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Steering Innovation Towards Sustainable

Electrochromic Displays: A Prospective Life Cycle

Assessment Study

A. Kamal Kamali^{1}, Daye Lee^{1*}, Romain Futsch², Edis Glogic¹, Aline Rougier^{2*}, Guido Sonnemann¹*

¹ Univ. Bordeaux, CNRS, Bordeaux INP, ISM, UMR 5255, Groupe Analyse du Cycle de Vie et Chimie Durable (CyVi), 351 Cours de la Libération, F-33405 Talence, Cédex, France

² Univ. Bordeaux, CNRS, Bordeaux INP, ICMCB, UMR 5026, 87 av du Dr Schweitzer, F-33600 Pessac, France

*A. Kamal Kamali - E-mail: ahmad-kamal.kamali@u-bordeaux.fr; Phone: +33-6-34-38-15-07

*Daye Lee - E-mail: daye.lee@u-bordeaux.fr; Phone: +33-7-68-25-73-90

*Aline Rougier - E-mail: aline.rougier@icmcb.cnrs.fr; Phone: +33-6-38-67-44-83

KEYWORDS

Electrochromic displays; Prospective; Ex-ante; Life cycle assessment; Scale-up; Emerging technology

ABSTRACT

Printed electrochromic displays (ECDs) have promising applications in visual communications. A cradle-to-gate ex-ante prospective life cycle assessment (LCA) was conducted on six ECD architectures to uncover the environmental implications of material and technological choices. Several materials were considered in ECD fabrication, including silver, carbon, and indium-tin-oxide (ITO) electrodes, plastic and paper-based substrates, and two electrolytes. The architectures differed in technology maturity levels, ranging from pilot-scale and lab-scale prototypes to conceptual designs. Regardless of their technological maturity, all architectures were scaled up to emulate impact burdens as if they were produced under an optimized industrial production scale. The analysis of ECD architectures at the early development stage, especially conceptual designs, determines their environmental viability without the need for experimental testing, resulting in significant economization of time and resources. The all-silver architecture was associated with the highest environmental impacts across all endpoint and midpoint indicators, except for the water consumption indicator. On the other hand, the all-carbon architecture exhibited the lowest environmental impacts, followed by the carbon-ITO architecture and the all-ITO architecture. Based on the environmental impact results, we could identify ECD architectures that merit further development and those that have limited potential for improvement, thus recommending to cease research and development of ECD architectures employing silver electrodes. The approach employed in this LCA guides scaling up and predicting the environmental impacts of conceptual ECD architectures. This may benefit LCA practitioners and researchers engaged in ex-ante prospective LCA studies. Furthermore, the findings of this LCA might be applicable to other electronic devices where silver, ITO, and carbon could be interchangeably used as electrodes.

Introduction

Printed electronics offer many practical advantages motivating their development as well as environmental assessment. Printing is an inexpensive and scalable method to produce lightweight electronics,¹ which increases overall material efficiency, minimizes waste, and reduces energy consumption during the production and use life cycle stages.² In efforts to substitute or reduce the dependency on critical and hazardous printable materials, the use of various bio-based and recyclable materials has been explored.² Electrochromic displays (ECDs) are one of the emerging examples of printed electronics. ECDs, known for their optical modulation under an applied voltage, have promising applications in visual communication, including smart labels, logistical monitoring, and consumer electronics.^{3,4} A multi-perspective application selection has been carried out determining applications, such as conventional low-voltage and low-frequency electronics with medium to low lifetimes, where current displays can be substituted with ECDs.⁵ Simple information is conveyed through the color change of electrochromic materials. ECDs can be fabricated on a flexible substrate, such as plastic-based or paper-based, and produced at a large scale using roll-to-roll (R2R) production processes.^{6,7} Environmental assessment can lead emerging technologies towards adopting more environmentally sustainable production routes.⁸⁻¹¹ Environmental assessment methods applied to technologies in early development stages can take advantage of high design freedom and ultimately facilitate an exploration of research pathways at a low cost, aiming to efficiently determine the optimal route to mitigating environmental burdens.¹² Ex-ante prospective life cycle assessment (LCA) is often used to evaluate the environmental performance of technologies at relatively low technology readiness levels (TRL), such as TRL 3-4 depicting technologies validated at a lab scale. The TRL system has been widely used to specify the maturity of emerging technologies.¹³ Ex-ante prospective LCA is carried out by modeling the

technology at a future point in time when technological maturity has been reached.¹¹ Data availability is the limiting factor when conducting Ex-ante LCA, as information on optimized use of materials and energy of emerging technologies requires adopting various assumptions of how systems might operate at the industrial level. Oftentimes, prospective LCAs are carried out on technologies implemented at a lab or pilot scale; this is because, at this level of technology maturity, sufficient understanding is reached to facilitate an informed scale-up to the industrial level.^{14,15} Nevertheless, conducting prospective LCA on conceptual designs (TRL 1-2) saves more time and valuable resources in developing environmentally conscious technologies, such as ECD.

Only a few studies have performed an environmental assessment on printed electronics, most of which were published after 2020. Studied printed electronic products include antennas,^{16,17} temperature sensors,¹⁸ solar cells,^{19,20} printed circuit boards,^{21,22} drug detection devices,²³ anticounterfeit labels,²⁴ and shock detection tags.²⁴ Published LCA studies aimed to identify environmental hotspots in printing materials and determine viable substituents. Observations relevant to ECDs can be identified due to the use of common materials such as nano-silver ink,^{18,21-23,25} poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS),^{19,20} polyethylene terephthalate (PET),^{19-21,25} and indium tin oxide (ITO).¹⁹ It is established that employing silver in printed electronics represents an environmental hotspot,²¹⁻²⁵ primarily due to its energy-intensive mining. A comparison of alternative conducting nanomaterials showed the superior environmental performance of copper and graphene-based inks.^{24,25} Furthermore, numerous substrates have been environmentally assessed. Nassajfar et al. studied the environmental impacts of PET, polylactic acid /glass fiber composite, paper, and fiber-glass-reinforced brominated epoxy resin substrates in printed circuit boards.²¹ Although the impact of the substrate proved to be relatively low compared to other components, the use of paper was found to be the least impactful choice.²¹ Similarly,

Prenzel et al. calculated the carbon emission stemmed from using polyether ether ketone, polyetherimide, PET, and paper substrates. The paper substrate was associated with the lowest environmental impacts,²⁵ while PET had the second lowest environmental burdens, emitting 6-8 times higher carbon emissions than paper.^{21,25} However, the pretreatment of the paper substrate in specific applications can substantially affect its environmental performance.^{22,23} For example, a cellulose nanocrystal-coated paper substrate used in a drug detection device renders the substrate an environmental hotspot.²³ It should be noted that all environmental comparisons of substrates and nano-inks made did not account for the functionality but were compared per single display.^{23,25} Consequently, for the proliferation of sustainable ECDs, further environmental research is needed, taking into account the application of the developed ECD. Thus, the functional unit should be revised to account for ECD's varied performance.

This work carries out an environmental assessment of various ECD technologies at different levels of technology maturity, including conceptual technological solutions prior to experimental testing. In the latter case, the analysis is put before the technology is even created in order to preserve valuable time and resources by steering research towards minimal environmental impacts early in the decision-making process, thus guiding researchers and entrepreneurs toward environmentally sustainable solutions. Six ECD architectures, denoted as S1-6, are environmentally assessed following the prospective ex-ante LCA method. This research is inspired by previous work on the implementation of ECD in anticounterfeiting devices, where ECD proved to be an environmental hotspot.²⁴ Therefore, five ECD architecture alternatives are investigated in addition to the one previously assessed (S1). These architectures vary in terms of materials usage, design choices, and, as mentioned, technological maturity. S1, S2, and S3 have been successfully produced on a pilot scale. S4 and S5 are theoretical concepts not tested experimentally, whereas S6 has been

experimentally validated at the lab scale. The environmental impacts associated with the six ECD architectures are discussed, focusing on key environmental concerns such as global warming potential, toxicity, and cumulative environmental damage to human health and the ecosystem. Examining the environmental impacts yields eco-design recommendations that economize resources in ECD development and enable directing it towards greener pathways. Moreover, a case study on emerging technology helps method developments in LCA by examining its challenges and approach.⁸

Materials and Methodology

LCA is a standardized method for assessing environmental impacts associated with products, processes, or services over their life cycle. LCA follows the framework of ISO standards (14040 and 14044), which entails four stages: goal and scope definition, life cycle inventory (LCI) analysis, life cycle impact assessment (LCIA), and interpretation.^{26,27} Ex-ante prospective LCA is a mode of LCA that allows practitioners to model an emerging technology in the future when it has reached maturity.¹¹ Numerous researchers have provided recommendations and proposed frameworks for this mode of LCA.²⁸

In this section, the system under investigation (i.e., ECD) is described, detailing the materials, production processes, and performance properties of the various proposed solutions. Next, the goal and objectives of the LCA are set, including selecting a functional unit allowing meaningful comparison of ECDs. Life cycle inventories are built and scaled up, accounting for all inputs (raw materials, auxiliary materials, and energy consumption) and outputs (emissions, waste) related to ECD industrial production. Section 3 discusses the LCIA results for midpoint and endpoint

indicators, providing eco-design recommendations for adopting and developing the most promising ECD solutions.

ECD environmental impacts are assessed following midpoint and endpoint approaches to better understand the results. Midpoint indicators are set early in the cause-effect chain to report the environmental effects caused by substance flows. For example, CO₂ emissions are classified into the global warming indicator. The environmental effect of substances is weighed using characterization factors.²⁹ Many midpoint indicators exist; however, seven are graphically presented in this study: global warming, human and ecotoxicity, energy demand, mineral resource depletion, ionizing radiation, and water consumption. Midpoint indicators are linked to one or two endpoint indicators, and their contribution is weighed via midpoint-to-endpoint factors. Endpoint indicators cover three areas of protection: human health, ecosystem quality, and resources. Endpoint indicators might embody a higher degree of uncertainty than midpoint indicators due to additional assumptions taken along the cause-effect chain to connect environmental flows to the three areas of protection. Despite the added uncertainty of the endpoint method, endpoint indicators aid in comprehending the environmental significance of input flows by relating them to areas of protection.³⁰ Furthermore, endpoint indicators are expressed in easily understandable units, ensuring straightforward and effective communication of the results to decision-makers. The impact on human health is quantified in disability-adjusted life years (DALYs). DALYs represent the years a person loses or lives with a disability caused by a disease or an accident. Effects on ecosystem quality are measured using species lost over time, indicating biodiversity decline. Lastly, the unit used to assess resource scarcity is the dollar, which provides a measure of the economic value associated with resource depletion.³¹

ECDs Investigated System: Materials, Manufacturing, and Performance

An ECD consists of several layers: an electrode, a counter electrode, an electrolyte, an electrochromic layer, an encapsulation layer, and a substrate. These components can be constructed using many different materials, material combinations, and deposition methods. The ECDs analyzed have explored electrode materials, two substrates, and two electrolytes, contrary to the fixed encapsulation and electrochromic material. A general schematic of the ECDs layer stacking is depicted in Figure 1. Information about the architectures' different layers and their material compositions is presented in

Table 1. Electrolyte A comprises LiTFSI:EmimTFSI and UV photopolymer, whereas electrolyte B incorporates TiO₂ as an additional component to Electrolyte A. Electrode materials investigated include silver (Ag), carbon (C), and indium tin oxide (ITO, i.e., In₂O₃:Sn).

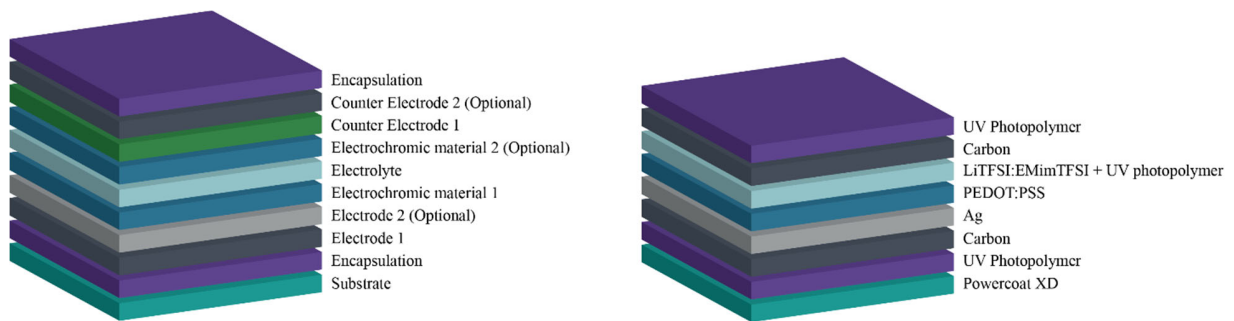


Figure 1. On the left: is a schematic illustrating ECDs' general layer configuration. On the right: a depiction of S1 layer organization.

Table 1. Overview of six ECD architectures: performance characteristics, technology readiness level, layers, and materials.

	S1	S2	S3	S4	S5	S6
Technology Readiness Level (TRL)	Pilot Scale (TRL 6-7)			Conceptual (TRL 1-2)		Lab Scale (TRL 4)
Operating voltage [V]	1.5	3.8	3.8			1.5
Optical contrast (ΔE^*)	17.3	17.5	10.6			
Switching time [Seconds]	2					
Minimum number of cycles	20*					
Encapsulation (Layer 2)	UV photopolymer					
Counter-Electrode (Layer 2)	N/A	Ag	N/A	N/A	Ag	N/A
Counter-Electrode (Layer 1)	C					ITO
Electrochromic Material (Layer 2)	N/A					PEDOT:PSS
Electrolyte	A ¹					B ²
Electrochromic Material (Layer 1)	PEDOT:PSS					
Electrode (Layer 2)	Ag	Ag	N/A			
Electrode (Layer 1)	C			ITO		
Encapsulation (Layer 1)	UV photopolymer					
Substrate	Paper-based			PET		

¹ Electrolyte A: LiTFSI:EmimTFSI + UV photopolymer

² Electrolyte B: Electrolyte A + TiO₂

* Assumed value

The selection of different architectures for ECD is based on expert knowledge and motivated by observations made in a previous LCA study.²⁴ In that previously published LCA, the environmental assessment of ECD was conducted at the pilot scale attributing more than 90% of S1 carbon emissions to silver electrodes.²⁴ In this research, various substituents have been considered in the electrodes and other ECD layers. Carbon is deemed as an acceptable substitute for silver in the electrode and counter electrode layer, which performance has been validated at the pilot scale. On the other hand, ITO is considered a viable electrode material as it is extensively used in ECDs,³² thanks to its high conductivity,³³ visible transmittance,³³ and proven functionality in applications such as smart windows,³⁴ offering performances comparable with silver electrodes.³² ITO can be deposited on a large scale using magnetron sputtering, the most used

technique in the industry,³⁵ that is compatible with temperature-sensitive substrates (plastic and paper).³⁶ Aside from electrode materials, PET substrates have been proposed as an alternative to paper-based. Trade-offs between design changes permitting size decrease and the compelling need for additional layers are also investigated. S6 is 70% smaller than other architectures but has the same active surface area, achieving the same visual function as other architectures. Although S6 requires an additional layer of PEDOT:PSS and TiO₂ as an additional component in the electrolyte. The ITO layer surface of S6 is slightly larger than the electrochromic surface to ensure contact with the electrode.

The architectures environmentally assessed in this work are at different research and development stages. The TRLs are specified for each architecture following recommendations on conducting prospective LCA.^{37,38} S1, S2, and S3 have been manufactured by Fedrigoni at a pilot scale (TRL 6-7). S4 and S5 are technological concepts untested experimentally (TRL 1-2), whereas S6 has been fabricated in the lab (TRL 4). On the pilot scale, the layers were printed using a sheet-to-sheet (S2S) semi-automatic flatbed screen printing with a blade speed between 300 and 500 mm/s. A vacuum was created on the screen-printing table to stabilize the substrate during printing. Patterns were created by polymerizing an emulsion of photo-patternable biodegradable polymer using UV light. Patterning is conducted on all ECD layers, which is required to create a pattern for communicating a predetermined message. Afterward, the unpolymerized emulsion is washed off and removed from the mask by drying. The electrolyte and encapsulation layers were cured using an Aktiprint T28-2 6 kW equipped with mercury lamps (120 W/cm) covering the full range of UV wavelengths. Other layers were thermally cured in an oven. All pilot-scale processes have been scaled up and adapted to the industrial level to facilitate the prospective environmental assessment.

The scale-up substitutes S2S with continuous R2R production. A process flowchart shows the continuous production steps envisioned on an industrial level (Figure 2).

ECD performance is determined by measuring three properties: switching time, optical contrast, and the number of operating cycles. ECD color change occurs due to a redox reaction in the electrochromic layer induced by applying a voltage in the range of 1.5-3.8 V. Switching time or latency corresponds to the time required for the active surface to change from the color of the oxidized state to the color of the reduced state. The specifications of the ECD architectures are available in

Table 1.

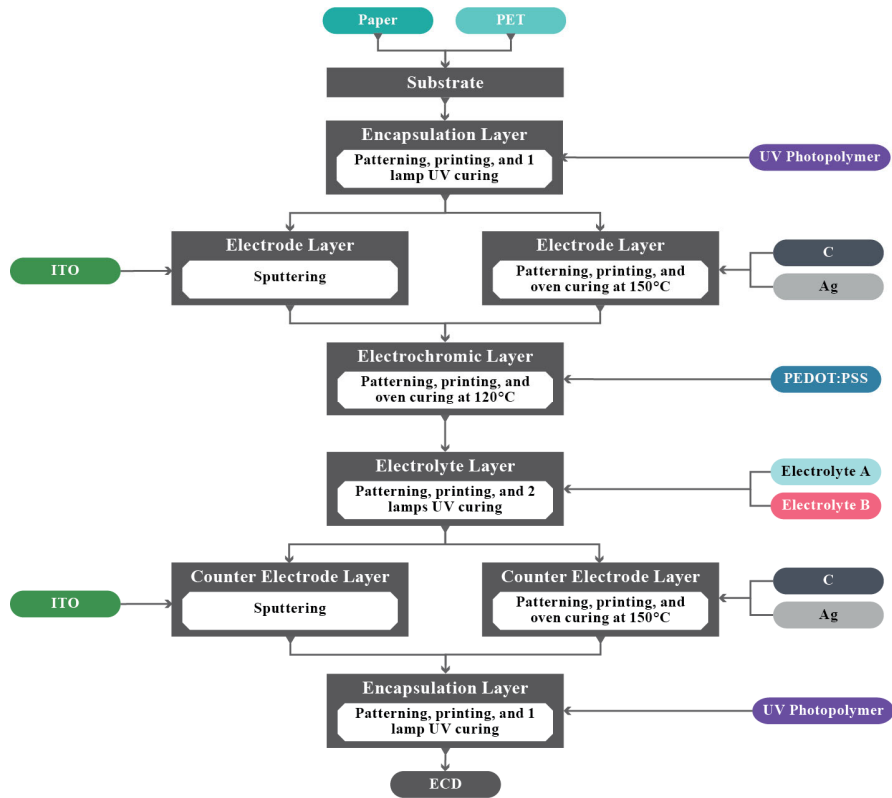


Figure 2. ECD production process flow chart: Different ECD structures utilize different materials as in Table 1.

Goal and Scope

Goal definition: This prospective LCA aims to explore research directions and technology development strategies in the field of ECDs to foster innovation that puts environmental sustainability in the forefront. Identification of materials, processes, and design variations leading to enhanced environmental performance is made possible by examining six ECD architectures (

Table 1). Before the analysis, the architectures are scaled up, permitting the prediction of their environmental impacts at industrial production conditions. Additionally, an alternative functional unit is proposed to examine the trade-offs involved in using materials associated with substantial environmental burdens but offer superior performance. A prospective LCA provides valuable information to technology developers, contributing to reducing research and development costs and ensuring the developed solution's sustainability.¹¹

Function and functional unit: The function of an ECD is to relay clear and easily readable visual information to consumers. To ensure that ECDs display a clearly visible message, it is recommended that ECD properties should achieve a minimum threshold of $10 \Delta E^*$ optical contrast. Other relevant aspects for the provision of ECDs' function are the time necessary to display the message (switching time) and how many times the messages can be displayed over the lifetime of the ECD (number of cycles). Taking into consideration the expected function from an ECD, the functional unit is defined as an ECD able to display a predefined message promptly (2 s latency) and clearly ($10 \Delta E^*$) for a minimum of 20 times. A similar functional unit has been adopted in a previous study in the context of anti-counterfeit labels.²⁴ Therefore, the reference flow needed to satisfy the defined functional unit is one ECD. The defined functional unit is valid for comparing ECDs with similar performance and for applications that do not benefit from the prolonged lifetime. However, for the functional unit to offer a fair comparison with ECDs exhibiting higher performance or with other display technologies, an alternative functional unit should be defined, see section 0.

System boundaries: A cradle-to-gate LCA was carried out, covering the production of six ECD architectures in Europe and France. The impacts associated with using and disposing of ECDs are not investigated, consistent with this study's goal of identifying environmentally sustainable

production routes. The production stage involves the acquisition and fabrication of ECD raw materials and the deposition of ECD layers: electrode, counter electrode, electrolyte, and electrochromic material. The substrate fabrication and its encapsulation are also part of the production. The environmental assessment of ECDs has been carried out using energy and material flows acquired from direct measurements at the pilot scale. The primary data collected have been adapted to reflect optimized industrial production by scaling up to emulate R2R industrial production. Industrial production is expected to be carried out in the next five years, rendering the use of current electricity mixes appropriate. The scaling up involves optimizing electricity consumption and solvent usage throughout ECDs' production. Further information on scaling up methodology and assumptions is described in the next section. Two production locations have been selected, permitting the investigation of electricity sources' effect on ECD production. An average of the European electricity mix and the French electricity mix were both considered.

The ECDs product system is composed of background and foreground subsystems. Background processes are defined here as the production of materials necessary for the fabrication of ECD. Materials production considered as background include substrate, electrode, electrochromic, and electrolyte materials fabrication. On the other hand, the foreground subsystem is noted as the ECD itself, encompassing materials and energy flows consumed during its production. Energy flows indicate all electricity consumed during the deposition and curing of ECD various layers and any concurrent processes. Primary data describe the foreground, while the background is described by secondary data such as literature.³⁹

LCIA Methods: Life cycle impact assessment was carried out using the Hierarchist perspective of ReCiPe 2016 following the midpoint and endpoint approaches,³¹ cumulative energy demand (CED),⁴⁰ and USEtox.⁴¹ Midpoint impact categories graphically presented in this study include

global warming potential, mineral resource scarcity (ReCiPe), cumulative energy demand (CED), and eco and human toxicity (USEtox). LCIA results for ReCiPe's remaining 16 midpoint indicators are available in Table 3, and interesting observations are reported. Classification and characterization of environmental impacts are aligned and dictated by the chosen LCIA methods. Ecoinvent database version 3.8 APOS⁴² was used to model background inventory processes, and modeling was done in OpenLCA 1.11.0 (GreenDelta, Berlin).

Life Cycle Inventory (LCI)

LCIs of the six studied ECDs are constructed for the foreground system based on primary data collected from Fedrigoni at the pilot scale and scaled accordingly (Table 2). The collected primary data are based on pilot scale production of three architectures (S1, S2, and S3). Inventories for the remaining architectures (S4, S5, and S6) were based on the LCIs of S1, S2, and S3, given similar production technology and materials. Data scale-up was carried out to emulate mass production conditions. The scale-up approach in this study optimizes solvent quantities used for cleaning the mask, assuming that the mask is cleaned after 1000 ECDs are printed (a total of 7.1 m²). The assessment also accounts for the degradable part of the mask (Table S1), which is assumed to produce 10 000 ECDs before requiring replacement. The electricity consumed during the production of ECD is optimized to emulate roll-to-roll (R2R) manufacturing. Moreover, all materials and electricity flows were optimized to account for the size reduction in S6.

LCIs are devised for the background system based on the inventory of S1.²⁴ S1 has been modeled in previous work as part of an anticounterfeit label,²⁴ where extensive LCIs are developed, encompassing all materials necessary for its fabrication. LCIs of the materials composing S1 are derived from literature for the following materials: nano-silver ink,⁴³ LiTFSI:EmimTFSI,⁴⁴

PEDOT:PSS,⁴⁵ and Thiophene,⁴⁶ while other materials such as the paper substrate, and carbon ink are derived from primary data measurements and expert opinion.²⁴

The LCI encompassed all electricity-consuming processes necessary to produce ECD: screen printing and oven thermal curing of the electrode, the counter electrode, and the electrochromic layer; screen printing and UV curing of the electrolyte and the two encapsulation layers; magnetron sputtering of ITO electrodes, including patterning and mask drying. While some processes are assumed to scale up linearly from S2S to R2R manufacturing, others require further optimization to reflect industrial production. For example, energy consumed by the compressor to create the vacuum was omitted as mechanical stabilization techniques were assumed to be followed in R2R manufacturing. Similarly, the oven warm-up stage was neglected from the analysis assuming continuous production. The European electricity mix is used to model the impact of these processes using the Ecoinvent dataset “market group for electricity, medium voltage”. The electricity consumption of these processes is aggregated into a single value shown in Table 2.

LCI of the ITO sputtering process was devised based on literature, as its Ecoinvent dataset was unsuitable for comparisons (Table S2). The Ecoinvent dataset for ITO sputtering titled “sputtering, indium tin oxide, for liquid crystal display” is not usable in this study. This is because ITO sputtering process is represented in the Ecoinvent database based on lab-scale data, and it has been instructed not to be compared with other deposition methods. Therefore, the dataset for ITO material titled “market for sputtering target, sintered, indium tin oxide” is used in this LCA. Additionally, Ecoinvent datasets representing auxiliary materials (i.e., argon) and energy consumption (i.e., electricity) required for ITO sputtering have been used for a comprehensive LCI of ITO sputtering. The materials efficiency of the sputtering process is deemed to be 15%. Electricity consumption during magnetron sputtering comprises plasma generated, a vacuum

pump, and other equipment.⁴⁷ The parameters of ITO sputtering for ECD have been taken from literature,⁴⁸ whereas the electricity consumption and argon gas flow have been calculated based on a previously published LCI.⁴⁷

Table 2. Life cycle inventories of the six electrochromic displays' architectures normalized for 1 ECD as output.

Component	Material	Unit	S1	S2	S3	S4	S5	S6
Substrate	Powercoat XD	g	0.113			N/A		
	PET	g	N/A			0.460	0.460	0.123
Electrodes	Silver	g	0.038	0.089	N/A	N/A	0.052	N/A
	Carbon	g	0.090			0.050	0.050	N/A
	ITO	cm ²	N/A			2	2	4
Electrochromic Material	PEDOT:PSS	g	0.015					0.032
Electrolyte	LiTFSI:EmimTFSI	g	0.030					0.008
	UV photopolymer	g	0.020					0.005
	Titanium Dioxide	g	N/A					0.001
Encapsulation	UV photopolymer	g	0.050					0.013
Energy Consumption	Electricity	W h	15.781	17.190	13.885	12.873	14.282	4.654
Cleaning Solvents	EGD	g	1.200	1.400	1.000	0.800	1.000	0.600
	Water	g	6.00					
Mask	Mask	Item	0.0001					2.68E-05

Results and Discussion

LCIA Results: Midpoint Indicators

Midpoint environmental impacts corresponding to a single ECD are detailed in Table 3 for six ECD architectures. Among these architectures, S2 has the highest environmental impacts across all midpoint indicators analyzed in this LCA, except for ionizing radiation and water consumption. In terms of ionizing radiation, S5 surpasses all other architectures due to the relatively high electricity required for the deposition of ECD layers and the embodied electricity in Argon gas

production represented as part of the ITO layer’s impact. In contrast, S6 has the highest environmental impact when considering water consumption, primarily attributed to water used in hydroelectric power plants for generating electricity consumed during Argon gas production. Conversely, S3 has the lowest environmental impact across all midpoint indicators. Upon scrutiny, the numerical values of carbon emissions and energy demand for S2 and S3 are as follows: S2 emits 46.21 g CO₂-eq and consumes 1.26 MJ of energy, approximately five times higher than S3’s emission of 10.22 CO₂-eq and energy consumption of 0.27 MJ. Moreover, the toxicity associated with S3 production is two orders of magnitude lower than that of S2, indicating a significant improvement in terms of effects on human health and biodiversity.

Table 3. Impact assessment results following the midpoint approach for the six ECD architectures considering 1 ECD as reference flow.

Impact Category	S1	S2	S3	S4	S5	S6	Unit
Global Warming	2.56E-02	4.62E-02	1.02E-02	1.68E-02	3.73E-02	1.81E-02	kg CO ₂ eq
Mineral Resource Scarcity	2.44E-03	5.78E-03	2.09E-05	3.44E-05	3.37E-03	3.76E-05	kg Cu eq
Ecotoxicity - total	3.70E+00	8.71E+00	6.61E-02	1.20E-01	5.13E+00	1.39E-01	CTU
Human Toxicity - total	9.79E-08	2.27E-07	3.70E-09	6.83E-09	1.36E-07	7.96E-09	CTU
Cumulative Energy Demand	6.93E-01	1.26E+00	2.70E-01	5.36E-01	1.10E+00	6.27E-01	MJ
Ionizing Radiation	5.33E-03	7.87E-03	3.29E-03	6.92E-03	9.47E-03	8.66E-03	kBq Co-60 eq
Water Consumption	3.85E-04	6.36E-04	1.95E-04	6.07E-04	8.58E-04	9.13E-04	m ³
Stratospheric Ozone Depletion	1.88E-08	3.61E-08	5.98E-09	9.88E-09	2.72E-08	9.82E-09	kg CFC11 eq
Ozone Formation Damage to Humans	1.29E-04	2.78E-04	2.05E-05	3.30E-05	1.82E-04	3.44E-05	kg NO _x eq
Particulate Matter Formation	6.36E-05	1.29E-04	1.54E-05	2.63E-05	9.17E-05	2.76E-05	kg PM _{2.5} eq
Terrestrial Acidification	1.51E-04	3.05E-04	3.71E-05	6.28E-05	2.17E-04	6.78E-05	kg SO ₂ eq
Toxicity Carcinogenic	3.07E-03	6.39E-03	6.27E-04	1.12E-03	4.44E-03	1.26E-03	kg 1.4-DCB
Toxicity Non-carcinogenic	4.59E-01	1.08E+00	1.08E-02	2.05E-02	6.38E-01	2.45E-02	kg 1.4-DCB
Ecotoxicity Freshwater	2.42E-02	5.69E-02	4.00E-04	7.38E-04	3.35E-02	8.67E-04	kg 1.4-DCB
Ecotoxicity Marine	3.81E-02	8.99E-02	5.32E-04	9.83E-04	5.28E-02	1.16E-03	kg 1.4-DCB
Ecotoxicity Terrestrial	2.53E-01	5.74E-01	1.88E-02	3.05E-02	3.52E-01	3.24E-02	kg 1.4-DCB
Freshwater Eutrophication	4.44E-05	9.54E-05	6.99E-06	1.38E-05	6.49E-05	1.68E-05	kg P eq
Marine eutrophication	1.59E-06	2.89E-06	6.09E-07	1.17E-06	2.48E-06	1.32E-06	kg N eq
Land Use	1.04E-03	2.07E-03	2.86E-04	7.12E-04	1.74E-03	5.86E-04	m ² a crop eq
Fossil resource scarcity	7.89E-03	1.36E-02	3.61E-03	5.40E-03	1.11E-02	5.48E-03	kg oil eq
Ozone Formation Damage to Ecosystems	1.32E-04	2.83E-04	2.11E-05	3.37E-05	1.85E-04	3.49E-05	kg NO _x eq

Comparative Assessment and Contribution Analysis of Electrochromic Displays

The environmental comparison and contribution analysis in Figure 3 reveal significant insights and findings regarding the six assessed ECD architectures. S2 employing two silver-based layers serving as an electrode and a counter electrode has the highest environmental impact, attributed to relatively high silver requirement compared to other ECD configurations (Figure 3). Similar to previous environmental research, silver was identified as an environmental hotspot.²¹⁻²⁵ The high impact of silver arises from its energy-intensive mining operations that emit significant amounts of greenhouse gases, as well as generating sulfide tailings that can contaminate air, water, and soil and have detrimental effects on human health,⁴⁹ thus causing the silver layer in ECD to dominate toxicity related impacts overshadowing other ECD materials (Figure 3-c, 3-e). Furthermore, silver is a hotspot in the mineral resource scarcity category due to its high depletion potential, given its rarity and cost (Figure 3-d). On the other hand, S3, S4, and S6 provide valid alternatives for S2. S3 surpasses the environmental performance of all other ECD architectural designs thanks to its carbon-based electrodes. Consequently, around 3-4% of S3's environmental impacts are caused by the electrodes, while more than 50% stem from electricity consumed during deposition methods. S6's compact design reduces the impact of its substrate, electrolyte and encapsulation layers compared to the other architectures by 40%. However, the impact of the ITO electrode layers is constant as their size stays the same at 2 cm² per layer, (Table 2). Thus, 90% of S6's environmental impacts are caused by the ITO electrodes, specifically from using argon gas during the sputtering deposition process. For S4, it is adopting an ITO electrode and a carbon-based counter electrode that positions the architecture's environmental impact in between that of S3 and S6. Upon scrutiny, around 50% of S4's environmental impact is caused by the single ITO electrode.

ECD architectures incorporating alternative electrode materials to silver demonstrate improved environmental performance. S5 and S1 partially substitute silver yielding 45% and 19% lower global warming potential than S2. (Figure 3-a). Similar decreases in environmental impacts are observed across all selected impact categories except for the ionizing radiation category. S5 usage of both ITO and silver electrodes contributes to the highest ionizing radiation level across all the architectures considered due to the electricity consumed for argon gas production used during ITO deposition (Figure 3-h). S3, S4, and S6 replace silver entirely, resulting in an improved environmental performance mainly attributable to that substitution. In the case of S3, adopting carbon-based materials for the electrodes lowers carbon emissions by 78% compared to S2. Thus, rendering S3 the lowest environmentally impacting design, emitting 10.22 g CO₂-eq and consuming 0.27 MJ of energy (Figure 3-a, 3-b). For S6, replacing silver with ITO in the electrodes lowers the global warming potential by 61% (Figure 3-a), emitting 18.15 g CO₂-eq and consuming 0.63 MJ of energy. In contrast, S6's water consumption is 30% higher than S2's and the highest among all considered architectures (Figure 3-i). Similarly, S6 is attributed to higher amounts of ionization radiation (Figure 3-h). Meanwhile, using ITO and carbon-based electrodes in S4 reduces carbon emissions by 64%, positioning it between S3 and S6 (Figure 3-a). In numerical terms, S4 emits 16.64 g CO₂-eq and consumes 0.54 MJ of energy. Notably, the single ITO electrode in S4 is responsible for increasing the impact of the architecture considerably in terms of ionizing radiation and water consumption (Figure 3-h, 3-i). Additionally, S3 has lower toxicity and resource depletion effects than S4 and S6. S4 and S6 higher toxicity-related impacts are attributed to the energy-intensive production of argon gas used as a medium to sputter ITO electrodes (Figure 3-6, 3-7). The toxicity from electricity generation is traced back to lignite mining, as Poland and other EU countries depend significantly on lignite for power generation.⁵⁰ Regarding the resource

depletion potential (Figure 3-d), the impact is also traced back to argon gas production. In this context, electricity consumed for argon gas production contributes to higher impacts in the resource depletion category due to nuclear fuel consumption and the materials utilized to construct electricity generation infrastructure.

Alternative materials were sought for S1 analyzed in previous work aiming to achieve a more sustainable ECD design.²⁴ The resulting material substitutions were environmentally analyzed to assess their overall impact on the architectures' environmental sustainability. Substituting paper substrates for PET of the same size had a 4-fold increase in global warming potential, emitting 0.74 g CO₂-eq compared to the previous 0.18 g (Figure 3-a). This is coherent with the literature, as paper substrates were continuously found to have a lower environmental impact than other substrates.^{21,25} In S6, incorporating titanium dioxide in the electrolyte (electrolyte B) and introducing a second layer of electrochromic material permits a size reduction of 70%. Logically, the additional material used in the electrolyte and the second layer of electrochromic material contributes to a higher environmental impact for those two components. However, it reduces the amount of solvent, substrate, and encapsulation required, as well as the electricity consumed during the production of the smaller ECD. As a result, the environmental impact of ECD without the electrode layers is reduced by around 40% (Figure 3). Adopting ITO electrodes results in higher toxicity-related impacts than carbon-based electrodes (Figure 3-f, 3-g). Furthermore, electricity consumption throughout ECD production stages constitutes a considerable part of the environmental damage caused by ECDs. Both electricity and solvent consumption depend on the number of layers an ECD has (ITO layer excluded as no solvent is needed to deposit ITO). Therefore, an ECD with fewer layers (ex. S4) has lower environmental impacts caused by these two factors (

Table 1).

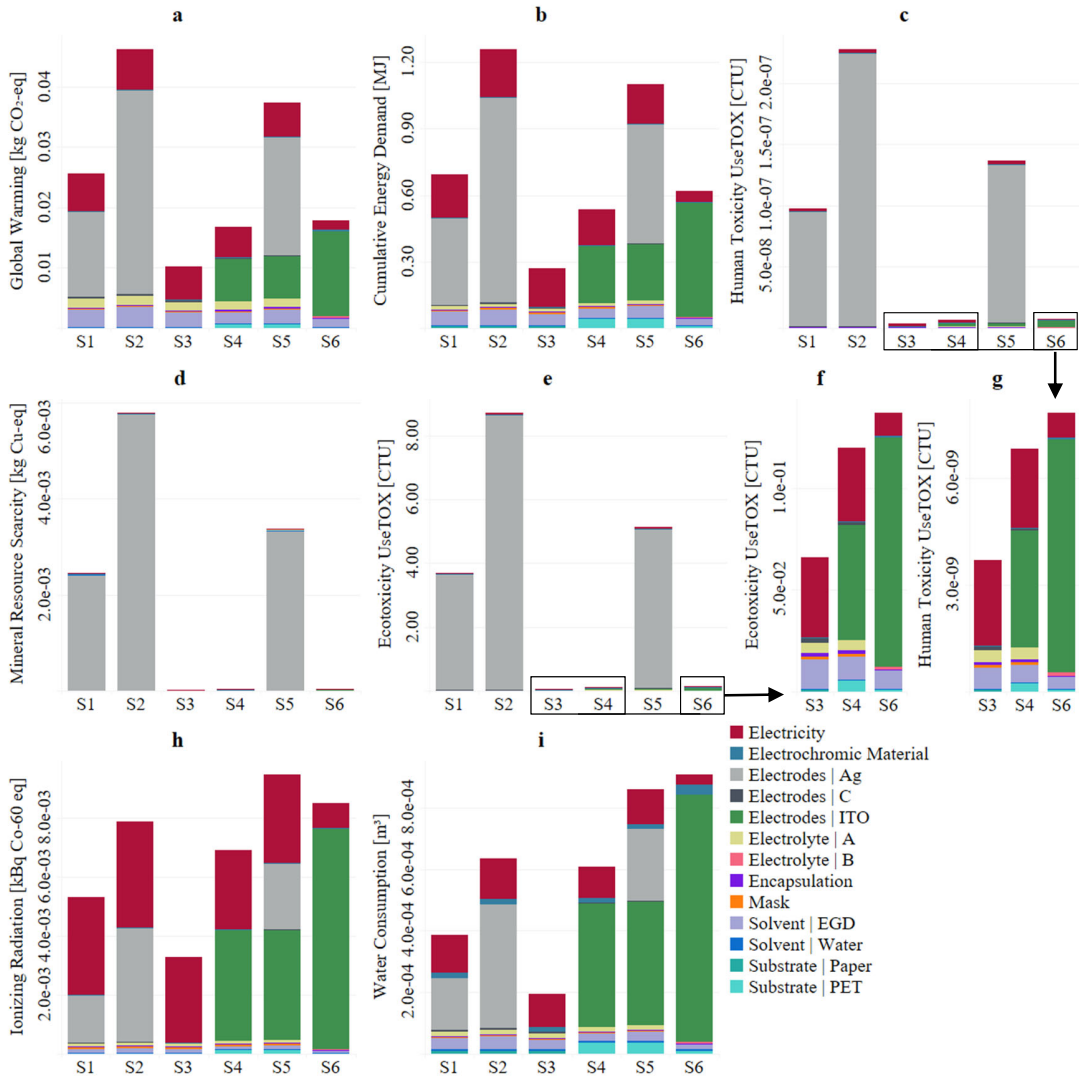


Figure 3. Midpoint environmental impacts comparison among assessed ECD architectures using a single display as a reference flow: a - global warming (ReCiPe Midpoint), b - cumulative energy demand (CED), c - human toxicity (UseTOX), d - mineral resource scarcity (ReCiPe Midpoint), e - ecotoxicity (UseTOX), h - Ionizing radiation (ReCiPe Midpoint) and i - Water consumption (ReCiPe Midpoint). Additional bar charts (f, g) are shown for the toxicity-related impacts of architectures (S3, S4, S6) to facilitate the comparison among silver-free architectures.

Influence of Management Decisions on Environmental Impacts: Exploring Plant Location

The impact of ECD production location on the overall environmental impacts was investigated by carrying out a scenario analysis considering two electricity grids. This investigation was prompted by recognizing the substantial impacts of electricity consumed during the ECD manufacturing stage. The two electricity grids considered in this scenario analysis were the European electricity mix involving a combination of fossil fuel, renewable, and nuclear energy generation, and the French electricity grid dominated by nuclear energy generation. A comparative bar chart depicting S3's greenhouse gas emissions and ionizing radiation illustrates the shift in environmental burdens between these two categories (Figure 4). French electricity grid exhibits five times lower greenhouse gas emissions and three times higher ionizing radiation per kWh compared to the European electricity mix. This shift of environmental burdens toward higher impact in the ionizing radiation category arises from the reliance on nuclear energy in the French electricity grid.⁵¹ Regarding ECD's overall environmental impact, adopting the French electricity mix contributes to a 15-45% reduction in global warming potential across the assessed architectures (Table S3). In contrast, ionizing radiation impacts are two times higher than that where the European electricity mix was used. Moreover, relying on nuclear dominantly generated electricity results in a marginal decrease in toxicity-related impacts, with a more prominent decrease for S3 and S4, reaching 33% and 17%, respectively. These findings indicate the significant impact of the electricity source on ECDs' environmental performance, resulting in trade-offs between different environmental categories. LCIA results for the six ECD architectures relying on the French electricity mix are available in Table S3.

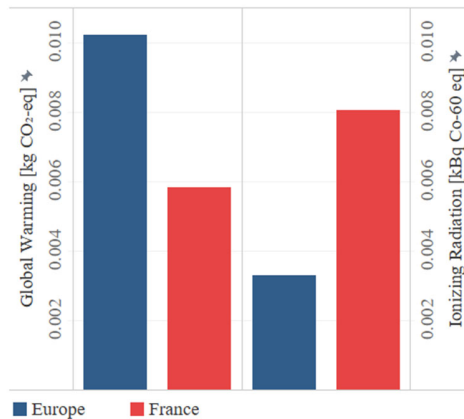


Figure 4. Global warming potential and ionizing radiation associated with ECD architecture S3 employing European or French electricity grids during the production stage.

LCIA Results: Endpoint Indicators

Expanding the environmental analysis to account for endpoint levels further supports the results obtained via the midpoint method. The endpoint environmental impacts confirm that S2, among analyzed ECD architectures, has the highest environmental impact, while S3 has the lowest environmental impact across all three areas of protection (Table S4). S2 resulted in an accumulated 3.9E-07 DALYs, 3.8E-10 Species.Year, and 5.0E-03 \$, in human health, ecosystem quality, and resources endpoint indicators, respectively. In contrast, S3 caused 5 to 16 times less environmental damage to the areas of protection. A notable finding is that S6, despite having higher ionizing radiation and water consumption impacts compared to S2 (Figure 3-h, 3-i), still causes around 80% less environmental damage than S2 (Figure 5). This discrepancy emphasizes the importance of considering endpoint levels in this analysis.

The influence of various midpoint indicators on the three areas of protection is examined within the considered ECD architectures. The human health endpoint level is predominantly influenced

by human toxicity midpoint indicators in architectures using silver as electrodes (Figure 5). This connection can be traced back to sulfide tailings generated during silver mining. Ecosystem quality is mainly impacted by global warming effects on terrestrial ecosystems, freshwater ecotoxicity, and freshwater eutrophication, accounting for over half of the damage caused. In terms of resources, the endpoint indicator is linked to two midpoint indicators: mineral and fossil resource scarcity. Notably, only fossil resource scarcity is relevant in the case of S3, S4, and S6, thanks to their silver-free design. Additionally, the contribution of ECD components on the endpoint level aligns with that of global warming (Figure S1). However, the contribution of both silver and ITO electrodes to the overall impact of the architectures is slightly higher than that observed at the midpoint level.

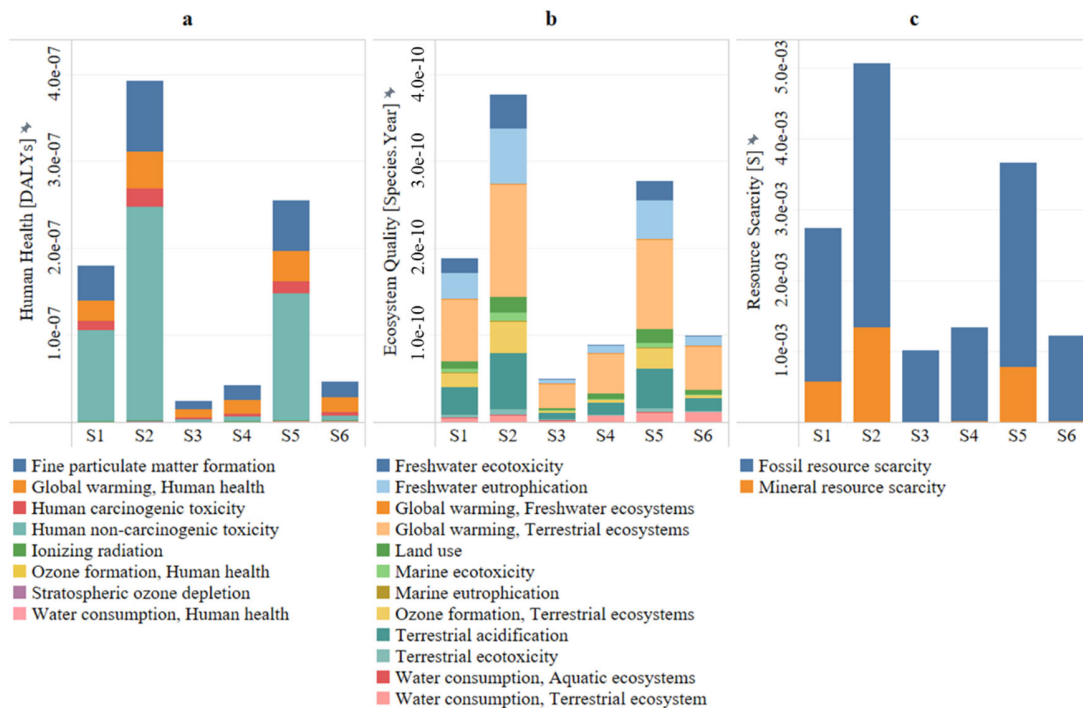


Figure 5. Endpoint environmental impacts comparison among assessed ECD architectures using a single display as reference flow according to ReCiPe Endpoint: a – human health, b – ecosystem quality, c – resource scarcity.

Recommendations Towards an Environmentally Sustainable ECD

The environmental assessment of ECD architectures, representing varying levels of technological maturity (Table 1), guides decision-makers towards either pursuing or abandoning their development. The objective of this section is threefold: firstly, summarizing the environmental impacts linked to the assessed architectures; secondly, providing recommendations for optimizing the environmental performance of ECD based on LCIA results; and finally, determining the environmental viability of pursuing further development for each ECD architecture.

Among ECD architectures, S3 all carbon design had the lowest impact across all midpoint and endpoint indicators (Figure 3 and Figure 5). Given the urgency of global warming as an environmental issue,⁵² it is imperative to prioritize technological solutions that minimize carbon emissions. In this light, S3 pilot-scale manufacturing should be further developed, capitalizing on its minimum carbon emissions. Insights from the other architectures can be adopted to further reduce S3's impacts. For instance, S6 compact size contributes to a 40% reduction in environmental impact attributed to its components, apart from the electrode, as the size of ITO electrodes stays the same. This suggests that reducing the size of S3 in a similar manner can potentially halve its overall environmental impact, as the contribution of the carbon-based electrodes is minimal.

S6, which employs an all-ITO architecture, exhibited the highest water consumption and the second-highest impact in terms of ionizing radiation (Figure 3-h), yet it demonstrated favorable performance in human health and ecosystem quality compared to S2 (Figure 5). Further research is warranted to optimize or substitute the ITO deposition process in S6 lab-scale processes, as it holds the potential to enhance its environmental performance beyond that of S3. Notably, the

majority of S6's impacts were attributed to the sputtering process and its reliance on argon gas, which means that exploring different deposition techniques may be environmentally rewarding. This may become technologically feasible if ink-jet printing of ITO is done on an industrial level.⁵³ An alternative solution to reduce S6's involves recirculating argon gas during the sputtering process or finding suitable substitutes. For example, nitrogen with around one-fifth the embodied carbon compared to Argon offers a potential substitute. Reports show positive results for the deposition of the ITO layer in a mixed Ar/N₂ or even a pure N₂ atmosphere.⁵⁴ Nonetheless, the implications of this substitution on ECD performance remains under investigation. It is worth mentioning that the argon gas quantity used in the LCI is based on literature, as S6 remains at the lab scale. Therefore, developing industrial processes for S6 may reveal specific optimizations leading to a significantly lower environmental impact.

S4 conceptual architectures demonstrated similar global warming potential and energy demand to S6 but caused 12% lower toxicity than S6. However, at the endpoint level, the environmental damage of S4 is quite similar to that of S6. The further development of S4 conceptual architectural design is of interest as it investigates combining silver-based electrode alternatives. S4 employs one sputtered ITO and one printed carbon layer exhibiting the same global warming potential as S6 but with lower toxicity-related impacts. Similarly, the impacts of S4 may be further reduced by optimizing or substituting the sputtering process.

Among ECD architectures that exploit silver are S1, S2, and S5. All these architectures have exhibited relatively high environmental impacts including the conceptual design of S5. S5 exhibits the second-highest environmental impact after S2. This is because S5 employs one printed silver electrode and one sputtered ITO electrode, which drives its environmental impacts relatively high. Consequently, further research and development of architectures employing silver electrodes,

including the conceptual design of S5, is recommended to be ceased, given the high environmental impact associated with silver usage and the limited potential for reducing this impact.

Several recommendations can be put forth to further reduce the environmental impacts of ECD during the manufacturing phase:

- Substituting silver for carbon-based or ITO electrodes, as demonstrated by S3, S4, and S6.
- Reducing the size of the ECD while maintaining the same functionality, as exemplified by S6.
- Recirculating, substituting, or reducing argon gas usage.
- Selecting the plant location where renewable or nuclear energy is available.

The findings of this LCA might be applicable to other electronic devices where silver, ITO, and carbon could be interchangeably used as electrodes.

The Methodological Approach: Perspectives and Considerations

A cradle-to-grave LCA predicts the environmental burdens associated with potential end-of-life scenarios for printed electronics. Fifty million tons of electronic waste was generated in 2019 and is rapidly growing, with a projected yearly increase of 3-4%, while recycling rates are estimated at 20-30%.⁵⁵ Low recycling rates and complex materials separation in printed electronics drive the adoption of biodegradable and/or renewable materials in ECD, such as paper and carbon.⁶ However, using precious and critical metals is imperative for enhanced performance and might be necessary for specific applications. Silver electrodes may be recovered through several hydrometallurgical routes,⁵⁶ thanks to the high demand for silver, making its recovery potentially economically viable.⁵⁷ Prior to commercialization and establishing logistical networks, many

uncertainties exist in ECD disposal practices. Printed electronics, especially ECDs, cannot be reused as they have a fixed and short lifetime, which leaves landfilling, incineration, or recycling as viable options.⁵⁸ Landfilled printed electronics may cause nanomaterials to leach, whereas its incineration produces toxic gases.⁵⁸ On the other hand, recycling is impeded by logistic issues, cost, and high-level integration of ECD.^{58,59} In a previous analysis, recycling paper-based anticounterfeiting labels utilizing ECD was explored; it was associated with paper waste streams. Hence, the paper substrate was recycled, and other components were landfilled.²⁴ Another LCA showed that a higher silver recycling rate could substantially improve the environmental performance of printed electronics.²¹ In ECDs utilizing precious silver or critical indium,⁶⁰ the recovery of these metals must supersede that of paper. Therefore, exploring scenarios that ease metals recovery is beneficial via cradle-to-grave LCA. In addition, it should be noted that impacts arising during ECD's use phase are expected to be minimal, considering the small amount of energy needed to operate the ECD over its lifetime.

Defining the functional unit in LCA requires careful consideration as it significantly affects the results and can easily alter the study's conclusion. In this study, the functional unit was defined as a single electrochromic display capable of communicating a predefined message clearly and promptly a fixed number of times. Thus, minimum values were fixed for relevant ECDs properties: optical contrast, switching time, and lifecycles. Fixing those values permitted the comparison of ECDs that have similar performance. Extending the comparison to include light emitting diode displays (LEDs), liquid crystal displays (LCDs), and ECDs exhibiting notably dissimilar characteristics obliges contemplating an alternative functional unit. However, when comparing with other display technologies it is imperative to select applications where the two displays compared can offer the same functionality. This is made possible by previous research identifying

applications where ECDs offer a viable substitute to currently used displays in clocks and other low-voltage electronics.⁵ Thus, it is possible to define a functional unit that accounts for variation in properties such as optical contrast, giving an environmental advantage to ECDs exhibiting superior ΔE^* and switching time. However, these properties are considered as positioning properties (i.e., attributes) that make an ECD more attractive to end users,³⁹ thus improving these properties beyond a certain threshold is not obligatory for ECD functionality. On the other hand, a display life cycle is an essential aspect from an environmental assessment point of view, as an extended life permits a prolonged use or reuse of the device. As a result, an alternative functional unit is proposed here that does not fix a minimum value for displays' life cycles but allows variables. The reference flow corresponding to the alternative functional unit can be calculated as in Equation 1. Incorporating these considerations into the functional unit facilitates an accurate and meaningful environmental comparison among other display technologies and ECDs of varying performance. Furthermore, it aids in reaching a more sustainable solution tailored to the application.

$$\text{Reference flow} = \frac{1}{\text{Lifecycles}} \quad \text{eq 1}$$

A sustainable ECD implies the consideration of its environmental impact, economic viability, and resource criticality. Therefore, integrating life cycle costing (LCC) and criticality assessment with LCA leads to a comprehensive understanding of an ECD architecture's sustainability. LCC examines an ECD's cost-effectiveness, considering raw materials, infrastructure, and disposal

costs.⁶¹ For instance, the cost of using carbon or ITO-based electrodes is evaluated beyond materials acquisition costs. It is extended to include equipment and integration costs arising from two different deposition methods, i.e., screen printing and sputtering. Furthermore, it is possible to evaluate the total costs attributed to a silver-based electrode throughout its life cycle and determine if it can compete with ITO or carbon-based, given a specific recycling rate. On the other hand, using indium, considered a critical material in ITO-based electrodes, motivates further criticality assessment. This is due to the limitation of the traditional LCIA method in assessing resource criticality⁶² and the emergence of several novel methods aiming to be integrated with LCA.^{63,64} Analyzing metal criticality during ECD production permits a meaningful comparison between all organic ECDs⁶⁵ and ECDs employing metals in terms of resource criticality and environmental performance.

Conclusion

This study evaluates six ECD architectures at varying technological maturity to identify environmentally viable research and development pathways. The environmental impacts of various materials and design choices were calculated by analyzing the six ECD architectures via ex-ante prospective LCA. The six ECD architectures, ranging from lab-scale prototypes to pilot-scale implementations, including conceptual designs, were scaled up to emulate industrial production. Accordingly, life cycle inventories were scaled up, and assumptions were made to optimize material flow and electricity consumption.

The LCIA results provided valuable insights into the environmental impact of materials utilized in ECD and helped to determine which ECD architectures are worthy of further development. For

instance, silver electrodes had a significant hotspot in the production of ECD, whereas carbon and ITO electrode substituents resulted in a lower environmental impact. While paper-based substrates are associated with lower environmental impacts than PET, their effect on the overall environmental performance of ECDs is negligible. In terms of the whole ECD architecture, S1, S2, and S5 are found to have the highest environmental impact due to its usage of silver electrodes, whereas S3, S4, and S6 architectures are identified as environmentally advantageous. S3 had a superior environmental performance compared to all other ECDs due to its use of carbon-based electrodes. On the other hand, S4 and S6 ranked second and third among ECD architectures in terms of environmental performance across all endpoint and midpoint indicators. Based on the environmental impact results of this LCA, S3, S4, and S6 were identified as ECD architectures that merit further development, particularly S6, which currently is at lab-scale level, as it has the potential to supersede S3 in terms of environmental performance. Additionally, it is recommended to cease research and development of ECD architectures employing silver electrodes, i.e., S1 and S2, including S5 conceptual design due to their high environmental impact and the limited potential for reduction. The environmental analysis of lab-scale and conceptual ECD architectures provided insightful eco-design recommendations for developing environmentally conscious solutions in the industrial production of ECD. One suggestion is to reduce the size of S3 while maintaining the same functionality, which can halve its environmental impact. Another suggestion is to replace the sputtering process of ITO or reuse the argon gas required during the sputtering process, which can significantly reduce the environmental impact of S6, as 90% of its impact stems from argon gas consumption.

The analysis of ECD architectures at the early development stage, especially conceptual designs, determines their environmental viability without the need for experimental testing, resulting in

significant economization of time and resources. The approach employed in this LCA provides guidance for scaling-up and predicting the environmental impacts of conceptual ECD architectures. This may benefit LCA practitioners and researchers engaged in ex-ante prospective LCA studies. Furthermore, the findings of this LCA might be applicable to other electronic devices where silver, ITO, and carbon could be interchangeably used as electrodes.

Nevertheless, steering ECD technology development toward sustainable directions requires a comprehensive approach to address economic viability, resource constraints, and environmental impacts throughout ECD's entire life cycle. In addition, LCC and novel criticality assessment methods may be conducted to evaluate ECDs' cost-effectiveness and resource criticality, respectively. These assessments can be integrated into LCA, thus guiding ECDs development toward a sustainable pathway. A comprehensive approach that merges results from these assessments aids in promoting sustainable practices in ECD's development.

Associated Content

Supporting Information

Life cycle inventory for 1 mask used in the screen-printing of ECD's layers: Table S1, Life cycle inventory for 1 cm² of indium-tin-oxide deposited: Table S2, Impact assessment results following the midpoint approach for the six ECD architectures considering 1 ECD as a reference flow and using French electricity mix during ECDs' production stage: Table S3, Impact assessment results following the endpoint approach for the six ECD architectures considering 1 ECD as a reference flow: Table S4, Comparison of assessed ECD architectures following ReCiPe Endpoint method featuring impacts of the three areas of protection, including an embedded contribution analysis

illustrating each component's relative environmental impacts. The environmental impacts are normalized for a single ECD as reference flow: Figure S1

AUTHOR INFORMATION

*A. Kamal Kamali - Univ. Bordeaux, CNRS, Bordeaux INP, ISM, UMR 5255, Groupe Analyse du Cycle de Vie et Chimie Durable (CyVi), 351 Cours de la Libération, F-33405 Talence, Cédex, France; E-mail: ahmad-kamal.kamali@u-bordeaux.fr

*Daye Lee - Univ. Bordeaux, CNRS, Bordeaux INP, ISM, UMR 5255, Groupe Analyse du Cycle de Vie et Chimie Durable (CyVi), 351 Cours de la Libération, F-33405 Talence, Cédex, France; E-mail: daye.lee@u-bordeaux.fr

Romain Futsch - Univ. Bordeaux, CNRS, Bordeaux INP, ICMCB, UMR 5026, 87 av du Dr Schweitzer, F-33600 Pessac, France

†Present Address: Luchrome, Elorprintec, B8, All. Geoffroy Saint-Hilaire, 33600 Pessac

Edis Glogic - Univ. Bordeaux, CNRS, Bordeaux INP, ISM, UMR 5255, Groupe Analyse du Cycle de Vie et Chimie Durable (CyVi), 351 Cours de la Libération, F-33405 Talence, Cédex, France.

*Aline Rougier - Univ. Bordeaux, CNRS, Bordeaux INP, ICMCB, UMR 5026, 87 av du Dr Schweitzer, F-33600 Pessac, France ; E-mail : aline.rougier@icmcb.cnrs.fr

Guido Sonnemann - Univ. Bordeaux, CNRS, Bordeaux INP, ISM, UMR 5255, Groupe Analyse du Cycle de Vie et Chimie Durable (CyVi), 351 Cours de la Libération, F-33405 Talence, Cédex, France.

Author Contributions

A. Kamal Kamali – Methodology, Data curation and analysis, Writing - Original draft

Daye Lee – Data curation and analysis, Writing - Original draft

Romain Futsch – Experiment, Collecting the data

Edis Glogic – Conceptualization, Collecting the data

Aline Rougier – Writing - review & editing

Guido Sonnemann – Writing - review & editing

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Notes

The authors declare no competing financial interest.

ABBREVIATIONS

ECD, Electrochromic Display; R2R, Roll-to-Roll; LCA, Life Cycle Assessment; TRL, Technology Readiness Level; PET, Polyethylene Terephthalate; ITO, Indium-Tin-Oxide; LCI, Life Cycle Inventory; LCIA, Life Cycle Impact Assessment; DALYs, Disability-Adjusted Life Years; CED, Cumulative Energy Demand; S2S, Sheet-to-Sheet; LCC, Life Cycle Costing;

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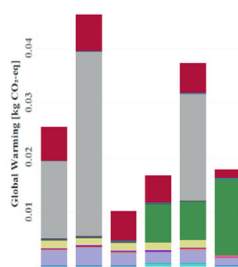
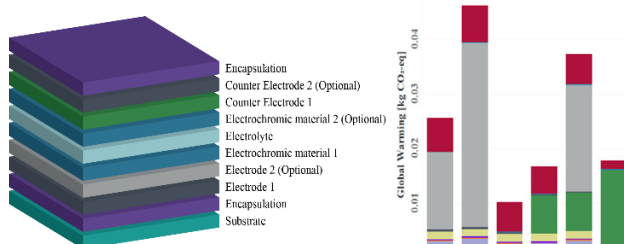
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The general structure of an electrochromic active-matrix displays along with the global warming potential of structures analyzed.