to the lack of information regarding the thickness of the product considered in the PEFCR, it was determined that the second value more accurately reflects reality. Using the ratios described above, finished leather has 1.04 kg/m<sup>2</sup>, which is a value supported by Notarnicola et al. (2011) and Grasso et al. (1990) in which it is stated that 200 kg of finished leather for footwear is equivalent to 185.8 m<sup>2</sup> of 1.3 mm thickness (1.08 kg/m<sup>2</sup>). Furthermore, industry professionals reaffirm the values described here.

**b**: Alternative Leather Co. (2023) states that the weight for cactus leather ranges from 540 g/m<sup>2</sup> to 650 g/m<sup>2</sup>. The average value, i.e., 595 g/m<sup>2</sup>, is adopted.

•: Kaplan & Bei (2018) show that post-processed mycelium leather-like materials have a density between 240 kg/m<sup>3</sup> and 800 kg/m<sup>3</sup>, with an average of 520 kg/m<sup>3</sup>. This means that a 1.2 mm thick mycelium sheet (1 m x 1 m x 0.0012 m) weights around 0.62 kg.

In line with the principle of transparency exposed by Hung et al. (2020) in the Lifecycle Screening of Emerging Technologies (LiSET) framework, a detailed list of all the sources employed in the LiSET analysis is presented below. In cases where divergent values were encountered, the average between the values was computed and utilized instead. This approach ensures a reasonable estimation while accounting for variations in reported data.

#### 1.1. Chromium-tanned bovine leather

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Sodium dichromate (mutagenic)	Red	MSDS
Water consumption (L/m <sup>2</sup> )	16,335 (farming) and between 136 and 180 (manufacturing)	16,335 + 160.3 = 16,495.3	Laurenti et al. (2017); UNIDO (2017) <sup>a</sup>
Carbon emissions (kgCO <sub>2-eq</sub> /m <sup>2</sup> )	105.6 (farming) and between 2.5 and 12 (manufacturing)	105.6 + 6.3 = 111.9	Laurenti et al. (2017); UNIDO (2017) <sup>b</sup>
Tear resistance (N/mm)	82.9	82.9	Meyer et al. (2021)
Non-plastic content	Acrylic resins coating is common in leather finishing.	Yellow	Bhattacharjee et al. (2014); Notarnicola et al. (2011)
Value creation from by-products and waste	Bovine hides are a co-product of the meat industry.	Yellow	Rosa-Giglio et al. (2018)
Energy demand (MJ/m <sup>2</sup> )	90 (farming) and between 30 and 49 (manufacturing)	90 + 41.37 = 131.37	Canals et al. (2002); Laurenti et al. (2017); Yu et al. (2021) <sup>3</sup>

 Table S4 - Lifecycle aspects for chromium-tanned bovine leather applied to footwear.

<sup>a</sup> Original measurements: The farming calculation comes from UNIDO (2017), in which it is stated that 16,500 L of water per square meter are used from cradle-to-gate, but only 1% (165 L/m<sup>2</sup>) is allocated to manufacturing. Laurenti et al. (2017) report between 136 L/m<sup>2</sup> and 180 L/m<sup>2</sup> from raw hide to finished leather. The average value (160.3 L/m<sup>2</sup>) is used.

<sup>b</sup> Original measurements: In line with the previous calculation, UNIDO (2017) also reports a carbon footprint (cradle-to-gate) of 110 kgCO<sub>2-eq</sub>/m<sup>2</sup>, from which 4% (4.4 kgCO<sub>2-eq</sub>/m<sup>2</sup>) is allocated to the manufacturing phase. This value aligns with the ones reported by Laurenti et al. (2017) which range from 2.5 to 12 kgCO<sub>2-eq</sub>/m<sup>2</sup> (raw hide to finished leather). The average value (6.3 kgCO<sub>2-eq</sub>/m<sup>2</sup>) is used.

<sup>c</sup> Original measurements: Canals et al. (2002) describe the energy footprint (cradle-to-gate) for producing 1000 kg of finished leather. The electricity values for agriculture, farming, and slaughtering are converted in megajoules and then summed to the values of thermal energy. They are normalized for 1 m<sup>2</sup>, using the conversion rates explained in **Table S3**. Yu et al. (2021) list 8,592 MJ of energy (thermal + electricity) as input for 480 kg of crust leather as output (around 200 kg of finishes leather, or 190.5 m<sup>2</sup>. Thus, 1 m<sup>2</sup> requires around 45.1 MJ/m<sup>2</sup>). This is in the range portrayed in Laurenti et al. (2017) that goes from 30 MJ/m<sup>2</sup> to 49 MJ/m<sup>2</sup> (from raw hides to finished leather). The average value (41.37 MJ/m<sup>2</sup>) is used.

#### 1.2. Vegetable-tanned bovine leather

Table S5 - Lifecycle aspects for vegetable-tanned bovine leather applied to footwear.

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Glutaraldehyde, formic acid (skin corrosion)	Yellow	MSDS
Water consumption (L/m²)	16.335 (farming) and between 100 and 214 (manufacturing)	16.335 + 157 = 16,492	Laurenti et al. (2017); UNIDO (2017) a
Carbon emissions (kgCO <sub>2-eq</sub> /m²)	105.6 (farming) and between 2.4 and 5 (manufacturing)	105.6 + 3.7 = 109.3	Laurenti et al. (2017); UNIDO (2017) b
Tear resistance (N/mm)	44.9	44.9	Musa & Gasmelseed (2013)
Non-plastic content	Acrylic resins coating is common in leather finishing.	Yellow	Bhattacharjee et al. (2014); Notarnicola et al. (2011)
Value creation from by-products and waste	Bovine hides are a co-product of the meat industry.	Yellow	Rosa-Giglio et al. (2018)
Energy demand (MJ/m²)	90 (farming) and between 15.4 and 57 (manufacturing)	90 + 36.2 = 126.2	Canals et al. (2002); Laurenti et al. (2017) °

<sup>a</sup> Original measurement: The farming water consumption was calculated as in **Table S4**. As for the manufacturing part, Baquero et al. (2021) stipulate an input of 9,300 L of water for 1000 kg of pickled hides (which will be turned in to 200 kg of finished leather, or 190.5 m<sup>2</sup>). This results in a footprint of 48.8 L/m<sup>2</sup>. This is quite below the two values presented in Laurenti et al. (2017), one of 100 L/m<sup>2</sup> and one of 214 L/m<sup>2</sup> (raw hide to finished leather). Since Baquero et al. (2021) only consider the post-tanning processes, the values are not included in this scope.

<sup>b</sup> Original measurements: The farming carbon emissions was assigned as explained in **Table S4**. The manufacturing carbon footprint is extracted from Laurenti et al. (2017), in which two values are reported, one of 2.4 kgCO<sub>2-eq</sub>/m<sup>2</sup> and one of 5 kgCO<sub>2-eq</sub>/m<sup>2</sup> (raw hides to finished leather).

<sup>c</sup> Original measurements: The farming and slaughtering energy demand followed the same rationale illustrated in **Table S4**. As for the manufacturing, Laurenti et al. (2017) showcase a value of 27 MJ/m<sup>2</sup> and one of 57 MJ/m<sup>2</sup> (raw hides to finished leather). Once again, data from Baquero et al. (2021) was not included due to differences in system boundary.

## 1.3. Polyurethane (PU) waterborne leather-like material

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Dimethylformamide (reproductive toxicity)	Red	MSDS
Water consumption (L/m <sup>2</sup> )	Between 134.2 and 401.9	268.05	Wool (2013) <sup>a</sup>
Carbon emissions (kgCO <sub>2-eq</sub> /m²)	Between 3.09 and 4.71	3.9	Bründl (2022); Schäfer et al. (2021); Wool (2013) <sup>b</sup>
Tear resistance (N/mm)	Between 17 and 79.2	48.1	Meyer et al. (2021); Saha et al. (2020)
Non-plastic content	PU is a fossil-based plastic polymer	Red	Lithner et al. (2011)
Value creation from by-products and waste	PU LLM is produced exclusively with virgin materials of fossil origin, not creating value from agricultural waste or by-products.	Red	Locker & Theregowda (2022); Schäfer et al. (2021); Wool (2013); Xia et al. (2010)
Energy demand (MJ/m <sup>2</sup> )	Between 92.76 and 102.5	97.93	Wool (2013) °

Table S6 - Lifecycle aspects for polyurethane (PU) waterborne leather-like material applied to footwear.

<sup>a</sup> Original measurement: Wool (2013) defines a water footprint between 20.1 gal/lb. and 60.2 gal/lb., or from 167.74 L/kg to 502.39 L/kg. The 0.326 kg/m<sup>2</sup> rate described in **Table S3Table S5Error! Reference source not found.** is not considered for data from Wool (2013), but instead, 0.8 kg/m<sup>2</sup>. Although no information is provided regarding the conversion of mass into area, it is assumed that this patent uses a heavier or denser material, so the results can support data found in other sources, like Schäfer et al. (2021). This results in a water consumption between 134.2 L/m<sup>2</sup> and 401.9 L/m<sup>2</sup>.

<sup>b</sup>Original measurements: Schäfer et al. (2021) model synthetic leather and obtain a carbon footprint between 3.09 kgCO<sub>2-eq</sub>/m<sup>2</sup> and 4.71 kgCO<sub>2-eq</sub>/m<sup>2</sup>, the last being the value used in Bründl (2022). As argued by the authors, the variation is due to the different fossil-based polymers as well as the reinforcement fabric. As for Wool (2013), the patent reports emissions between 5,04 lb.CO<sub>2-eq</sub>/lb. and 5.60 lb.CO<sub>2-eq</sub>/lb. (from 5.04 kgCO<sub>2-eq</sub>/kg to 5.60 kgCO<sub>2-eq</sub>/kg). With the conversion rate of 0.8 kg per square meter, a carbon footprint between 4 kgCO<sub>2-eq</sub>/m<sup>2</sup> and 4.4 kgCO<sub>2-eq</sub>/m<sup>2</sup> is obtained. <sup>c</sup>Original measurements: Wool (2013) elaborates that the energy demand for PU synthetic ranges between 12,571 kcal/lb. and 13,892 kcal/lb., or between 115.96 MJ/kg and 128.15 MJ/kg. With the 0.8 kg/m<sup>2</sup> conversion rate, this results in an energy demand ranging from 92.76 MJ/m<sup>2</sup> and 102.5 MJ/m<sup>2</sup>.

# 1.4. Polyvinylchloride (PVC) leather-like material

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Butyl benzyl phthalate (reproductive toxicity)	Red	MSDS
Water consumption (L/m <sup>2</sup> )	Between 15.09 and 45.33	30.21	Wool (2013) <sup>a</sup>
Carbon emissions (kgCO <sub>2-eq</sub> /m²)	Between 2.72 and 3.14	2.93	Schäfer et al. (2021); Wool (2013) <sup>b</sup>
Tear resistance (N/mm)	Between 14 and 81	47.5	۹ Altindag & Akdogan (2021)
Non-plastic content	PVC is a fossil-based plastic polymer.	Red	Lithner et al. (2011)
Value creation from by-products and waste	PVC LLM is produced exclusively with virgin materials of fossil origin, not creating value from agricultural waste or by-products.	Red	Baitz et al. (2004); Schäfer et al. (2021)
Energy demand (MJ/m <sup>2</sup> )	Between 66.38 and 89.18	77.78	Wool (2013) <sup>d</sup>

 Table S7 - Lifecycle aspects for polyvinylchloride (PVC) leather-like material applied to footwear.

<sup>a</sup> Original measurements: Wool (2013) elaborates that the water consumed in the PVC manufacturing is between 2.26 gal/lb. and 6.79 gal/lb., or 18.86 L/kg and 56.67 L/kg, respectively. Assuming a conversion of 0.8 kg per square meter, this equals to 15.09 L/m<sup>2</sup> as the minimum and 45.33 L/m<sup>2</sup> as the maximum.

<sup>b</sup> Original measurements: Wool (2013) defines a carbon footprint for PVC that goes from 3.4 lb.CO<sub>2-eq</sub>/lb. and 3.92 lb.CO<sub>2-eq</sub>/lb. (or 3.4 kgCO<sub>2-eq</sub>/kg and 3.92 kgCO<sub>2-eq</sub>/kg, respectively). With the 0.8 kg/m<sup>2</sup> rate, this translates into 2.72 kgCO<sub>2-eq</sub>/m<sup>2</sup> and 3.14 kgCO<sub>2-eq</sub>/m<sup>2</sup>, which agree with the values stipulated by Schäfer et al. (2021).

<sup>c</sup> Observation: Altindag & Akdogan (2021) report values of tear strength of 17±3 N/mm, 40±11 N/mm, and 76±5 N/mm. As discussed by the authors, the differences come from the type of plasticizes and its concertation, or level. For this framework, the lowest and highest numbers are adopted, using an average between these values.

<sup>d</sup> Original measurements: An energy consumption between 8,995 kcal/lb. (82.97 MJ/kg) and 12,571 kcal/lb. (111.47 MJ/kg) is conveyed in Wool (2013). Using the 0.8 kg/m<sup>2</sup> rate gives us an energy footprint between 66.38 MJ/m<sup>2</sup> and 89.18 MJ/m<sup>2</sup>.

# 1.5. Pineapple leaf fibers (PALF) leather-like material

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Sodium hydroxide (skin corrosion)	Yellow	MSDS
Water consumption (L/m <sup>2</sup> )	Between 144 and 14,400	144	Ananas Anam Ltd. (2021); Hildebrandt et al. (2021) ª
Carbon emissions (kgCO <sub>2-eq</sub> /m²)	2.69	2.69	Ananas Anam Ltd. (2021) <sup>b</sup>
Tear resistance (N/mm)	31	31	Meyer et al. (2021)
Non-plastic content	20% wt. is PLA, 5% by bio-based PU, and 5% by fossil-based PU	Red	Ananas Anam Ltd. (2021); Hijosa et al. (2013) °
Value creation from by-products and waste	The material uses the fibers of pineapple leaves, which is a waste from the fruit production.	Green	Padzil et al. (2020); Pandit et al. (2020); Siakeng et al. (2020)
Energy demand (MJ/m <sup>2</sup> )	107.46	107.46	Hildebrandt et al. (2021) <sup>d</sup>

Table S8 -Lifecycle aspects for pineapple leaf fibers (PALF) leather-like material applied to footwear.

<sup>a</sup> Original measurements: Hildebrandt et al. (2021) discuss that the water used during pineapple cultivation should be allocated to the fruit production process. However, it is crucial to consider the water use in polylactic acid (PLA) production, which in the case of Piñatex<sup>®</sup> means considering irrigation in corn production (the main raw material for PLA). The paper gives a range between 0.144 and 1.44 m<sup>3</sup> of water per square meter of PALF leather-like material, if the corn is rainfed or irrigated, respectively. Corn production in the Philippines is majorly rainfed (Anuada et al., 2022), which means that the value of 0.144 m<sup>3</sup>/m<sup>2</sup> or 144 L/m<sup>2</sup> should be adopted, assuming a local production of PLA. The value is the same described by Ananas Anam Ltd. (2021) in their LCA study.

<sup>c</sup> Original measurements: Ananas Anam Ltd.(2021) states that the carbon footprint of Piñatex<sup>®</sup> in 2020 was 2.69 kgCO2-eq/m<sup>2</sup>. Although this is considered as grey literature, the only peer-reviewed paper that measures the environmental performance of PALF leather-like material (Hildebrandt et al., 2021) provides an aggregated result that makes it impossible to isolate the carbon footprint. Thus, the values in the report by Ananas Anam Ltd. was considered as the best estimation for this lifecycle aspect.

<sup>d</sup> **Observation:** PALF 'leather' uses polyurethane (10% wt.) for achieving properties like water resistance and durability (Ananas Anam Ltd., 2021). Even though half of the polyurethane comes from biological sources (from sugarcane), the other half is still from fossil origin. Furthermore, around 20% (wt.) is composed by PLA, which is a biodegradable plastic polymer that is not free from environmental burdens and can create microplastics, as shown in Tong et al. (2022).

• Original measurements: Hildebrandt et al. (2021) show that 1 m<sup>2</sup> PALF leather-like material demands 1.77 kWh of energy, 0.58 kWh for PLA production and it has a cumulative energy demand (CED) of 27.5 kWh/m<sup>2</sup>. The sum converts to 107.46 MJ/m<sup>2</sup>.

#### 1.6. Cactus leather-like material

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Sodium hypochlorite (Strong irritating to mucous membranes)	Yellow	MSDS
Water consumption (L/m <sup>2</sup> )	7% of PU's water footprint	18.8	Adriano di Marti (2022) ª
Carbon emissions (kgCO <sub>2-eq</sub> /m <sup>2</sup> )	29% of PU's carbon footprint	1.1	Adriano di Marti (2022) ª
Tear resistance (N/mm)	37.2	37.2	Meyer et al. (2021)
Non-plastic content	Cactus LLM contain up to 80% (wt.) in plastic polymer like PU or PVC.	Red	Cázarez Duarte & Lopez Arriaga Lopez Velarde (2021) <sup>b</sup>
Value creation from by-products and waste	Cactus is primarily produced for manufacturing leather-like material.	Red	Adriano di Marti (2022)
Energy demand (MJ/m <sup>2</sup> )	37% of PU's energy footprint	36.23	Adriano di Marti (2022) ª

 Table S9 - Lifecycle aspects for cactus leather-like material applied to footwear.

<sup>a</sup> Original measurement: Adriano di Marti (2022) provides results of the environmental performance analysis for water, carbon, and energy of Desserto<sup>®</sup>, a commercially available cactus LLM, and compares it with polyurethane and bovine leather. Although this study is considered as grey literature and cannot be replicated without additional information, its results were used to calculate percentages, that are applied to the data presented in **Table S6**. The calculated values are found to be closely aligned with the reported in the original document, with the estimated water consumption being 18.8 L/m<sup>2</sup> (calculated) compared to 20 L/m<sup>2</sup> (original), carbon emissions being 1.1 kgCO<sub>2-eq</sub>/m<sup>2</sup> (calculated) as opposed to 1.39 kgCO<sub>2-eq</sub>/m<sup>2</sup> (original), and energy consumption being 36.23 MJ/m<sup>2</sup> (calculated) as compared to 34.33 MJ/m<sup>2</sup> (original).

<sup>b</sup> Observation: Cázarez Duarte & Lopez Arriaga Lopez Velarde (2021) show that liquid polyurethane is used in the manufacturing process of cactus leather-like material which increases the potential of microplastic generation. Alternative Leather Co. (2023) shows that up to 80% of the final weight of cactus-based leather-like material is polyurethane.

## 1.7. Kombucha bacterial cellulose (KBC) leather-like material

Table S10 - Lifecycle aspects for Kombucha bacterial cellulose (KBC) leather-like material applied to footwear.

Lifecycle aspect	Literature values	Value used	Reference
Exposure to chemical hazards (based on the MSDS)	Sodium hydroxide (preventable skin corrosion)	Yellow	MSDS
Water consumption (L/m <sup>2</sup> )	Between 60 and 77 + 200 L (finishing)	268.5	Fernandes et al. (2019); Hildebrandt et al., (2021); Zhao et al. (2018) ª
Carbon emissions (kgCO2-eq/m <sup>2</sup> )	12.27	12.27	Bründl (2022) <sup>b</sup>
Tear resistance (N/mm)	25.44	25.44	Nguyen et al. (2021)
Non-plastic content	Many studies use natural coating with beeswax or vegetable oils.	Green	da Silva Junior et al. (2022); Damsin (2019)
Value creation from by-products and waste	Bacterial cellulose is a by-product from the food industry (kombucha production), and it is often taken as waste.	Green	Rathinamoorthy & Kiruba (2020); Tang et al. (2021)
Energy demand (MJ/m <sup>2</sup> )	Between 6.84 and 217.99	112.42	Bründl (2022); Hildebrandt et al. (2021) •

<sup>a</sup> Original measurements: Fernandes et al. (2019) show that a dry sheet of  $1.56 \times 10^{-4} \text{ m}^3$  (in volume) represents 11 grams of dry bacterial cellulose. By adopting the yield of 1.18 g/L of broth described by Zhao et al. (2018), one finds that 1 L of fermentation broth yields in a material sheet with  $1.67 \times 10^{-5} \text{ m}^3$ . A 1.2 mm thick sheet (1 m x 1 m x 0.0012 m) requires 84.62 g of bacterial cellulose which demands 71,71 L of fermentation broth. The water demand in accordance with Hildebrandt et al. (2021), who state that one 1 m<sup>2</sup> of KBC leather-like material demands 60 L of water for fermentation. Bründl (2022) reports a value of 77 L/m<sup>2</sup>, which is close of the values described in the other sources. Hildebrandt et al. (2021) also report 200 L of water being used for finishing.

<sup>b</sup> Observation: This is the correspondent value of Celium<sup>™</sup>, a commercial bacterial cellulose material.

<sup>c</sup> Original measurements: Bründl (2022) show that minimal electricity demand for Celium<sup>™</sup> is 1.9 kWh/m<sup>2</sup> (6.84 MJ/m<sup>2</sup>). Hildebrandt et al. (2021) state that bacterial material has an energetic demand of 26 kWh (93.6 MJ) per liter of fermentation. Considering 68.5 L, the energy demand is around 6,411.6 MJ. As Mohammadshirazi & Bagheri Kalhor (2016) show, only 3.4% of the energetic demand is allocated to microorganisms, which translates to 217.99 MJ/m<sup>2</sup>.

#### 1.8. Mycelium leather-like material

 Table S11 - Lifecycle aspects for mycelium leather-like material applied to footwear.

Lifecycle aspect	Literature values	Value used	Reference
· ·			
Exposure to chemical hazards (based on the MSDS)	Genipin adipic or phenol (germ cell mutagenicity)	Red	MSDS
Water consumption (L/m <sup>2</sup> )	Between 0.76 and 1.14	0.95	Silverman (2018) ª
Carbon emissions (kgCO <sub>2-eq</sub> /m²)	Between 6.2 and 24.3	15.25	Bründl (2022); Williams et al. (2022) <sup>b</sup>
Tear resistance (N/mm)	52.6	52.6	Jones et al. (2021)
Non-plastic content	A low-in-plastic material is possible, although most alternatives do contain significant amounts of fossil-based polymers.	Yellow	Jones et al. (2021)
Value creation from by-products and waste	Mycelium is produced with agro-industrial waste, like sawdust.	Green	Elsacker et al. (2020); Lelivelt et al. (2015)
Energy demand (MJ/m <sup>2</sup> )	Between 22.62 and 136.3	79.46	Kaplan & Bei (2018); Silverman (2018); Williams et al. (2022) <sup>c</sup>

<sup>a</sup> Original measurements: Silverman (2018) reports a water consumption between 1.22 L/kg and 1.83 L/kg, which translates to 0.76 L/m<sup>2</sup> and 1.14 L/m<sup>2</sup>, if the weight of 0.62 kg/m<sup>2</sup> from Table S3 is assumed.

<sup>b</sup> **Original measurements:** While Bründl (2022) obtains a carbon footprint of 24.3 kgCO<sub>2-eq</sub>/m<sup>2</sup>, Williams et al. (2022) show that MycoWorks' Reishi<sup>m</sup>, a mycelium composite leather alternative, has a carbon footprint of 6.2 kgCO<sub>2-eq</sub>/m<sup>2</sup> in the current pilot scale. This gap can be explained by the type of fungal growth, as Reishi<sup>m</sup> has a passive growth process with no consume of CO<sub>2</sub> to prevent the growth of the fungi reproductive bodies.

<sup>c</sup> Original measurements: Silverman (2018) stipulate that mycelium composites have an embodied energy use of 38.1 MJ/kg. If 1 m<sup>2</sup> of material with 1.2 mm thickness weights 0.62, as explained before, the mycelium leather-like material has an energy demand of 23.62 MJ/m<sup>2</sup>. However,

Williams et al. (2022) shows that in the current scale, 27 kWh (94.5 MJ) of electricity is spent per mycelium sheet with 0.43 kg. Assuming similar density as the previous cases, a 0.62 kg sheet (correspondent to  $1 \text{ m}^2$ ) would require 136.3 MJ.

# 1.9. Sensitivity analysis



Figure S1 - Sensitivity analysis of the LiSET matrix for water consumption (in L/m<sup>2</sup>)



Figure S2 - Sensitivity analysis of the LiSET matrix for carbon emissions (in  $kgCO_{2-eq}/m^2$ )



Figure S3 - Sensitivity analysis of the LiSET matrix for tear resistance (in N/mm)



Figure S4 - Sensitivity analysis of the LiSET matrix for energy demand (in MJ/m<sup>2</sup>)

#### 2. Life Cycle Assessment

The following section presents the data we used in a streamlined comparative life cycle assessment for seven technologies, including bovine leather and leather-like materials. The model incorporates certain general assumptions concerning heating, transportation, regionalization, and downstream processes.

#### Heating

All the systems being compared utilize heating, primarily for water heating purposes. We assumed that all systems use industrial heating from natural gas. The only exception is the material derived from kombucha bacterial cellulose (KBC), which utilizes water heating from electrical devices. This assumption is deemed accurate due to the current level of development of this technology, which is primarily at the laboratory scale. In all cases, an 80% efficiency (20% heat loss) is assumed, unless stated otherwise.

#### Transportation

We did not include transportation as an individual process in each system. This exclusion was due to poor data availability regarding the distances to be adopted. Instead, the model utilizes the market mix activities from *ecoinvent*, which already provide an average estimation of transport used per product. Consequently, adding an extra process to account for transportation could potentially result in double counting.

#### Regionalization

Regionalization significantly influences the performance of materials. In the case of bovine leather, both chromium tanning and vegetable tanning predominantly occur in Spain, which is one of the leading global producers of bovine leather (Notarnicola et al., 2011). For cactus and pineapple leaf fiber leather-like materials, we considered the place of feedstock production, leading to manufacturing in Mexico and the Philippines, respectively. These countries are also home to Adriano Di Marti<sup>™</sup> and Ananas Anam<sup>™</sup>, the two major companies producing such materials. In contrast, the production of polyurethane, polyvinylchloride, and bacterial cellulose-based materials is not restricted to specific locations, so the global market mix is adopted for these alternatives.

#### Downstream processes

As discussed by Hung et al. (2020), excluding downstream processes is one approach to streamline an LCA. Due to data availability concerning direct emissions, wastewater and solid residues treatments are not included in the scope of this research. It is considered that the downstream impacts, as well as direct emissions, do not cause significant alterations in the overall results, except for chromium-tanned bovine leather. This is attributed to the chrome (III) content present in the solid waste, as demonstrated in the studies by Buljan & Král (2019) and Yu et al. (2021). This is the only direct emission considered in the LCA.

# 2.1. Bovine leather

Figure S5 shows a general representation of bovine leather production (cradle-to-gate). The process starts with farming, both for milk and meat. It is very common that after some lactation cycles, the dairy cows are sent for slaughtering (Moreira et al., 2021). In slaughtering, the live weight is converted in beef, offal, and non-edible products. Hides account for between 6% and 8% of the live weight, as demonstrated in Table S2.

Then the valuable skins are sent to preservation with a salt brine, being ready for the preparation steps in the beamhouse. This includes removing hair, fat layers, vestige meat, trimming, and treating the hide with lime, which yields a pelt ready to be tanned (Buljan & Král, 2019). Up to this point, the treatment for both chromium-tanned and vegetable-tanned leather is the same. **Table S12** presents a complete inventory with the inputs and outputs for these processes.



*Figure S5* - General bovine leather production system. Processes with allocation are market with an 'A.' The allocation factors (percentage) are highlighted in green.

	Process	Activity	Value	Unit	Reference
	Slaughtering	Live cattle for slaughtering <sup>d</sup>	65.63	kg	Notarnicola et al. (2011) ª
		Water	126.8	kg	Notarnicola et al. (2011) <sup>a</sup>
		Fuel oil	8.55	kg	Notarnicola et al. (2011) ª
		Diesel	4.27	MJ	Notarnicola et al. (2011) <b>a</b>
		Heat (natural gas)	4.27	MJ	Notarnicola et al. (2011) <sup>a</sup>
		Electricity	0.08	kWh	Notarnicola et al. (2011) <sup>a</sup>
	Preservation -	Sodium chloride (12% w/w)	0.63	kg	Buljan & Král (2019) Þ
	Brine salt	Water (42% w/w)	2.2	kg	Buljan & Král (2019) <sup>b</sup>
		<i>p</i> -dichlorobenzene (0,3% w/w - biocide)	0.016	kg	Buljan & Král (2019) <sup>b</sup>
		Electricity	0.08	kWh	Author's calculation
	Washing	Water (100% w/w)	5.25	kg	Buljan & Král (2019) <sup>b</sup>
		Sodium hydroxide (1.1% w/w)	0.058	kg	Buljan & Král (2019) <sup>b</sup>
	Dirty soaking	Water (250% w/w)	13.12	kg	Buljan & Král (2019) <sup>b</sup>
<u></u>	Soaking	Water (150 % w/w)	7.88	kg	Buljan & Král (2019) <sup>b</sup>
INPL		Sodium hydroxide (1.1% w/w – alkali)	0.009	kg	Buljan & Král (2019) <sup>b</sup>
		<i>p-</i> dichlorobenzene (0,3% w/w - biocide)	0.008	kg	Buljan & Král (2019) <sup>b</sup>
	Immunization	Water (80% w/w – 28°C)	4.2	kg	Buljan & Král (2019) <sup>b</sup>
		Lime (1.5% w/w)	0.079	kg	Buljan & Král (2019) <sup>b</sup>
		Heat (natural gas)	0.11	MJ	Author's calculation
		Electricity (low power agitation)	0.2	kWh	Author's calculation
	Unhairing	Sodium sulfide (1.5% w/w)	0.074	kg	Buljan & Král (2019) <sup>b</sup>
	Reliming	Lime (0.9% w/w)	0.05	kg	Buljan & Král (2019) <sup>b</sup>
		Sodium sulfide (0.2% w/w)	0.01	kg	Buljan & Král (2019) <sup>b</sup>
		Water (75% w/w)	3.94	kg	Buljan & Král (2019) <sup>b</sup>
	Washing	Water (300% w/w)	15.75	kg	Buljan & Král (2019) <sup>b</sup>
	Splitting	Electricity (splitting machine)	0.2	kWh	Buljan & Král (2019)
	Deliming	Ammonium sulphate (2.5% w/w)	0.131	kg	Buljan & Král (2019) <sup>b</sup>
	Bating	Enzymes (1.5% w/w)	0.079	kg	Buljan & Král (2019) <sup>b</sup>
оитрит	All processes	Pelt ready for tanning	3.39	kg	Based on Notarnicola et al. (2011) °

**Table S12** - Life cycle inventory for cattle slaughtering, hides preservation, and hides preparation (at beamhouse) steps, normalized for  $1 m^2$  of finished leather.

<sup>a</sup>: The values from the original source were normalized for 65.6 kg of live weight, which is the amount required for 1 m<sup>2</sup> of finished leather.

**b**: The percentage values relate to the weight of wet salted hides.

: Notarnicola et al. (2011) report a weight loss of 35.5% (w/w) from salted hides to pelts.

d: The activity "Live cattle for slaughtering (GLO)" from *ecoinvent* already account for resources demanded for cattle farming, including feeding and breeding.

### 2.1.1. Chromium tanning

**Figure S6** illustrates the tanning process (in shades of green) for chromium-tanned leather. The figure is based on information and diagrams from Brugnoli (2012), Buljan & Král (2019), Laurenti et al. (2017), Notarnicola et al. (2011), UNIDO (2017), and Yu et al. (2021).



Figure S6 – Manufacturing process system for 1 m<sup>2</sup> of chromium-tanned leather

After the processes conducted in the beamhouse, the pelt undergoes pickling in a solution containing sodium chloride, followed by tanning with basic chromium sulfate (BCS). Since *econinvent* does not provide an activity for BCS, the chemical synthesis from sodium dichromate is included within the scope, following the system description by Yu et al. (2021). The inventory for this stage is displayed in **Table S13**. At this point, the chromium-tanned hide is referred to as wet-blue and is considered a commodity that can be commercialized (Buljan & Král, 2019).

The post-tanning steps include rewetting, retanning, and fatliquoring, among others. These steps are depicted in Yu et al. (2021) for which an inventory is available. The post-tanning sub-system yields crust leather, which can also be commercialized like wet-blue (Buljan & Král, 2019). The product system concludes with the finishing process, which aims to condition, buffer, and coat the leather. As this process is not included in Yu et al. (2021), data from Notarnicola et al. (2011) is considered, even though the latter is an older source. **Table S14** presents the complete inventory for chromium-based tanning, while **Table S16** shows the proxies used in the model.

	Process	Activity	Value	Unit	Reference
	Chrome	Sodium dichromate	500	kg	Yu et al. (2021)
	production	Glucose	150	kg	Yu et al. (2021)
5		Sulfuric acid	450	kg	Yu et al. (2021)
NP N		Water (85°C)	1000	kg	Yu et al. (2021)
		Electricity	203.9	kWh	Yu et al. (2021)
		Heat (natural gas - 4h)	5292	MJ	Yu et al. (2021)
Ουτρυτ	Chrome powder production	Chrome powder	1000	kg	Yu et al. (2021)

 Table S13 - Life cycle inventory for chrome powder production.

**Table S14** - Life cycle inventory for tanning  $1 m^2$  of bovine leather with chromium.

	Process	Activity	Value	Unit	Reference
	Pickling	Pelt ready for tanning	3.39	kg	Table S12
		Water (25°C)	10.50	kg	Yu et al. (2021) ª
NPUT		Sodium chloride	0.74	kg	Yu et al. (2021) <b>a</b>
_	Tanning	Water (25°C, 40°C)	21.00	kg	Yu et al. (2021) <b>a</b>
		Chrome powder	0.74	kg	Table S13

		Sodium formate	0.11	kg	Yu et al. (2021) <sup>a</sup>
		Sodium bicarbonate	0.12	kg	Yu et al. (2021) ª
	Rewetting	Water (35°C)		kg	Yu et al. (2021) <sup>a</sup>
	Retanning	Water (35°)	25.20	kg	Yu et al. (2021) <sup>a</sup>
		Sodium formate	0.04	kg	Yu et al. (2021) ª
		Sodium bicarbonate	0.05	kg	Yu et al. (2021) <sup>a</sup>
		Water (35°)	4.20	kg	Yu et al. (2021) <sup>a</sup>
		Acrylic resin	0.13	kg	Yu et al. (2021) ª
		Dicyandiamide resin	0.08	kg	Yu et al. (2021) ª
		Mimosa extract	0.17	kg	Yu et al. (2021) ª
		Dyestuff	0.08	kg	Yu et al. (2021) ª
	Fatliquoring	Water (50°C, 25°)	14.70	kg	Yu et al. (2021) <sup>a</sup>
		Fatliquor	0.59	kg	Yu et al. (2021) <sup>a</sup>
	All processes in the tanyard	Heat (natural gas)	4.80	MJ	Yu et al. (2021) <sup>a</sup>
		Electricity (machinery operation)	2.41	kWh	Yu et al. (2021) <sup>a</sup>
	Finishing	Aniline	0.01	kg	Notarnicola et al. (2011) <sup>b</sup>
		Casein	0.01	kg	Notarnicola et al. (2011) <sup>b</sup>
		Wax	0.005	kg	Notarnicola et al. (2011) <sup>b</sup>
		Acrylic polymer	0.04	kg	Notarnicola et al. (2011) <sup>b</sup>
		Dye	0.004	kg	Notarnicola et al. (2011) <sup>b</sup>
		Lacquer	0.02	kg	Notarnicola et al. (2011) <sup>b</sup>
		Electricity	3.35	kWh	Joseph & Nithya (2009)
Ουτρυτ	All processes	1 m² of finished bovine leather (chromium-tanned)	1.05	kg	Based on <b>Table S3</b>
EMISSIONS	Tanning and post-tanning	Chromium (III)-containing solid waste	1.58	kg	Yu et al. (2021) ª

a: The original values are normalized for 5.25 kg of wet-salted hides.
b: The original values are normalized for 5.25 kg of raw hides, which has the same weight of wet-salted hides, as shown in Table S2.

# 2.1.2. Vegetable tanning

**Figure S7** represents the vegetable tanning processes (in shades of brown and orange). This figure is inspired by the descriptions given in Baquero et al. (2021), Buljan & Král (2019), Laurenti et al. (2017), and Ömür et al. (2016).



Figure S7 - Manufacturing process system for  $1 \text{ m}^2$  of vegetable-tanned leather

Right after leaving the beamhouse, the animal pelts are washed and proceed directly to the tanning process. Falcão & Araújo (2018) demonstrate that there is a wide range of plant-based tannins suitable for vegetable tanning. In this model, the use of quebracho (wood chips) and mimosa (bark) is assumed, following the inventory described by Baquero et al. (2021). However, since this source only accounts for the post-tanning processes such as neutralization, retanning, fatliquoring, and washing, the inventory is

complemented with data from Ömür et al. (2016). The finishing process is assumed to be the same as that of chromium-tanned leather, as depicted by Notarnicola et al. (2011). **Table S15** compiles the life cycle inventory for the vegetable treatment, with proxies presented in **Table S16** - Proxies for activities in the chromium-tanned and vegetable-tanned product systems**Table S16**.

	Process	Activity	Value	Unit	Reference
	Washing	Pelts ready for tanning	3.39	kg	Table S12Table S12
		Water (200% w/w – 22°C)	6.78	kg	Ömür et al. (2016)
	Tanning	Water (200% w/w – 22°C)	6.78	kg	Ömür et al. (2016)
		Synthetic tanning product (2% w/w)	0.068	kg	Ömür et al. (2016)
		Tannin (10% w/w)	0.339	kg	Ömür et al. (2016)
		Water (60% w/w – 50°C)	2.034	kg	Ömür et al. (2016)
		Synthetic fatliquor (6% w/w)	0.203	kg	Ömür et al. (2016)
		Heat (water heating for 10 min)	0.25	MJ	Ömür et al. (2016)
		Electricity (machinery operation)	0.5	kWh	Author's assumption
	Neutralization	Sodium formate	0.03	kg	Baquero et al. (2021) ª
		Water (20°C)	6.30	kg	Baquero et al. (2021) <sup>a</sup>
		Electricity (2h15min)	0.40	kWh	Baquero et al. (2021)ª
NPUT	Retanning	Sodium polyphosphate	0.04	kg	Baquero et al. (2021) <b>a</b>
_		Synthetic retanning products	0.32	kg	Baquero et al. (2021) ª
		Acrylic copolymers	0.21	kg	Baquero et al. (2021) <sup>a</sup>
		Synthetic phenolic retaining products	0.42	kg	Baquero et al. (2021) ª
		Bark of mimosa (atomized)	1.68	kg	Baquero et al. (2021)ª
		Wood chips of quebracho (atomized)	0.42	kg	Baquero et al. (2021)ª
		Biopolymer	0.21	kg	Baquero et al. (2021) ª
		Water (30°C)	5.25	kg	Baquero et al. (2021) ª
		Heat (natural gas)	0.22	MJ	Baquero et al. (2021) ª
		Electricity (11h15min)	2.50	kWh	Baquero et al. (2021) <sup>a</sup>
	Fatliquoring	Synthetic retanning products	0.34	kg	Baquero et al. (2021) <sup>a</sup>
		Fat liquors agents	0.42	kg	Baquero et al. (2021) a
		Fungicide	0.01	kg	Baquero et al. (2021) ª

**Table S15** - Life cycle inventory for tanning  $1 m^2$  of bovine leather with vegetable tannins.

		Formic acid	0.11	kg	Baquero et al. (2021) ª
		Water (30°C, 45°C)	16.28	kg	Baquero et al. (2021) ª
		Heat (natural gas)	1.44	MJ	Baquero et al. (2021) ª
		Electricity (3h40min)	0.71	kWh	Baquero et al. (2021) ª
	Washing	Oxalic acid	0.02	kg	Baquero et al. (2021) ª
		EDTA (ion sequestering agent)	0.02	kg	Baquero et al. (2021) ª
		Water (20°C, 30°C)	21.00	kg	Baquero et al. (2021) ª
		Heat (natural gas)	0.44	MJ	Baquero et al. (2021) ª
		Electricity (40min)	0.16	kWh	Baquero et al. (2021) ª
	Finishing	Aniline	0.01	kg	Notarnicola et al. (2011) <sup>b</sup>
		Casein	0.01	kg	Notarnicola et al. (2011) <sup>b</sup>
		Wax	0.005	kg	Notarnicola et al. (2011) <sup>b</sup>
		Acrylic polymer	0.04	kg	Notarnicola et al. (2011) <sup>b</sup>
		Dye	0.004	kg	Notarnicola et al. (2011) <sup>b</sup>
		Lacquer	0.02	kg	Notarnicola et al. (2011) <sup>b</sup>
		Electricity	3.35	kWh	Joseph & Nithya (2009)
OUTPUT	All processes	1 m² of finished bovine leather (vegetable-tanned)	1.05	kg	Based on Table S3

<sup>a</sup>: The original values are normalized for 5.25 kg of wet-salted hides.

**b**: The original values are normalized for 5.25 kg of raw hides, which has the same weight of wet-salted hides, as shown in **Table S2**.

## 2.1.3. Allocations

**Figure S5** illustrates that certain processes in the leather manufacturing system generate multiple outputs, demanding the use of allocation methods in accordance with the ISO 14044:2006 guidelines (European Committee for Standardization, 2006). While the allocation rules differ across product systems, the Product Environmental Footprint Category Rules (PEFCR) for leather (Rosa-Giglio et al., 2018) provide the allocation factors presented in the same figure.

According to the PEFCR, milk farming impacts are allocated as follows: 88% to milk production and 12% to the live animal that will be slaughtered. During the slaughtering process, only 3.5% of the impacts are allocated to hides and skins. In the core processes, 9% of the impacts are attributed to hair, while flesh splits receive an allocation of 31%. Consequently, 60% of the impacts from tanning are allocated to finished leather derived from raw hides.

	Activity	Proxy	Reference
Table S14	Acrylic resin	Market for acrylic binder, with water, in 54% solution state (RER)	Rosa-Giglio et al. (2018)
	Dicyandiamide resin	Market for anionic resin (RER)	Rosa-Giglio et al. (2018)
	Mimosa extract	Market for phenol (RER)	Yu et al. (2021)
	Dyestuff	Market for aniline (RER)	Rosa-Giglio et al. (2018)
	Fatliquor	Market for fatty acid (GLO)	Rosa-Giglio et al. (2018)
Table S15	Synthetic tanning product	Market for formaldehyde (RER)	Rosa-Giglio et al. (2018)
	Tannin	80% Market for bark chips, wet, measured as dry mass (Europe without Switzerland), 20% Market for wood chips, wet, measured as dry mass (Europe without Switzerland) <sup>a</sup>	Author's assumption
	Synthetic fatliquor	Market for fatty acid (GLO)	Rosa-Giglio et al. (2018)
	Sodium polyphosphate	Market for sodium tripolyphosphate (GLO)	Rosa-Giglio et al. (2018)
	Synthetic retanning products	Market for formaldehyde (RER)	Rosa-Giglio et al. (2018)
	Acrylic copolymers	Market for acrylic binder, with water, in 54% solution state (RER)	Rosa-Giglio et al. (2018)
	Synthetic phenolic retaining products	Market for polycarboxylates, 40% active substance (RER)	Rosa-Giglio et al. (2018)
	Bark of mimosa (atomized)	Market for bark chips, wet, measured as dry mass (Europe without Switzerland)	Falcão & Araújo (2018)
	Wood chips of quebracho (atomized)	Market for wood chips, wet, measured as dry mass (Europe without Switzerland)	Falcão & Araújo (2018)
	Biopolymer	Market for polyester-complexed starch biopolymer (GLO)	Rosa-Giglio et al. (2018)
	Fungicide	Market for benzo[thia]diazole-compound (GLO)	Rosa-Giglio et al. (2018)
Both Table S14 and	Casein	Market for acrylic binder, with water, in 54% solution state (RER)	Rosa-Giglio et al. (2018)
Table S15	Wax	Market for paraffin (GLO)	Rosa-Giglio et al. (2018)
	Acrylic polymer	Market for acrylic binder, with water, in 54% solution state (RER)	Rosa-Giglio et al. (2018)
	Dye	Market for aniline (RER)	Rosa-Giglio et al. (2018)
	Lacquer	Market for acrylic binder, with water, in 54% solution state (RER)	Rosa-Giglio et al. (2018)

Table S16 - Proxies for activities in the chromium-tanned and vegetable-tanned product systems

<sup>a</sup>: The vegetable tannins are modelled like in Baquero et al. (2021), with the proportion of 80% (w/w) of mimosa bark to 20% (w/w) of quebracho wood chips.

# 2.2. Polyurethane (PU) leather-like material

# 2.2.1. Manufacturing process

**Figure S8** depicts the manufacturing for synthetic leather from polyurethane (PU). The system is an adaptation of the wet processes described in Sphera Solutions GmbH (2020), Sur et al. (2018) and Xia et al. (2007). **Table S17** exposes the complete inventory for this system, while **Table S18** disclosures the list of proxy activities used in the model.



Figure S8 - Manufacturing process system for polyurethane (PU) leather-like material

As Xia et al. (2010) elaborate on 'Example 1', the manufacturing process starts with the polymer preparation. This solution contains a polyol (7% w/w), 1,4-butyleneglycol (3% w/w), diphenylmethane-4,4'-diisocyanate (14% w/w), a colorant (5% w/w), and dimethylformamide (DMF) (70% w/w) as the main organic solvent (Sur et al., 2018). The mix is homogenized under stirring for 15 minutes, at 100°C for 1h. This consumes 0.945 kWh, divided between electricity for heating and operating the machinery, considering a homogenizer like the model <u>KG-P2009 by Quzhou KingGo Machinery Co., Ltd</u> (power equals 1.5 kW).

This solution is poured in a nonwoven or knitted fabric. It is assumed that the basis material is a 100% nonwoven polyester, which is one of the cheapest options of textiles available in the market. The soaked fabric is then submerged in a solution of 4% (w/w) DMF. A liquor ratio of 10:1 (solution to final material weight) is estimated, which translates into 3.26 L of water for one square meter of material. The polymerization happens in 5 minutes at 30°C, which consumes 0.15 MJ for water heating, assuming an efficiency of 80%.

The polymerized material is then washed in water at 55°C for 40 minutes. Once again, the liquor ratio of 10:1 (solution to final material weight) is adopted. 0.94 MJ is required to heat 3.96 L water, accounting for heat losses (assumed to be 20%). The manufacturing process ends with a drying process that consumes 1.23 kWh, if we use a drier like the model <u>HGQ10 by Jiangsu Sunflower Machinery Co., Ltd</u>. It has 3.7 kW of power, and it is operated for 20 minutes at 100°C, as recommended by Xia et al. (2007).

## 2.2.2. Allocation

Allocation is not necessary for PU synthetic, as the system does not yield multiple products.

	Process	Activity	Value	Unit	Reference
	Polymer preparation	Polyester polyol	0.023	kg	Based on Xia et al. (2010)
		Colorant	0.016	kg	Based on Xia et al. (2010)
		1,4-butyleneglycol	0.01	kg	Based on Xia et al. (2010)
		Dimethylformamide (DMF)	0.22	kg	Based on Xia et al. (2010)
		Diphenylmethane-4,4'-diisocyanate	0.044	kg	Based on Xia et al. (2010)
F		Electricity (homogenizer for 15 min + heating to 100°C for 1.5 hours)	0.945	kWh	Based on Xia et al. (2010)
NPU	Coating	Polyblend fabric (1 m²)	0.15	kg	Based on Xia et al. (2010)
	Submersion	Dimethylformamide (DMF)	0.13	kg	Based on Xia et al. (2010)
		Water (30°C)	3.26	kg	Based on Xia et al. (2010)
		Heat (for 5 min)	0.15	MJ	Based on Xia et al. (2010)
	Washing	Water (55°C)	3.26	kg	Based on Xia et al. (2010)
		Heat (for 40 min)	0.94	MJ	Based on Xia et al. (2010)
	Drying	Electricity (drying at 100°C for 20 min)	1.23	kWh	Based on Xia et al. (2010)
ουτρυτ	All processes	1 m² of polyurethane leather-like material	0.326	kg	Author's calculation

**Table S17**- Life cycle inventory to produce  $1 m^2$  of polyurethane leather-like material (wet process)

Table S18 - Proxies for activities in the PU lea	ather-like material product system
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	Activity	Proxy	Reference
Table S17	Polyester polyol	Market for polyol (RoW)	Author's assumption
Colorant 1,4-butyleneglycol		Market for aniline (RoW)	Rosa-Giglio et al. (2018)
		Market for diethylene glycol (RoW) <sup>a</sup>	Author's assumption
	Dimethylformamide (DMF)	Market for N, N-dimethylformamide (GLO)	Author's assumption
	Diphenylmethane-4,4'- diisocyanate	Market for methylene diphenyl diisocyanate (RoW)	Sphera Solutions GmbH (2020)

<sup>a</sup>: Other chemicals like propylene glycol, ethylene glycol, and triethylene glycol are also considered. However, no real changes in the results are observed. Thus, diethylene glycol is used for chemical similarity with 1,4-butyleneglycol.

# 2.3. Polyvinylchloride (PVC) leather-like material

# 2.3.1. Manufacturing process

**Figure S9** depicts the manufacturing process for synthetic leather made of polyvinylchloride (PVC), a fossilbased polymer, as an extension of the descriptions given by Baitz et al. (2004) and Gurera & Bhushan (2018).



Figure S9 - Manufacturing process system for polyvinylchloride (PVC) leather-like material

In step one, about 100 grams of a solution containing 55% (w/w) of fossil-based polymer, i.e., PVC, 40% (w/w) plasticizer, 1% (w/w) stabilizer, and up to 5% (w/w) of fillers is poured on 1 m<sup>2</sup> of silicone-coated paper. The filler is composed by polysiloxane (surface modifier and releasing agent) and anionic surfactants (coloring dyes). The amount in kilograms of each chemical is presented in **Table S19**, as well as proxies for these, in **Table S21**. Homogenizing the chemical demand approximately 0.25 kWh, assuming a processing time of 2.5 minutes in an equipment with 1.5 kW of power, like the model <u>KG-P2009 by Quzhou KingGo</u> Machinery Co., Ltd.

The thin layer is pressed in a calender. We adopted a specialized machine like the ones commercialized by <u>Zhangjiagang Sino-Tech Machinery Co., Ltd</u>. Similar machines process around 30 meters of PVC synthetic leather per minute, which means 1 meter every 2 seconds. Assuming power of 320 kW like the example, the machine demands approximately 0.18 kWh to process one square meter.

After heating the base layer, additional 100 g of polymer solution plus a riser (2% w/w) is poured on top of it. After heating, this layer expands, creating a foamy material. The energy demanded in this process is both for homogenizing the solution and riser, and for calendering the material once again.

The next step is to reinforce the material with a fabric backing. Schäfer et al. (2021) discuss that several types of fabrics can be used for backing synthetic leather, being the polyester-based ones the most common. For this model, we chose a 100% polyester backing with a glue coating, as mentioned by Gurera & Bhushan (2018). The material goes once again through the calender, consuming 0.18 kWh.

The last step is to coat the material with a varnish for shine, or waterproofing. Acrylic varnish is assumed here, in a proportion of 1% of final weight, i.e., 0.326 kg for 1 m<sup>2</sup>. This value is in line with what is used in the industry, as explored by Bhattacharjee et al. (2014). The process finishes with another round through the calender, which means the consumption of additional 0.18 kWh.

## 2.3.2. Allocation

Co-production is not in visualized in the system displayed in Figure S9, thus allocation is not necessary.

_	Process	Activity	Value	Unit	Reference
	Polymer	Polyvinylchloride (PVC) (polymer)	0.11	kg	Baitz et al. (2004)
	production	Non-ionic surfactant (plasticizer)	0.08	kg	Baitz et al. (2004)
5		Stabilizer	0.002	kg	Baitz et al. (2004)
NP		Polysiloxane (filler)	0.004	kg	Baitz et al. (2004)
		Anionic surfactant (filler)	0.006	kg	Baitz et al. (2004)
		Electricity (homogenizer for 5 min)	0.25	kWh	Author's calculation
оитрит	Polymer solution production	Polymer solution	0.2	kg	Author's calculation

Table S19 - Polymer solution preparation for polyvinylchloride leather-like material

	Process	Activity	Value	Unit	Reference
	1 <sup>st</sup> pour	Polymer solution	0.1	kg	Author's calculation
		Silicone coated paper (1 m <sup>2</sup> )	0.07	kg	Author's calculation
		Electricity (heated calender)	0.18	kWh	Author's calculation
	2 <sup>nd</sup> pour	Polymer solution	0.1	kg	Author's calculation
		Riser (2% w/w)	0.002	kg	Based on Gurera & Bhushan (2018)
INPUT		Electricity (homogenizer for 5 min + heated calender)	0.428	kWh	Author's calculation
	Backing	Polyester fabric, non-woven	0.15	kg	Author's calculation
		Vinyl acetate (glue)	0.5	kg	Author's calculation
		Electricity (heated calender)	0.178	kWh	Author's calculation
	Finishing	Acrylic varnish (1 wt.)	0.0033	kg	Bhattacharjee et al. (2014)
		Electricity (heated calender)	0.178	kWh	Author's calculation
ουτρυτ	All processes	1 m <sup>2</sup> of polyvinylchloride (PVC) leather like material	0.326	kg	Author's calculation

 Table S20 - Life cycle inventory to produce  $1 m^2$  of polyvinylchloride leather-like material

 Table S21 - Proxies for activities in the PVC leather-like material product system

	Activity	Proxy	Reference
Table S19	Non-ionic surfactant	Non-ionic surfactant, fatty acid derivate, GLO	Cortés et al. (2021)
	Stabilizer	Bisphenol A, powder, GLO	Doworkin (1989)
	Polysiloxane	Polydimethylsiloxane, GLO	Author's assumption
	Anionic surfactant	Alkylbenzene sulfonate, linear, petrochemical, GLO	Cortés et al. (2021)
Table S20	Silicone coated paper	Paper, woodfree, coated, RoW	Author's assumption
	Riser	Dioctyl terephthalate, GLO	Howick (1998)
	Acrylic varnish	Acrylic varnish, with water, in 53% solution state, GLO	Author's assumption

# 2.4. Pineapple leaf fibers leather-like material

# 2.4.1. Manufacturing process

**Figure S10** shows the product system for pineapple leaf fibers (PALF) LLM. The flowchart derives from Hijosa et al. (2013), a patent owned by Ananas Anam<sup>™</sup>, and it is supplemented with other sources like Chollakup et al. (2017), Hazarika et al. (2017), Li et al. (2011), and Salsabila et al. (2021). **Table S22** displays the full life cycle inventory of this product system.





After harvest, the pineapple leaves go through a decorticating process, in which the fibers are extracted. It is estimated that 1 m<sup>2</sup> requires 480 pineapple leaves (Ananas Anam Ltd, 2021). Each leaf weights between 15 and 50 grams, according to Franck (2005). For this study, it is assumed that the average leaf weight is 20 g, with a fiber production yield of 3% per leaf (Leao et al., 2009). This results in a demand for 9.6 kg of pineapple leaves, from which 0.288 kg is fibers, which is enough to fulfill the requirement of 70% (0.283 kg) of PALF content in the final leather-like material. The mechanical decorticator model <u>SL-400 by</u> <u>Zhengzhou Shuliy Machinery Co., LTD.</u> was adopted. It processes between 500 and 1000 kg of material per hour. The value of 750 kg/h is used for this analysis and motor power of 7.5 kW. For 9.6 kg of pineapple leaves, 0.768 minutes are necessary. We found that 0.1 kWh is demanded for this model.

A chemical degumming step follows, to remove the gum that keeps the fibers together, composed of pectin (Padzil et al., 2020). Degumming can be done chemically or biologically using enzymes (Pandit et al., 2020)

but in this case, alkali degumming is considered, using sodium hydroxide as a chemical agent. Hijosa et al. (2013) describe the use of a 2% (w/v) solution of NaOH. This contrasts with other sources like Hazarika et al. (2017) that adopt concentrations of 1% (w/v), and Chollakup et al. (2017) that uses a 4% (w/v) NaOH solution plus a 4% (w/v) sodium carbonate liquor.

The liquor ratio to fiber content also varies through diverse sources. While the patent for Piñatex® describes a ratio of 25:1 (liquor to fiber content), the literature also shows 10:1 (Hazarika et al., 2017; Zhang et al., 2021), 15:1 (Poletanovic et al., 2021), 20:1 (Chollakup et al., 2017; Li et al., 2011), and even 100:1 (Munawar et al., 2008). This information is critical because the degumming process is conducted in hot water. Thus, the volume of alkali solution per mass of fibers determines the energy consumption for water heating. The time and temperature of degumming are variable as well. The literature reports temperatures between 80°C (Hijosa et al., 2013; Li et al., 2011) to 100°C (Chollakup et al., 2017), for between 30 minutes (Hijosa et al., 2013) to up to 12 hours (Chiliveri et al., 2016). For this study, we adopted the original protocol described by Hijosa et al. (2013), with a 2% (w/v) NaOH solution in the liquor ratio (solution to fiber) of 25:1. This means 240 L of distilled water is demanded. The energy for heating it and keeping it at 80°C for 30 minutes equals 87.48 MJ. For rinsing, a proportion of 1:1 (distilled water to fiber) is assumed.

The air laying and needle punching machine connects the fibers, which yield a PALF mat. The model <u>TDL-FZ by Qingdao Tongda Textile Machinery Co., Ltd.</u>, can process between 300 kg and 5 tons of material per day (used: 2,650 kg/day or 110.42 kg/h). For processing 0.283 kg (the 70% fiber content in the final product), 0.1521 minutes are demanded. Its motor power ranges between 7 and 11 kW, with 9 kW being adopted in this study. This results in 0.02 kWh of electricity demand.

Later, the fibers mat is fused with a plasticized (in the case of Piñatex<sup>®</sup>, it is polylactic acid (PLA) from corn). The final product contains a 20% (w/w) content of PLA (Ananas Anam Ltd., 2021), which translates in 0.081 kg of plasticizer. The PLA is cured on a hot press, at 120°C for 30 seconds. The model under use is similar to <u>TMYJ02 by Shandong Hummingbird machinery Co., Ltd.</u>, with power equals to 26 kW. Thus, operating the equipment for 30 seconds demand 0.22 kWh of electricity.

The fused mat receives a coating layer of plastic polymer. According to Ananas Anam Ltd. (2021), the plasticizer is polyurethane (PU), with a concentration of 10% (w/w) of the final product, i.e., 0.04 kg. However, since *ecoinvent* does not present an activity that matches the PU in polymer solution, the model adopts polyvinylchloride (PVC) instead. This small alteration is supported by Hijosa et al. (2013), which points PVC as a possible plastic material for leather-like material made of pineapple leaf fibers.

The material is fused into the mat through hot press, in the same temperature and time as the previous step. Hence, the energy demand is the same as the fusing step.

Finally, the non-woven material is finished with leather finishing products. Hijosa et al. (2013) propose the use of Astacin Finish PFM TF<sup>™</sup> from BASF<sup>™</sup>, which is a commercial acrylic compound. Acrylic resins and varnishes are suggested as a leather finishing product that could proxy the product by BASF<sup>™</sup> (Alonso et al., 2005). Since Hijosa et al. (2013) do not recommend any specific quantity of the product, 0.004 kg (1% of final weight) is assumed. This quantity agrees with Bhattacharjee et al. (2014) that show that leather finishing is usually applied in an amount between 2 and 100 g/m<sup>2</sup> of material. After the coating, the mat goes through a calender. The model <u>XY-2I1120 by Qingdao Fineyear Industry Co., Ltd.</u>, process 1.5 m/min. For 1 m of material, 0.67 minutes of operation is demanded. The power of this model is 40 kW, which results in an electricity demand of 0.447 kWh. The LLM is then dried in a tumbler drier. This study uses the model <u>HGQ10 by Jiangsu Sunflower Machinery Co., Ltd.</u>, with motor power of 3.7 kW. Thus, operating it for 1 minute requires 0.062 kWh of electricity.

#### 2.4.2. Allocation

All upstream impacts, which regards to pineapple farming, are allocated to fruit production. Currently, the fruit is only part of the plant with a commercial value (Prado et al., 2020). As part of the harvest process, farmers must remove the leaves from the pineapple bush to promote plant regrowth and flourishing (Pandit et al., 2020). These leaves are detached manually or mechanically, and they are taken as agriculture waste (Siakeng et al., 2020), being burned most part of the time (Padzil et al., 2020). Therefore, PALF should not carry environmental burdens from farming and harvesting. This assumption aligns with Hildebrandt et al. (2021) and Salsabila et al. (2021) that do LCAs for applications of PALF as leather alternatives, and paper, respectively.

	Process	Activity	Value	Unit	Reference
	Decorticating	Pineapple leaves	9.6	kg	Based on Ananas Anam Ltd., (2021)
		Electricity (Operation for 0.8 min)	0.10	kWh	Based on Hijosa et al., (2013)
	Chemical degumming	Sodium hydroxide 2% (in a 50% solution) <sup>a</sup>	9.6	kg	Based on Hijosa et al., (2013)
		Distilled water (95°C)	230.4	kg	Based on Hijosa et al., (2013)
		Distilled water (22°C - rinsing)	9.6	kg	Based on Hijosa et al., (2013)
		Heat (80°C for 30 min)	87.48	MJ	Based on Hijosa et al., (2013)
F	Air laying	Electricity (Operation for 0.15 min)	0.02	kWh	Based on Hijosa et al., (2013)
NPU	Fusing	Polylactic acid (PLA)	0.081	kg	Based on Hijosa et al., (2013)
		Electricity (Press at 120°C for 30 sec)	0.22	kWh	Based on Hijosa et al., (2013)
	Curing	Polyvinylchloride (PVC)	0.04	kg	Based on Hijosa et al., (2013)
		Electricity (Press at 120°C for 30 sec)	0.22	kWh	Based on Hijosa et al., (2013)
	Calendering	Acrylic resin (finishing product)	0.004	kg	Based on Hijosa et al., (2013)
		Electricity (Operation for 1 min)	0.447	kWh	Based on Hijosa et al., (2013)
	Drying	Electricity (Drying at 160°C for 1 min)	0.062	kWh	Based on Hijosa et al., (2013)
Ουτρυτ	Tumbling	PALF leather-like material	0.404	kg	Based on Hijosa et al., (2013)

 Table S22 - Life cycle inventory to produce 1 m² of pineapple leaf fibers leather-like material

<sup>a</sup>: describes the use of a 2% (w/v) solution of NaOH for chemical degumming. However, the *ecoinvent* activity for NaOH is a 50% (v/v) solution in water. Thus, it is necessary to apply the chemical relation  $C_1 \cdot V_1 = C_2 \cdot V_2$ , in which  $C_1$  and  $C_2$  represent the initial and final concentration of NaOH, respectively, while  $V_1$  represents the volume of the 50% solution that is necessary in the final volume (V<sub>2</sub>). For a  $V_2 = 240$  L of solution, 9.6 L of the 50% NaOH solution is used. The remainder (230.4 L) is completed with distilled water.

## 2.5. Cactus leather-like material

# 2.5.1. Manufacturing process

**Figure S11** presents the manufacturing process for cactus leather-like material. This process has never been described in the scientific literature before and it is retrieved for the patent by Cázarez Duarte & Lopez Velarde (2021). When necessary, the system is complemented with Agarwala et al. (2007), Ramírez-Arpide et al. (2018), and Vergel-Rangel et al. (2021), which regards to other applications of nopal cacti, like dietary supplement, biogas, and as food source. The life cycle inventory is presented in **Table S23.Table S23** - Life cycle inventory to produce 1 m<sup>2</sup> of cactus leather-like material



Figure S11 - Manufacturing process system for cactus leather-like material.

As DESSERTO<sup>®</sup> (2023) shows, 1 meter of cactus LLM requires 3 mature cladodes (nopal's leaves) with 6 to 8 months of age. Hernández-Becerra et al. (2022) describes that nopal leaf after 200 days (approximately 6.5 months) weights on average 700 g. Thus, it is reasonable to assume that 2.1 kg of cacti is required per square meter of finished material. The organic farming described by Vergel-Rangel et al. (2021) is adopted, with the inputs being normalized for 2.1 kg of output. After the harvest, the cladodes are cleaned and the glochids (hair-like spines structures) are removed manually. The cleaning process occurs with a 2% (v/v) sodium hypochlorite solution in a liquor ratio of 1:1 (solution to cactus), as supported by Agarwala et al. (2007). The cladodes soak in the solution for 10 minutes and then are milled in rough granule of 1 to 2 cm of diameter (Cázarez Duarte & Lopez Velarde, 2021). The machine for this process is the model <u>SG40 by Shandong Dexi Machine Co., Ltd.</u>, that processes between 0.3 and 0.8 tons per hour (used: 550 kg/h). For 2.1 kg, it needs 0.23 minutes. With a motor power of 7.5 kW, it consumes 0.029 kWh for this operation.

This material dries in the sun, which saves energy, and then is milled again to a thinner powder, consuming 0.029 kWh, once again. The powder is sifted with an in industrial vibration sifter, and smaller particles are selected. The model <u>XZS-500 by Xinxiang Yongquing Screen Machinery Co., Ltd</u>, processes 600 kg/h. Thus, it takes 0.21 minutes to process 2.1 kg. With a motor power of 0.25 kW, it requires 0.001 kWh for this process. After sifting, only 6% of the original weight of the cactus cladodes is left.

Then, the polymer addition happens in a ratio of 3:1 (polyurethane to cactus powder). As *ecoinvent* does not have an activity for polyurethane binder or resin, polyvinylchloride (PVC) was adopted as a proxy. The original patent by Cázarez Duarte & Lopez Velarde (2021) supports this decision, as the formulation could use different polymers, including PVC. As Alternative Leather Co.,(2023) display on their website, 1 m<sup>2</sup> of Desserto<sup>®</sup> is composed by 80% of PU and 20% of cactus content. This source also shows that Desserto<sup>®</sup> weights between 540 and 650 g/m<sup>2</sup>. Assuming an average weight of 595 g/m<sup>2</sup>, the PU content is 476 g/m<sup>2</sup>.

The mix is homogenized for 2.5 minutes by a machine like the model <u>KG-P2009 by Quzhou KingGo</u> <u>Machinery Co., Ltd,</u> with 5 L capacity and power of 1.5 kW. This consumes 0.06 kWh of electricity. Then the composite is cured in hot press (adopted model: <u>TMYJ02 by Shandong Hummingbird Machinery Co., Ltd.</u>, with power of 26 kW) at 150°C for 1 minute, consuming 0.43 kWh. The application of the mix repeats for four times, each time adding a new layer on top of the previous one.

Finally, a layer of fabric is fused to the composite through sublimation. 150 g of 100% non-woven polyester is assumed, which is the average weight for  $1 \text{ m}^2$  of fabric. The energy used to operate a sublimation machine at 140°C (assumed power: 1 kW) is 0.17 kWh.

#### 2.5.2. Allocation

The impacts for farming, cleaning, and manufacturing are 100% allocated to the cactus leather-like material as co-production does not happen.

	Process	Activity	Value	Unit	Reference
	Agriculture ª	<i>Opuntia ficus-indica</i> cladodes	1.62	kg	Vergel-Rangel et al. (2021)
		Water	289.3	kg	Vergel-Rangel et al. (2021)
		Manure	0.252	kg	Vergel-Rangel et al. (2021)
		Potassium nitrate	0.003	kg	Vergel-Rangel et al. (2021)
		Monoammonium phosphate	0.0005	kg	Vergel-Rangel et al. (2021)
		Calcium nitrate	0.001	kg	Vergel-Rangel et al. (2021)
		Potassium sulphate	0.014	kg	Vergel-Rangel et al. (2021)
		Diesel	0.019	kg	Vergel-Rangel et al. (2021)
	Cleaning	Water	2.1	kg	Agarwala et al. (2007)
IPUT		Sodium hypochlorite 15% (final concentration: 2%)	0.28	kg	Agarwala et al. (2007)
Z	Milling (Twice)	Electricity (milling machine for 0.25 min)	0.057	kWh	Cázarez Duarte & Lopez Velarde (2021)
	Sifting	Electricity	0.001	kWh	Cázarez Duarte & Lopez Velarde (2021)
	Coating (Four	Liquid polyurethane	0.476	kg	Cázarez Duarte & Lopez Velarde (2021)
	times)	Electricity (homogenizer for 2.5 min)	0.25	kWh	Cázarez Duarte & Lopez Velarde (2021)
	Curing (Four times)	Electricity (hot press at 150°C for 1 min)	1.73	kWh	Cázarez Duarte & Lopez Velarde (2021)
	Sublimation	Polyester fabric, non-woven	0.15	kg	Cázarez Duarte & Lopez Velarde (2021)
		Electricity (sublimation machine for 10 min)	0.17	kWh	Cázarez Duarte & Lopez Velarde (2021)
UTPUT	All processes	1 m² of leather-like material from cactus	0.595	kg	Author's calculations
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 Table S23 - Life cycle inventory to produce  $1 m^2$  of cactus leather-like material

## 2.6. Kombucha bacterial cellulose leather-like material

2.6.1. Manufacturing process

**Figure S12** demonstrates the processes to produce a leather-like material from bacterial cellulose coming from kombucha fermentation. da Silva Junior et al. (2022) propose a step-by-step system, who aimed for a naturally dyed material made of reconstituted bacterial cellulose. The product system is complemented with sources like Damsin (2019) and Fernandes (2021).



Figure S12 – Manufacturing process system for kombucha bacterial cellulose (KBC) leather-like material

The process starts with static cultivation. Kombucha fermentation medium is straightforward: sucrose as the source of energy (50 g/L), green tea (10 g/L) as the source of nitrogen, phosphorous, and trace elements, citric acid (1.15 g/L) as pH stabilizer, water, and a symbiotic culture of yeast and bacteria (SCOBY) (10% w/w). The amount of water followed the same rationale exposed in **Table S10**, i.e., 72 L. This volume yields 69 L of kombucha, and 3.95 kg (wet weight) of microorganism's biofilm, which is enough for 1 m<sup>2</sup> of leather-like material.

The SCOBY ferments this mix for between 5 to 16 days, i.e., 14 days, at 30° C (da Silva et al., 2021). During this period, the microorganisms enlarge the SCOBY biofilm, an entangled structure of cellulose that will float in the medium. The energy is primarily used for heating the tea medium to 30°C. A water heater for

bioreactors with 44 kW power was assumed. Hence, its operation for 336h requires 14.8 kWh. The overall inventory is displayed in **Table S24**.

The gelatinous structure is neutralized with 0.1 M solution of NaOH. In this research, whenever the bacterial cellulose biofilm needs to be submerged, a ratio of 3:1 (water to biofilm) is assumed. Rinsing is assumed to take 10 L of water. Thus, the amount of NaOH was calculated using the molar mass (39.997 g/mol). After, being neutralized, it is pre-dyed with potassium alum 99.5% (20 g/L) and hot water (90°C, for 30 minutes, under agitation). The agitator in use is the model <u>NV0608 by Taizhou City Novia Tools Co., Ltd.</u>, with a power of 0.726 kW (1 HP). To operate it for 30 minutes the energy requirement is 0.37 kWh. Additionally, it is assumed that water is heated with electricity, since this protocol refers to a laboratory scale innovation.

Then it is dyed with natural dyes, described in **Table S25**. It is assumed that the natural dyes can be reused twice, based on the opinion of the creator of the production protocol. The natural dyes are fixed with sodium chloride (10 g/L). The process is carried under agitation at 90°C for 1h. The electricity demand is calculated for heating water and operating the agitator (0.746 kWh for the same model as the previous step). A 20% heat loss in water heating is assumed.

After that, the biofilm is smashed in a blender for the reconstitution process. According to da Silva Junior et al. (2022), reconstitution is used to provide a uniform structure and thickness to the final product. This is done by using an industrial blender. The model <u>DH903-310 Ding-Han Machinery Co., Ltd</u> (20 L) has a motor power of 1.45 kW (2 HP), and it requires 0.77 kWh to be operated for 5 minutes.

After air drying, the mat receives waxes and oil treatments, which assure waterproof characteristics. The original paper describes the use of homogenized wax from *Copernicia prunifera* and oil from *Melaleuca alternifolia*. The quantity is not described by da Silva Junior et al. (2022) , but it is assumed 100 grams of waterproofing mix per square meter. The reinforcement step is not described by da Silva Junior et al. (2022). However, using reinforcement materials is common when it comes to kombucha LLMs, as discussed by Damsin (2019). Celium™, for example, uses a polyblend backing (Bründl, 2022), while other independent producers like Ponto Biodesign, uses 100% cotton. To prevent underestimation of impacts the worst option available is assumed, meaning 100% non-woven polyester fabric, with the average weight of 0.15 kg/m<sup>2</sup>.

An overview of the inventory is showed in **Table S26**. **Table S27** indicates the proxy activities used in the model for this product system.
## 2.6.2. Allocation

Hildebrandt et al. (2021) consider kombucha as a by-product of bacterial cellulose and perform economical allocation to sugar from lignocellulosic feedstock biorefineries. However, as Ramírez Tapias et al. (2022) elaborate, the SCOBY is a by-product of kombucha production, and it is often discarded, representing a lost opportunity as it could be applied in many fields like bio-textiles (Cottet et al., 2020). Therefore, this study assumes a physical allocation following what is proposed by Mohammadshirazi & Bagheri Kalhor (2016), who shows that only 3.4% of electricity is transferred for the biofilm. It is, therefore, reasonable to assume that 3.4% of the environmental burdens of kombucha production is allocated for the creation of bacterial cellulose material.

	Process	Activity	Value	Unit	Reference
INPUT	Kombucha	Green tea leaves	0.72	kg	da Silva Junior et al. (2022)
	production	Symbiotic Culture of Bacteria and Yeast (SCOBY)	7.2	kg	da Silva Junior et al. (2022)
		Citric acid	0.08	kg	da Silva Junior et al. (2022)
		Sucrose	3.6	kg	da Silva Junior et al. (2022)
		Mineral water (22°C)	72	kg	da Silva Junior et al. (2022)
		Electricity (Bioreactor at 30°C for 14 days)	14.8	kWh	da Silva Junior et al. (2022)
OUTPUT	Kombucha	Kombucha	69	kg	Author's calculations
	production	Bacterial cellulose (wet weight)	3.55	kg	Author's calculations

Table S24 - Life cycle inv	entory for kombu	ucha production
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Table S25 - Life cycle inventory for natural dyes production

_	Process	Activity	Value	Unit	Reference
	Natural dyes	Deionized water	3.3	kg	da Silva Junior et al. (2022)
	production	Ethanol (70%)	1.11	kg	da Silva Junior et al. (2022)
5		Eucalyptus globulus (dry leaves)	0.22	kg	da Silva Junior et al. (2022)
INPL		Allium cepa L. (bulb bark)	0.22	kg	da Silva Junior et al. (2022)
		Punica granatum (dried fruit)	0.22	kg	da Silva Junior et al. (2022)
		Electricity (Boiling for 30 min)	0.75	kwh	da Silva Junior et al. (2022)
Ω	Natural dyes	Natural dyes	4.4	kg	da Silva Junior et al. (2022)
DUTF	production				
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<sup>a</sup>: da Silva Junior et al. (2022) stipulate the use of 1.25 L of natural dyes per kilogram (wet weight) of bacterial cellulose. Thus, 4.4 L of dyes is required. With that, it is possible to calculate the inputs quantity, based on the protocol provided by the same authors, in which ethanol (70%) composes 25% of the final volume, and the concentration of each plant source is 50 g/L. An 80% efficiency in water heating with electricity is assumed.

	Process	Activity	Value	Unit	Reference
	Cellulose	Bacterial cellulose biofilm	3.55	kg	Table S24
	purnication	Sodium hydroxide 0.1 M	0.043	kg	da Silva Junior et al. (2022)
		Deionized water (22°C)	10	kg	da Silva Junior et al. (2022)
		Water (70°C)	10.64	kg	da Silva Junior et al. (2022)
		Electricity (water heating for 1h) $^{f b}$	1.48	kWh	da Silva Junior et al. (2022)
	Pre-dyeing	Potassium Alum (99.5%)	0.21	kg	da Silva Junior et al. (2022) <sup>b</sup>
		Water (90°C)	10.64	kg	da Silva Junior et al. (2022)
		Electricity (water heating for 30 min + agitator operation) <sup>b</sup>	1.95	kWh	da Silva Junior et al. (2022)
	Dyeing	Natural dyes	4.4	kg	
Ţ					Table S25
IN P		Water (22°C, 90°C)	20.64	kg	da Silva Junior et al. (2022)
		Sodium chloride	0.11	kg	da Silva Junior et al. (2022) <sup>b</sup>
		Electricity (water heating for 1h + agitator operation) <sup>b</sup>	2.8	kWh	da Silva Junior et al. (2022)
	Cellulose Reconstitution	Electricity (industrial blender for 5 min)	0.12	kWh	da Silva Junior et al. (2022)
	Waterproofing	Wax from Copernicia prunifera	0.05	kg	da Silva Junior et al. (2022) <sup>b</sup>
		Oil from Melaleuca alternifolia	0.05	kg	da Silva Junior et al. (2022) <sup>b</sup>
		Electricity (wax homogenization at 75°C for 15 min)	0.77	kWh	da Silva Junior et al. (2022)
	Reinforcement	Polyester fabric, non-woven	0.15	kg	Author's calculations
		Vinyl acetate (glue)	0.05	kg	Author's calculations
Ουτρυτ	All processes	Kombucha bacterial cellulose leather- like material	0.23	kg	Author's calculations

 Table S26 - Life cycle inventory to produce  $1 m^2$  of kombucha bacterial cellulose leather-like material

<sup>a</sup>: The calculations of NaOH 0.1M uses the molar mass of 39.997 g/mol for the final volume (10.64 L).

**b**: An 80% efficiency in water heating with electricity is assumed.

<b>Table S27</b> - Proxies for activities in the KBC leather-like material product system	
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	Activity	Proxy	Reference
Table S24	Green tea leaves	Market for tea, dried, GLO	Author's assumption
	Symbiotic Culture of Bacteria and Yeast (SCOBY)	Market for fodder yeast, GLO	Author's assumption
Table S25	Ethanol (70%)	Market for ethanol, without water, in 99,7% solution state, from fermentation, GLO <sup>a</sup>	Author's assumption
	Eucalyptus globulus (dry leaves)	Market for mulberry leaves, GLO	Khan (2012)
	Allium cepa L. (bulb bark)	Market for onion, GLO	Author's assumption
	Punica granatum (dried fruit)	Market for pomegranate, GLO	Chang & Lo (2010)
Table S26	Potassium Alum (99.5%)	Market for aluminum sulphate, powder, GLO	Ohno et al. (2013)
	Wax from Copernicia prunifera	Market for coconut oil. Crude, GLO	Damsin (2019)
	Oil from Melaleuca alternifolia	Market for coconut oil. Crude, GLO	Damsin (2019)

**a**: By using 0.779 L of ethanol 99,7% plus 0.33 L of deionized water, one obtains the 1.11 L of ethanol 70% that is required.

## 2.7. Uncertainty



**Figure S13** - Monte Carlo simulation of the LCA results. A: Global warming potential; B: Cumulative energy demand; C: Water consumption potential; D: Terrestrial acidification potential; E: Particulate matter formation potential (< 2.5  $\mu$ m); F: Freshwater eutrophication potential.

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