Cost, Energy and Emissions Assessment of Organic Polymer Light-Emitting Device Architectures

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Graphical abstract

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Abbreviation	Unit	Definition	Description
lm	-	lumen	SI unit of luminous power
cd	-	candela	SI unit of luminous intensity brightness
W	-	watt	SI unit of power
h	-	hour	Unit of time
d	-	day	Unit of time
a	-	year	Unit of time
kW h	-	kilowatt hour	Unit of energy
C_{dev}	\$/m ²	device cost per area	The total cost of materials and cost to manufacture a device architecture with an area of 1 m ²
C_{op}	\$/a	yearly operating cost	Cost to operate the device for 8 h/d over a period of 1 a
C_{lm}	\$/h	optical power cost	Cost to continuously generate 1000 cd/m ² for 1 h
$GHG-CO_{2,dev}$	kg	mass of CO ₂ emissions from device production	Mass of CO ₂ greenhouse gas emissions from raw materials extraction and manufacturing of a device with an area of 1 m ²
E_{mat}	MJ/kg	material embodied energy	Embodied energy of the raw materials in a device per mass
E_{man}	MJ/kg	direct process energy	Energy consumed during device manufacturing per mass
I_{CO_2}	kg/kWh	CO ₂ emission intensity	Average CO ₂ emission intensity
$GHG-CO_{2,op}$	kg/a	emission mass flow rate for CO ₂	Yearly CO ₂ emissions produced from a device with an area of 1 m ² operating for 8 h/d
C_{mat}	\$	cost of materials in the device	Cost of raw materials used for a device with an area of 1 m ²
C_{man}	\$	manufacturing cost	Cost to manufacture a device with an area of 1 m ²
C_{use}	\$	use phase cost	The cost to electrically power each device architecture
L	h	operational lifetime	The length of time it takes for device luminous efficacy to degrade by 50%
C_{elec}	\$(kW h)	cost of electricity in the USA	The cost of electricity in the USA per energy
E_{use}	kW h or GJ	use-phase energy consumption	The electrical energy needed to operate a device over its entire use phase
P_{in}	W	power in	Electrical power applied to the device
M_v	lm/m ²	luminous exitance	Light output power emitted by a device

B_f	lm/W	luminous efficacy	Electrical-to-optical power conversion efficiency
k	0.75	correction factor	Accounts for device operation at 75% of its initial luminous efficacy, on average, due to degradation and efficiency roll-off during its operational lifetime
B	cd/m ²	luminance	Used to quantify the brightness of a light-emitting device per area

Abstract

Proponents for sustainable alternative lighting and display options advocate for organic light-emitting diodes (OLEDs), particularly polymer-based organic light-emitting diodes (P-OLEDs), because of their potential for low-cost fabrication, more versatile device formats and lower power consumption compared to traditional options. Here, an economic, energy and CO₂ emissions assessment is carried out for four different laboratory-scale, blue-emitting P-OLED device architectures: bottom-emitting conventional; bottom-emitting inverted; top-emitting conventional; and top-emitting inverted. Additionally, comparisons with a standard, commercial-scale, blue inorganic light-emitting diode (LED) device architecture are made. The various P-OLED device architectures are investigated due to their potential to increase operational lifetime (inverted) and light out-coupling efficiency (top-emitting). The following metrics are used in this assessment: device cost per area, yearly operating cost, optical power cost, CO₂ emissions from device production, and yearly operating CO₂ emissions. We show that the top-emitting inverted device architecture significantly reduces the device cost per area, yearly operating cost, optical power cost and CO₂ emissions for the P-OLED devices, due to elimination of indium tin oxide and its comparatively high luminous efficacy and longer lifetime. In addition, the top-emitting inverted P-OLED device architecture performs competitively at the laboratory scale with commercial-scale inorganic LEDs for all metrics. However, if top-emitting P-OLEDs are to be manufactured on a large scale, the luminous efficacy assumed for laboratory-scale devices needs to remain constant throughout development to remain competitive.

1. Introduction

OLEDs are being investigated as sustainable alternative display and lighting options, as opposed to compact fluorescent lamps, incandescent lighting, and LEDs because of their low temperature growth conditions and potential earth-abundant constituent elements (while organic phosphorescent OLEDs usually contain rare-earth elements to harvest triplet excitons, fluorescent OLEDs have active layers that are primarily composed of carbon and hydrogen). However, current small-molecule OLEDs on the market are fabricated under high vacuum using thermal deposition, thus making the fabrication process expensive [1-9]. Therefore, low-cost, large-scale fabrication options are needed to make OLED technologies more marketable. P-OLEDs are an emerging sub-section of OLED technologies that are more amenable to solution-based processing which may enable more straight-forward, vacuum-free fabrication of the devices and, hence, lower cost and lower process energy consumption [2,10-13].

However, the lower efficiency (i.e., luminous efficacy) and the shorter operational lifetime of blue P-OLEDs compared to red and green P-OLEDs, slows the commercialization of a full-color P-OLED for general lighting and display purposes [2]. The lower efficiency of blue P-OLEDs can be attributed to the difficulty of charge injection into blue-emitting fluorescent polymers which have low highest-occupied molecular orbital (HOMO) energies (ca. -5.9 eV) and high lowest-unoccupied molecular orbital (LUMO) energies (ca. -2.1 eV) [14]. Common approaches that have been used to remedy these issues are incorporation of a high-work-function hole injection layer, such as poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), and a low-work-function electron transport layer, such as calcium (Ca) or lithium fluoride (LiF) into the device structure [15-17]. Additionally, while internal quantum efficiency can be optimal (~100% for phosphorescent OLEDs and phosphorescent P-OLEDs) [18], light-extraction efficiency is quite low (~20 % to 31%) [18-20], especially for blue-emitting devices and is a significant barrier to high-efficiency P-OLEDs. The shorter operational lifetime of conventional blue, fluorescent P-OLEDs is due to a combination of: (1) higher drive voltages due to the low light extraction efficiency; (2) corrosion of indium tin oxide (ITO) due to the

acidity of the PEDOT:PSS layer [3,21] and (3) degradation of PEDOT:PSS and Ca carrier injection properties due to exposure to water vapour and oxygen during device fabrication or operation [3].

Proposed approaches to tackling efficiency and operational lifetime issues are: (1) light management approaches, such as use of top-emitting device architectures, and addition of metallic or dielectric nanostructures to promote more efficient light extraction [22]; (2) inverted device architectures [23]; (3) improved encapsulation methods [24]. While these approaches have been shown to have the potential to increase device efficiency and operational lifetime [22-24], they may in turn affect the overall cost and environmental impact. Thus, further assessment of these approaches must be completed before any definitive conclusions can be drawn as to their usefulness. The goal of this study is to carry out an cost, energy and CO₂ emissions assessment, based on life-cycle assessment (LCA) methodologies, for four laboratory-scale, prototypical, fluorescent, blue-emitting P-OLED device architectures (conventional bottom-emitting, inverted bottom-emitting, conventional top-emitting and inverted top-emitting architectures) to determine which architecture is more effective in terms of device cost per area, yearly operating cost, optical power cost, energy consumption, and CO₂ emissions. Additionally, comparisons are made with the more ubiquitous, commercial-scale, blue inorganic LED device. As discussed earlier, the inverted P-OLED architectures increase the operational lifetime and the top-emitting P-OLED architectures increase the electrical-to-optical power conversion efficiency (i.e., luminous efficacy) [3,8,23].

Several economic and LCA studies have been conducted for LEDs and organic photovoltaic (OPV) devices which form the basis of our study for P-OLEDs [4-7,25-39]. A case study for the LCA of LED downlight luminaires concluded that the environmental impact of LEDs is dominated by the use-stage energy consumption and data gaps exist in LED product manufacturing and its environmental impacts; thus, resulting in a need for further research and assessments in order to compare LED-based luminaires with existing lighting technologies [28]. The U. S. Department of Energy has carried out detailed LCAs of energy and environmental impacts of LED lighting products, which show that the average life-cycle energy consumption is similar for both compact fluorescent lamps and LEDs, with it

being greater for incandescent lamps [7]. While economic, energy and environmental assessments have been completed for inorganic LEDs not much emphasis has been placed on the organic counterpart, which motivates this study. We draw comparisons with polymer-based OPV device economic and LCA studies, where applicable, since there have been a significant number of such studies [4-6,22,27-39] and, while OLEDs and OPV devices are operationally different, their device compositions and architectures are similar. As a result, one can study prior work on OPVs in order to draw inspiration to base future organic polymer-based cost and LCA studies due to the similarities in device structure and material type with the main difference being that polymer-based OPV devices produce electrical energy from sunlight while P-OLEDs consume electrical energy to produce light.

Furthermore, numerous OPV studies have focused on identifying approaches to lower device cost and the effects of increasing the efficiency and operational lifetime on energy and greenhouse gas (GHG) emission metrics that are also pertinent to P-OLEDs. For example, a life-cycle and cost assessment of OPV devices by Emmott et al. explored various transparent conductor alternatives to ITO in which they found that material alternatives, such as silver nanowires and high-conductivity PEDOT:PSS, have the potential to reduce the energy-payback time (EPBT) and financial cost of organic photovoltaic devices [4]. Espinosa et al. conducted a LCA of organic tandem solar cells where they investigated the economic and environmental feasibility of manufacturing a tandem solar cell versus a single junction solar cell. They found that the tandem solar cell has to be 20% better performing than a single-junction device in order to improve cost and sustainability metrics [27]. A review paper by Lizin et al. of LCA studies of OPVs focused on environmental aspects such as cumulative energy demand (CED), EPBT, and the GHG emission factor of single-junction, organic, bulk-heterojunction P3HT:PC₆₀BM polymer-based solar cells [5]. The top environmentally performing solar cell had a CED of 37.58 MJ/m², EPBT from 3.54 months to 6.24 months, and cell efficiency of 2% with lower GHG emission factors than current power plants. They concluded that the often-used linear relationship between increasing operational lifetime or efficiency and improved sustainability, CED and EPBT, is not a sufficient model because improvements in these areas are heavily dependent on the device materials and architectures [5]. Darling et al. conducted

a LCA to estimate the CO₂ emission factor for a OPV device with an area of 1 m², with 1% solar power conversion efficiency (PCE) and one year operational lifetime (which are achievable today), and a hypothetical future OPV device with 15% PCE and a 20 year operational lifetime [6]. They estimated a ~10% decrease in CO₂ emissions due to the increase in PCE and the longer operational lifetime. Furthermore, they suggest that improvements can be made to operational lifetime through encapsulation with materials with low water and oxygen transport rates and use of air-stable alternative materials. Additionally, in order to transfer OPV technology from laboratory-scale to larger scales for commercialization, efficiency, scalability of manufacturing processes, and knowledge of degradation mechanisms and their impacts on operational lifetime are critical factors that have been identified through economic and LCA studies [5,6,29-39].

The aforementioned OPV studies allow us to draw some conclusions that are applicable to P-OLED devices, such as: eliminating ITO from the device architectures, increasing multilayer device performance, and use of stable device materials and encapsulants should assist in making P-OLED performance more comparable with the performance of current LEDs on the market. However, comparisons between certain aspects such as life-cycle CO₂ emissions and cost assessments of photovoltaic devices and light-emitting devices are not appropriate or straight forward. For example, once a photovoltaic system is installed, the main yearly cost is associated with system maintenance, while, for a lighting system there are significant additional costs because it consumes electricity during operation (as opposed to generating electricity from a free natural resource, i.e. the sun, as in the case of a photovoltaic system). Therefore, in our study we develop some alternative assessment methodologies and metrics that are relevant to P-OLED devices but not to OPV devices.

2. Methodology

2.1 Goal and Scope

As mentioned in Section 1, the goal of this study is to carry out a cost, energy and CO₂ greenhouse gas (GHG-CO₂) emissions assessment, based on LCA methodologies, for four laboratory-scale, prototypical, fluorescent, blue-emitting P-OLED device architectures to determine which architecture is more effective in terms of device cost per area, yearly operating cost, optical power cost, energy consumption, and CO₂ emissions. LCA is used as a tool to assess the energy and environmental impacts of a product, process or activity throughout its life cycle; from the extraction of raw materials through to processing, transport, use and disposal [4,5,7,25-42]. LCA is a standard international ISO 14040 series method that consists of four distinct components: (1) goal and scope, (2) inventory analysis, (3) impact assessment, and (4) interpretation. First, the aim of the study, central assumptions, and system boundaries are chosen. Next, during the life-cycle inventory analysis (LCI) phase, the inputs and outputs for the emissions and resources are quantified. Then a life cycle impact assessment (LCIA) is conducted to evaluate the potential environmental impact of the previous quantified values. Finally, an interpretation of results are presented in a clear concise manner[27,41,42].

We complete a **cradle-to-grave assessment**, i.e., from the inception of raw materials to the end of use, of a fluorescent, blue-emitting P-OLED device (i.e., one that uses, for example, a polyfluorene-based light-emitting active layer) as there are numerous results reported in the literature for such devices [1,3,8-14]. As discussed in Section 1, while photovoltaic studies can be used as a guide to base LCA studies of **light-emitting devices**, they differ in terms of their operation. Unlike OPVs, P-OLEDs consume electrical power for operation and produce optical power (i.e., light). As a result, the functional unit should be determined by the basis of the optical power produced, which in our case we are assuming to be the brightness per area or luminance. Therefore, here, we employ a luminance of 1000 cd/m² as our functional unit which is a commonly reported luminance for OLEDs [2,24,43-50]. Each device architecture is assumed to produce this constant brightness, and in order for this to be achieved either the electrical input power or the power efficiency (i.e., luminous efficacy) of the device can be varied. Note that in contrast, for OPV devices a constant optical input power (or irradiance) is applied during performance testing (i.e., 100 mW/cm² (1 sun)); therefore, to generate a particular target electrical power quantity,

device area or efficiency is varied. We determine the following metrics: the device cost per area, including materials and manufacturing costs; yearly operational cost; cost to continuously generate 1000 cd/m² for one hour; and the GHG-CO₂ emissions, for both device production and yearly operation, from the point of view of the user. The assessment carried out here is meant to provide perspective on the cost, energy and emissions impact of blue-emitting P-OLEDs relative to the more mature inorganic semiconductor LED technologies. Additionally, the P-OLED life-cycle stages and materials which are expected to have the greatest cost and emissions impacts are identified.

2.2 Central Assumptions

In this study, we completed a cradle-to-grave assessment including the following stages (Fig. 1): (1) raw materials extraction and production; (2) PLED device fabrication and (3) PLED device use. The following inputs and outputs are considered for each stage where relevant: material inputs; electrical energy inputs; GHG-CO₂ emissions outputs and optical energy output, and P-OLED device use stages. A life-cycle inventory is compiled and analyzed for the materials, production and fabrication, and use-phase operating cost of the PLEDs in order to carry out the assessment. In our assessment, we ignore all transport, installation, and disposal phase costs associated with the life-cycle of the P-OLED because these costs are assumed to be small compared to device and use-phase costs [4, 31, 40]. Furthermore, we have not included in the assessment the housing, electrical connections, heat sinks, or other items involved in the mounting of the P-OLED as it is assumed to be similar amongst the different P-OLED architectures regardless of the final product (e.g., lighting, display) because they are all planar, thin-film surface-emitting optoelectronic devices and our functional unit (i.e., 1000 cd/m²) is the same for each architecture. Conversely, for the inorganic LED different housing, mounting and peripheral components (e.g., electrical connections, heat sinks) could certainly be employed. However, given that the blue inorganic LED is the most ubiquitous blue light-emitting device, it is useful as a standard against which blue OLEDs can be compared (similar to how a silicon solar cell is the standard against which all newer solar cell technologies are compared regardless of eventual differences in mounting, housing, etc. [6]).

Fig. 1. System boundary diagram for a LCA of OLEDs. In this study, our assessment considers only the stages of the life cycle highlighted in by the dashed red boxes: raw materials extraction and production, OLED device fabrication, and OLED device use.

Our background system (i.e., the information needed to carry out this study) is defined through extensive reviews of published literature and supplier catalogues. Embodied energy and direct process energy values were obtained from published literature that included relevant embodied energy data from LCA databases such as Ecoinvent and Gabi for the more common materials (e.g., glass, silver). However, in some cases, material embodied energy values were assumed values based on more commonly-available materials within the same material class. For example, material embodied energies for poly(3-hexylthiophene) were used instead of those for poly(9,9-dioctylfluorene)), PFO, for the organic conjugated polymer active layer due to lack of available embodied energy data for PFO. This is a reasonable approach because both P3HT and PFO are conjugated (i.e., semiconducting) polymers that have rigid

molecular backbones that consist of molecular monomers or long chains of carbon-based repeat units connected by covalent bonds. In addition, both polymers are synthesized from a solution in a similar catalytic fashion [51,52]. The cost per mass data for materials were obtained from supplier online catalogues and from published literature. References to the source websites (including date accessed) and the relevant papers are included at the appropriate location for each cost per mass value. For the thickness of layers in the different P-OLED devices (Tables 1 and 2), information was obtained from published literature in which device prototypes were fabricated and tested, as well as Department of Energy solid-state lighting technical reports (referenced below).

The performance parameters (Table 3) are essential to calculating the metrics defined in Section 2.3 used for the assessment of the different device architectures, which comprise our foreground system. Note that only lab-scale and pilot-scale fluorescent blue polymer OLED devices have been reported to date. Therefore, all of our metrics for the OLED devices are for lab/pilot-scale devices. First, the average operational lifetimes of conventional blue fluorescent OLEDs were determined from references [2,43,44]. Then, based on publications in which direct comparisons between the lifetime of a conventional and inverted OLED were made [45,46], a scaling factor was determined. From this assessment we found that the inverted OLEDs have operational lifetimes that are 1.75 times longer than the conventional OLEDs. We then multiplied the average conventional operational P-OLED lifetime by the scaling factor to determine the inverted OLED operational lifetimes for both bottom and top emitting device structures.

The luminous efficacy values were calculated in a similar fashion to the operational lifetime. To determine the luminous efficacy values for the different device architectures, first, we averaged the luminous efficacy values for conventional bottom emitting fluorescent blue OLEDs from references [10,44,52] at a luminance of 1000 cd/m². Then, in a similar way to how the operational lifetime of the inverted devices was determined, a scaling factor was taken from reports that directly compared inverted to conventional bottom emitting OLEDs [45,54-57], top emitting to bottom emitting conventional OLEDs [58], and the top emitting to bottom emitting inverted OLEDs [56,59-61]. We then used these scaling factors to calculate the efficacy values from the averaged conventional bottom emitting OLED luminous

efficacy for each device configuration. Note that the luminous efficacy value of the inorganic LED was based on reported values for mass-produced inorganic LEDs that included light extraction structures and housing [48,50,62-65], which may aid in increasing efficacy values.

2.3 Definition of Metrics

Our metrics for this assessment are as follows: (1) device cost per area, C_{dev} , which is the upfront cost to the user at the initial purchase; (2) yearly operating cost, C_{op} , which is the cost to operate the device for 8 h/d over a period of one year; (3) optical power cost, C_{lm} , which is the cost required to generate the functional unit of 1000 cd/m² of optical power for one hour; (4) GHG-CO₂ emissions from raw materials extraction and device manufacturing, $GHG-CO_{2,dev}$; (5) yearly GHG-CO₂ emissions produced from a device operating for 8 h/d, $GHG-CO_{2,op}$. We define C_{dev} as:

$$C_{dev} = C_{mat} + C_{man} \quad (1)$$

where C_{mat} is the materials cost for all device layers and C_{man} is the manufacturing cost. Estimation of C_{mat} for each device architecture studied here will be discussed in the next section using information obtained from materials suppliers and is the largest contribution to C_{dev} .

C_{man} is determined from a percentage range of the total device costs reported for solution-processed OPV device manufacturing costs on the lab/pilot scale, i.e., 21 % to 40 % [6,27,29-51]. Note due to the current state of P-OLED development, large-scale manufacturing methods and practices are currently not optimized or standardized. Additionally, comparisons between manufacturing costs for reel-to-reel processed devices and manufacturing costs for devices fabricated on ridged substrates have shown only a slight increase in the percentage contribution of manufacturing costs to total device costs (~50 % is an upper estimate for OPV devices on glass substrates compared to 21 % to 40 % for OPV on PET substrates). Therefore, we have assumed an average percent contribution of manufacturing costs to total device costs of 30% [6,27,29-51]. Therefore, to determine C_{man} , such that C_{man} contributes to 30% of

C_{dev} , C_{man} was taken to be a 43% of C_{mat} . We do not account for possible differences in manufacturing costs between lighting and display P-OLED technologies such as types of capital equipment (e.g., spray coaters versus ink-jet printers). However, in both cases the active layer, hole transport layer (HTL) and ETL are assumed to be fully solution processed on glass [11,12]. Additionally, as stated earlier, this study focuses on device costs per area; costs associated with (P-O)LED housing, electrical connections, heat sinks and electronic drivers (i.e., balance of system costs) are not included.

We define C_{op} as:

$$C_{op} = \frac{C_{use}}{L} * 8 * 365 \quad (2)$$

where L is the operational lifetime of the device and C_{use} is the use-phase cost. L is taken to be the time it takes for the luminous efficacy to drop to 50% of its initial value[68]. C_{use} is the cost of operation for a device (with area of 1 m²) over the device's operational lifetime and is defined as:

$$C_{use} = C_{elec} * E_{use} \quad (3)$$

where C_{elec} is taken to be the cost of electricity in the United States (assumed to be 0.0984 \$/(kW h) [69]), and E_{use} is the use-phase energy consumption defined as:

$$E_{use} = \frac{P_{in} * L}{1000} \quad (4)$$

where P_{in} is operating electrical power for a device with an area of 1 m². P_{in} is calculated as follows:

$$P_{in} = \frac{M_v}{B_f} k \quad (5)$$

where B_f is the luminous efficacy and k is a correction factor, which accounts for the performance degradation of the device over time. Here, we assume a constant applied voltage is applied to each device, therefore, $k = 0.75$ (i.e., on average, the device operates at 75% of its initial luminous efficacy over its operational lifetime, L) [68]. M_v is the luminous exitance (i.e., the light output power) and is defined as

$$M_v = B * \pi \quad (6)$$

where B is luminance which we have taken to be 1000 cd/m^2 , as discussed earlier in Section 2.1, as it is the standard value used when reporting operational lifetimes of OLED devices [2,24,41-50]. We assume C_{lm} in $\$/h$ is then calculated as:

$$C_{lm} = \frac{M_p * C_{elec}}{B_f * 1000} \quad (7)$$

Next, CO_2 emissions from device production, $GHG\text{-CO}_{2,dev}$, which includes the $GHG\text{-CO}_2$ emissions from raw materials and device fabrication in kg of CO_2 is defined as:

$$GHG\text{-CO}_{2,dev} = (E_{mat} + E_{man}) * I_{CO_2} \quad (8)$$

where E_{mat} , in MJ/m^2 , is the embodied energy of the raw materials in the devices and E_{man} , in MJ/m^2 , is the direct process energy consumed during device manufacturing. E_{man} , which is taken to be 1.05 times E_{mat} , is determined from averaged ratios of direct process energy to embodied energy in the material from relevant OPV literature [27,29-31]. The average CO_2 emission intensity from fossil fuels, I_{CO_2} , between 1997 and 2012 for the United States from electricity generation is taken to be $1.90 \text{ kg/(kW h) of CO}_2$ (equal to $0.53 \text{ kg/MJ of CO}_2$) [70]. Finally, the emission mass flow rate for CO_2 , $GHG\text{-CO}_{2,op}$, which is the yearly $GHG\text{-CO}_2$ emission produced from a device (area of 1 m^2) operating 8 h/d (in kg/a of CO_2) is defined as:

$$GHG\text{-CO}_{2,op} = \frac{(E_{use} * I_{CO_2} * 8 * 365)}{L} \quad (9)$$

3. Life-Cycle Inventory - Device Architectures and Materials

The bottom-emitting conventional P-OLED was analysed initially as a foundation with which to compare the bottom-emitting inverted, the top-emitting conventional and the top-emitting inverted P-OLED architectures, and the blue inorganic LED. Schematics of the different P-OLED architectures and the inorganic LED are shown in Fig. 2a-e.

Fig. 2. Schematics of: (a) a bottom-emitting conventional P-OLED, (b) a bottom-emitting inverted P-OLED, (c) a top-emitting conventional P-OLED, (d) a top-emitting inverted P-OLED, and (e) a blue inorganic LED. The blue arrows with $h\nu$ (photon energy) labels represent the direction of light emission.

The inventory of materials and average layer thicknesses for each device architecture, obtained from published literature, is included in Table 1.

Table 1. Table of bottom-emitting (conventional and inverted) and top-emitting (conventional and inverted) P-OLED device layer materials, layer thicknesses and corresponding layer functions. The thickness values are an average of values reported in the corresponding referenced literature with the standard deviation shown after the average value. Glass substrates with thickness of 0.192 mm were assumed for all P-OLED device architectures.

Device Architecture	Thickness (nm)	Function	References
Bottom-Emitting Conventional			
ITO	108 ± 50	Anode	[4,22,50]
PEDOT:PSS	57 ± 13	HTL	[4,22,52,71-75]
PFO	108 ± 45	active layer	[4,22,50,72-76]
LiF	2.3 ± 2	ETL	[4,71,76]
Al	142 ± 53	Cathode	[4,22,50,73,74,76]
Bottom-Emitting Inverted			
ITO	108 ± 50	Cathode	[4,22,50]
TiO ₂	45 ± 40	ETL	[77,78]
PFO	108 ± 45	active layer	[4,22,50,72-76]
MoO ₃	11 ± 7	HTL	[8,50,77-84]
Ag	35 ± 30	Anode	[82-84]
Top-Emitting Conventional			

Ag	125 ± 29	Anode	[8,58,85]
PEDOT:PSS	57 ± 13	HTL	[4,22,52,71-75]
PFO	108 ± 45	active layer	[4,22,50,72-76]
LiF	2.3 ± 2	ETL	[4,71,76]
Al	2	Cathode	[85]
Ag	17.8 ± 2.17	Cathode	[8,85]
Top-Emitting Inverted			
Ag	125 ± 29	Cathode	[8,58,85]
TiO ₂	45 ± 40	ETL	[77,78]
PFO	108 ± 45	active layer	[4,22,50,72-76]
MoO ₃	11 ± 7	HTL	[8,50,77-84]
Ag	17.8 ± 2.17	Anode	[8,85]
Blue Inorganic LED			
sapphire	10 ⁶	substrate	[86,87]
undoped GaN	600	buffer layer	[86,87]
n-doped GaN	1500	ETL	[86,87]
p-doped GaN	500	HTL	[86,87]
InGaN	200	emitter layer	[86,87]

4. Cost Assessment

4.1 Device Cost Per Area

To determine the C_{dev} , we first carried out materials cost calculations to determine C_{mat} using the mass per area for each layer of the device and material cost per mass as shown in Table 2. Using the data obtained from Table 2, the estimated cost for each layer per area (C_A) in a device was calculated as follows:

$$C_A = m * C_m \quad (10)$$

where m is the mass per area and C_m is the cost per mass of the material in each layer of the device.

Table 2. The mass of each layer for a device (area of 1 m²), the material cost per mass values and the cost per layer in a particular device are represented for bottom-emitting conventional P-OLED, bottom-emitting inverted P-OLED, top-emitting conventional P-OLED, top-emitting inverted P-OLED, and blue inorganic LED architectures [88-97]. Only the materials that are used in a particular device architecture are represented in the respective column. The cost per layer for the encapsulant is a generic value taken from Ref. [32]. Each material layer function is represented by the following superscript characters: ^ substrate, # anode, * HTL, + active layer, ~ ETL, * cathode.

Materials	Mass per area (g/m ²)	Cost per mass	Cost per layer (\$/m ²)
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	(\$/g)		Bottom-Emitting P-OLED		Top-Emitting P-OLED		Inorganic LED
			Conventional	Inverted	Conventional	Inverted	
Glass [^]	474.24	0.09	42.92	42.92	42.92	42.92	-
ITO	0.73 ± 0.34	114	83.68 ± 38.53 [#]	83.68 ± 38.53 [*]	-	-	-
Al [*] (142 nm)	0.38 ± 0.14	0.24	0.09 ± 0.03	-	-	-	-
Al [*] (2 nm)	0.005	0.24	-	-	0.001	-	-
Ag [#] (35 nm)	0.37 ± 0.32	6.39	-	2.35 ± 2.01	-	-	-
Ag (17.8 nm)	0.19 ± 0.02	6.39	-	-	1.19 ± 0.15 [*]	1.19 ± 0.15 [#]	-
Ag (125 nm)	1.31 ± 0.30	6.39	-	-	8.38 ± 1.94 [#]	8.38 ± 1.94 [*]	-
PEDOT:PSS [×]	0.06 ± 0.01	9.02	0.52 ± 0.12	-	0.52 ± 0.12	-	-
MoO ₃ [×]	0.05 ± 0.03	10.84	-	0.57 ± 0.36	-	0.57 ± 0.36	-
PFO ⁺	0.11 ± 0.05	391	43.79 ± 18.38	43.79 ± 18.38	43.79 ± 18.38	43.79 ± 18.38	-
LiF [~]	0.01 ± 0.005	31.30	0.19 ± 0.16	-	0.19 ± 0.16	-	-
TiO ₂ [~]	0.19 ± 0.17	3.16	-	0.59 ± 0.53	-	0.59 ± 0.53	-
Sapphire [^]	398	0.52	-	-	-	-	206.96
GaN	15.99	17.55	-	-	-	-	280.62
GaN ⁺	0.86	17.55	-	-	-	-	15.11
InN ⁺	0.41	188	-	-	-	-	76.89
Encapsulant	-	-	11.49	11.49	11.49	11.49	11.49
<i>C_{mat}</i> (\$/m ²)	-	-	182.68 ± 57.22	185.39 ± 59.81	108.48 ± 20.75	108.93 ± 21.36	591.07

By adding the estimated price for each layer (calculated using Equation 10) we determined the estimated C_{mat} of a device with an area of 1 m² (excluding manufacturing costs) for each architecture: 183\$/m² ± 57\$/m (bottom-emitting conventional); 185\$/m ± 60\$/m (bottom-emitting inverted); 108\$/m ± 21\$/m (top-emitting conventional); 109\$/m ± 21\$/m (top-emitting inverted); and 591\$/m (inorganic blue LED). In this way the contribution of each layer to the overall C_{mat} amount for each device could be examined.

Fig.3. The percentage cost for each layer in the: (a) bottom-emitting conventional P-OLED; (b) bottom-emitting inverted P-OLED; (c) top-emitting conventional P-OLED; and (d) top-emitting inverted P-OLED. The materials cost, C_{mat} , for each device (area of 1 m²) is shown above the corresponding pie chart (i.e., manufacturing costs, C_{mat} , not included).

As shown in Figure 3 the ITO, PFO and glass layers contributed the most to C_{mat} for bottom-emitting P-OLEDs (approximately 45%, 24% and 23%, respectively) and the PFO and glass layers contributed the most to C_{mat} for top-emitting P-OLEDs (both ~40%). Despite layer thicknesses of only ~100 nm for both ITO and PFO (Table 1) they were the most expensive materials, per mass, hence the significant percentage contribution to C_{dev} . Conversely, glass was one of the cheapest materials per mass; however, it was also the thickest layer (0.192 mm) which resulted in the significant overall cost per area. The metal layers (Ag and Al) accounted for less than 9% of the total cost of the materials in the devices. The ETL and HTL layers were negligible in cost compared to the other layers. As a result, there was very little change in cost on going from a conventional to an inverted device architecture. However, since the top-emitting architectures eliminated ITO, the value of C_{mat} was reduced by ~41% compared to the bottom-emitting devices. C_{mat} is 591 \$/m² for the blue inorganic LED device architecture (1 m² device), with C_{mat} calculated in a similar fashion to the P-OLED architectures (see Fig. 2 and Table 1). Therefore, C_{mat} for the blue inorganic LED was 5.4 times more than that of the top-emitting P-OLED architecture. This makes the top-emitting architecture a viable option in terms of C_{mat} for solid-state lighting or display applications. C_{dev} , which included a material cost, C_{man} , that was calculated as a percentage of C_{mat} (43%) such that C_{man} contributed to 30% of C_{dev} (see Section 2.3), was then determined and the assessment of C_{dev} is included in Section 4 below.

4.2 Use-Phase Cost

We now determine the performance data (operational lifetime and luminous efficacy) obtained from the literature for each device architecture (Table 3) and how much it would cost to electrically power each architecture in the United States (cost of electricity of 0.0984 \$/(kW h) [69]) over the useful life of each device, i.e., the use-phase cost (C_{phase}) using Equations 3-6.

Table 3. The operational lifetime, L , and luminous efficacy, B_f , are represented for the bottom-emitting conventional P-OLED, bottom-emitting inverted P-OLED, top-emitting conventional P-OLED, top-emitting inverted P-OLED and blue inorganic LED.

Performance Parameters	Bottom-Emitting P-OLED		Top-Emitting P-OLED		Inorganic LED	Sources
	Conventional	Inverted	Conventional	Inverted		
L (h)	16000	28000	16000	28000	50000	[2,24,41-46,50]
B_l (lm/W)	2.5	3.8	5.1	7.8	7.5	[10,44-65]

As shown in Table 3 the inverted P-OLED architecture has an approximately 75% longer operational lifetime than the conventional P-OLED architecture (9.6 years compared to 5.5 years, assuming the P-OLED device operated for 8h/d). The top-emitting inverted device is the most energy efficient of all of the devices (luminous efficacy of 7.8 lm/W), and consumes at least 1.5 times less power than the other P-OLED architectures during operation. However, the blue inorganic LED has a factor of 1.8 longer operational lifetime than the longest operating P-OLED (17.1 years for the inorganic LED). Using the operational lifetime and luminous efficacy values reported in Table 3 along with Equations 3-6, the use-phase cost (C_{phase}) for each architecture was determined; see Figure 4.

Fig. 4. The corresponding C_{use} and C_{op} values for each P-OLED device architecture and a blue inorganic LED.

We find that the bottom-emitting inverted P-OLED device had the highest use-phase cost (1717\$;

see Fig.4) of all of the devices and the top-emitting conventional device had the lowest use-phase cost of the P-OLED devices (2.4 times smaller than the bottom-emitting inverted architecture) primarily due to its higher luminous efficacy and shorter operational lifetime (Table 3). The use-phase cost of the blue inorganic LED was ~2.1 times greater than the top-emitting conventional P-OLED (729\$ and 1537\$, respectively) primarily due to its longer operational lifetime (50000 h). When the use-phase cost is normalized by the device operational lifetime, we obtain C_{op} , the yearly operating cost, for each device (Fig. 4). The top-emitting inverted P-OLED had the lowest operating cost of all P-OLEDs (261\$/a) and cost 8\$/a lower than the blue inorganic LED (269\$/a) due to the higher luminous efficacy of the former (7.8 lm/W).

4.3 Economic Impact

The following metrics are presented in Table 4 for light-emitting devices with areas of 1 m²: C_{dev} , C_{op} and C_{lm} . C_{dev} (i.e., including materials and manufacturing costs) was the lowest for the top-emitting P-OLEDs and was approximately 5 times cheaper than the blue inorganic LED.

Table 4. The metrics C_{dev} , C_{op} and C_{lm} , for each P-OLED device architecture and the blue inorganic LED (device areas are 1 m² in all cases).

Device Architectures	C_{dev} (\$/m ²)	C_{op} (\$/a)	C_{lm} (\$/h)
Bottom-emitting Conventional P-OLED	261 ± 82	267	0.12
Bottom-emitting Inverted P-OLED	265 ± 85	179	0.08
Top-emitting conventional P-OLED	155 ± 30	133	0.06
Top-emitting Inverted P-OLED	156 ± 31	87	0.04
Inorganic LED	844	90	0.04

This indicates that the top-emitting P-OLEDs are the most attractive device type in terms of up-front costs to the user. The top-emitting inverted P-OLED had the lowest C_{op} of all of the devices due to its high luminous efficacy (Fig.4), while the bottom-emitting conventional P-OLED had the highest C_{op} . For C_{lm} , the

top-emitting conventional architecture (0.04\$/lm) was the best performing of the P-OLEDs and cost the same as the blue inorganic LED. In short, the top-emitting inverted P-OLED is the most promising P-OLED device architecture in terms of total cost because: (1) it eliminates one of the most expensive layers (ITO) in the device composition; and (2) it has high luminous efficacy in comparison to the other P-OLED device architectures. Furthermore, while the top-emitting P-OLED architecture has a slight advantage over the blue inorganic LED in terms of C_{op} , it is significantly cheaper in terms of C_{dev} (~5 times cheaper). Therefore, even considering the longer lifetime of the inorganic LED (1.8 times longer), the top-emitting P-OLED would still have a lower total cost when factoring in lifetime and replacement device costs (neglecting P-OLED housing and light extraction structures).

5. Energy and CO₂ Emissions Assessment

5.1 Device Embodied Energy

Powering light-emitting optoelectronic devices is tied to GHG emissions through the indirect production of CO₂ during electricity consumption. Furthermore, GHG emission is linked to the use-phase energy of the P-OLED devices, which is expected to be the most significant energy-consumption stage of the P-OLED life-cycle. As referenced in the Department of Energy study on lighting technology it has been shown that the use phase is the largest contributor to the overall energy consumption of such devices as fluorescent, incandescent, and LED lamps [7]. To illustrate this point, we determined the embodied energy, E_{mat} , in P-OLED devices (areas of 1 m²) using literature values for the embodied energy of each constituent layer material and the mass of each layer of the device (Table 5).

Table 5 Embodied energy in MJ/kg of materials in P-OLED devices.

Materials	Embodied Energy (MJ/kg)	Source
ITO	355753	[30,31]
PEDOT:PSS	131	[30,33]
PFO*	1843	[30,33]
LiCl [#]	220	[103]

Al	171	[25]
Ag	128	[104]
MoO ₃	80	[33]
TiO ₂	118	[33,104,105]
Glass	16	[25]
Encapsulant (1 m ²)	10 [‡]	[31]

*Data for P3HT used here as an approximation for PFO (PFO embodied energies not available), [#]LiCl used here instead of LiF as embodied energy data was limited for LiF; [‡]value is in units of MJ/m²

As can be seen in Table 5, ITO and PFO have the highest embodied energies 355753 MJ/kg and 1843 MJ/kg, respectively. We then accounted for the mass of each layer in each P-OLED architecture (Table 6).

Table 6. Embodied energy (in MJ) from raw material extraction per layer of material in P-OLED devices with areas of 1 m² (direct layer process energy not included).

Layer	Bottom-Emitting P-OLED		Top-Emitting P-OLED	
	Conventional	Inverted	Conventional	Inverted
ITO	45.89	45.89	-	-
PEDOT:PSS	0.008	-	0.008	-
PFO	0.207	0.207	0.207	0.207
LiCl [#]	0.001	-	0.001	-
Al	0.078	-	-	-
Ag	-	0.047	0.192	0.192
MoO ₃	-	0.004	-	0.004
TiO ₂	-	0.022	-	0.022
Glass	39.52	39.52	39.52	39.52
Encapsulant	9.96	9.96	9.96	9.96
Total(<i>E_{mat}</i>)	95.67	95.65	49.90	49.93

The resulting embodied energies for P-OLED devices (areas of 1 m²) were ~96 MJ and ~50 MJ for the bottom-emitting and top-emitting P-OLEDs, respectively. The major contribution to the larger bottom-emitting device embodied energy was ITO, making up approximately 48% of the embodied energy. The other layers that exhibited significant embodied energies were the glass and encapsulant layers (~39 MJ and 10 MJ, respectively); however, these were still significantly smaller than the ITO embodied energy (which was almost 46 MJ). Additionally, the embodied energies of all P-OLED devices were approximately an order of magnitude smaller than the embodied energy estimated for the

inorganicLED(4650 MJ from Ref. 25). However, the total device embodied energy(E_{mat}) for each P-OLED architecture was substantially lower than the use-phase energy(E_{use} ; converted to GJ by multiplying its value in kW h by 0.0036) which ranged from 53.4 - 62.8GJ for the bottom-emitting P-OLEDs, was a value of 26.7 GJ for the top-emitting conventional P-OLED, and was 30.5 GJ and 56.2GJ for the top-emitting inverted P-OLED and the blue inorganic LED, respectively. Therefore, lowering the use-phase energy should have the greatest effect on reducing environmental impacts caused by energy consumption during operation of the P-OLEDs.

5.2. GHG-Carbon Footprint

The CO_2 emissions from device production, $GHG\text{-CO}_{2,dev}$, were calculated for the four different architectures and the blue inorganic LED using Equation 8 and the embodied energies for raw material extractions shown in Table 6. In addition, $GHG\text{-CO}_{2,op}$, was calculated using Equation 9 and the devices' luminous efficacy and operational lifetime values from Table 3. Both metrics are displayed graphically in Fig. 5.

Fig.5. The corresponding $GHG\text{-CO}_2$ emissions for raw material extraction and device (1 m^2) manufacturing ($GHG\text{-CO}_{2,dev}$) and yearly $GHG\text{-CO}_2$ emissions from operation ($GHG\text{-CO}_{2,op}$) for each P-OLED device architecture and the blue inorganic LED.

The $GHG\text{-CO}_{2,dev}$ is the lowest for the top-emitting P-OLEDs (54 kg of CO_2) due to the low embodied

energy for the top-emitting architecture (Table 6). The blue inorganic LED had a substantially higher $GHG-CO_{2,dev}$ (5031 kg of CO_2) compared to all of the P-OLED devices due to the large amount of energy embodied in the materials and the correspondingly high direct process energy. The $GHG-CO_{2,op}$ was lowest for the top-emitting inverted and blue inorganic LED devices (~1678 kg/a of CO_2 and ~1734 kg/a of CO_2 respectively) because they are more efficient at converting electrical input power to light (Table 3). The top-emitting inverted P-OLED produced approximately 67% less CO_2 during operation than the bottom-emitting conventional P-OLED. Therefore, the top-emitting inverted is the most promising P-OLED architecture for maintaining a low carbon footprint.

The post-use environmental effects of both P-OLEDs and inorganic LEDs also need to be considered; however, materials toxicity and degradation, as well as materials recycling, are complex and relatively underdeveloped topics in the context of optoelectronic devices. As a result, data is lacking on the environmental impacts and embodied energy associated with processes being used or under development for disposing of or recycling advanced electronic materials [98]. However, metals can be recycled from both types of devices (including indium, tin, silver, gallium and aluminium) either directly or as a by-product [100]. Costs of purifying the recycled metals is likely to be a compounding issue. Furthermore, it has been shown for OPV the glass substrate can be removed and reused with almost no difference in efficiency, and the polymer layers can biodegrade without leaving harmful elements in the environment [34]. While it would be ideal to recycle the P-OLED and LED devices, the energy required to recycle should be considered. Typically, the energy required to recycle a material is less than that for production of the virgin material [25]. All else being equal, based on the embodied energy (Table 6), the energy required to recycle the inorganic LED (4650 MJ) would still be significantly greater than that of the P-OLEDs (~50-96 MJ) which would make the P-OLED devices the more sustainable choice.

Although recycling removes some of the contaminants; unfortunately, optoelectronic devices (recycling rate of 10%) are not recycled at the same rate as other hazardous consumer products (recycling rate of 24% to 90%) [99], and large amounts of optoelectronic materials and devices still end up in landfills or recycling centers where they can adversely affect human health and the environment due to

leaching and evaporation of hazardous substances such as heavy metals [101]. The actual amount of hazardous materials depends on the type of optoelectronics, but as a result of these health and environmental risks governmental agencies have begun to regulate optoelectronic recycling [102].

6. Interpretation, Scenario Analysis and Conclusions

6.1. Interpretation

While improvements in the operational lifetime of P-OLEDs must be made to be competitive with the comparatively long operational lifetime of inorganic blue LEDs, the top-emitting inverted P-OLED device architecture appears to be the most promising device in terms of projected electrical-to-optical power efficiency, with a high luminous efficacy of 7.8 lm/W compared to the 7.5 luminous efficacy for the inorganic blue LED. Furthermore, the device costs per area of P-OLEDs were between 3-5 times cheaper than inorganic LED device costs per area, which would make P-OLED devices more immediately appealing. The embodied energy in the blue inorganic LED was significantly higher than that for all P-OLED device architectures. However, since the embodied energy was only a small fraction (~0.2% for P-OLEDs and 8% for the inorganic blue LED) of the use-phase energy of each P-OLED device, the use-phase was deemed to be the most critical stage to focus on to reduce energy consumption and environmental impacts associated with GHG-CO₂ emissions.

6.2. Scenario Analysis

The above interpretation compares laboratory small-scale prototype P-OLEDs to commercial-scale (i.e., mass-produced) blue inorganic LEDs with the P-OLEDs are already cheaper in terms of C_{dev} , and it is likely to remain the case during scale-up of device fabrication. However, OLED large-scale fabrication methods are not well developed and need to be further refined. One of the current obstacles to widespread commercialization of OLEDs is their overall high cost due to small-scale manufacturing and use of ridged substrates and vacuum deposition methods during the OLED or P-OLED device fabrication.

In order to produce P-OLEDs on a large scale and at low cost, the rigid substrate would need to be replaced with a flexible substrate like polyethylene terephthalate (PET) to enable reel-to-reel processing, materials wastage would need to be decreased, and material types and processing methods would need to be reevaluated. For example, the glass substrate contributed ~25% and ~44% (bottom-emitting and top-emitting, respectively) to the C_{mat} and if it was replaced with PET, assuming a thickness of 0.143 mm [31, 35, 108], and at a cost of ~0.16 \$/g, the C_{mat} for each device would be reduced by approximately 6% and 10%, respectively [32]. Furthermore, flexible substrates extend the range of applications for P-OLEDs into not only lighting and standard display options, but also such markets as wearable electronics. In addition, the U.S. Department of Energy estimates that the material utilization rate is as low as 30% for vapour deposition and as high as 90% for solution deposition fabrication [9]. In this study, we did not consider fully solution-based fabrication of all layers, thus the deposition of certain layers (e.g., the metal and ITO) by vacuum methods would result in high amounts of materials wastage and increase our predicated C_{dev} . If we account for wastage, C_{dev} is 548 \$/m $\pm$ 384 \$/m and 561 \$/m $\pm$ 225 \$/m for the conventional and inverted bottom-emitting P-OLED architectures, respectively; which represent a factor of 2.1 more than without wastage. However, C_{dev} for the top-emitting device architecture does not increase as significantly when wastage is accounted for because most layers are solution processed (i.e., ITO is eliminated) with C_{dev} increasing only by a factor of 1.25 (to 195 \$/m $\pm$ 41 \$/m) for the top-emitting conventional and inverted device architectures.

Fully solution processed fabrication of P-OLEDs would require alternative material choices to some of those listed in Table 1-2. For example, bulk silver would need to be replaced with silver ink. This alternative material would not have much effect on the overall price as the silver layer(s) do not contribute significantly to C_{dev} ; see Fig. 3. However, full solution processing would only be possible for the top-emitting devices due to the vacuum deposition needed for deposition of ITO for conventional P-OLED devices. In addition, for fully-solution-processed, large-scale production the cost of the materials would decrease by a factor of 2, at least, compared to those reported in Table 2 (resulting in

C_{mat} between 63\$/m² and 123\$/m² for the bottom-emitting P-OLED and between 44\$/m² and 65\$/m² for the fully-solution-processed top-emitting P-OLED) because of the bulk purchase of material from the suppliers [32-38]. These estimates are on target with estimates reported by Azzopardi et al., Powell et al. and others for the total device cost of a commercial-scale OPV module which ranges from 45\$/m² to 264\$/m² [32-38].

When addressing the scalability of OLEDs, changes to device luminous efficacy and operational lifetime are important considerations. Large-scale production is likely to yield devices with lower luminous efficacy and operational lifetimes compared to those for small-scale prototypes, due to the increased likelihood of non-uniformities over large active areas as a result of defects, layer thickness variations and/or electrical shorts [39,109]. Reductions in luminous efficacy, in particular, are expected to increase the yearly operating cost of these P-OLED devices. While it is expected that there will be a reduction in both luminous efficacy and operational lifetime due to large-scale production, P-OLEDs are currently manufactured at the lab- and pilot-scale which makes quantification of the reduction in performance difficult. Therefore, in order to estimate the efficiency and lifetime reduction caused by large-scale production, we draw comparisons with reports on the commercial scale-up of OPVs. Lab-scale efficiencies for optimized polymer-based OPVs have been reported to be between 10% and 12% (fully solution processed and vacuum evaporated) and OPV modules produced using large-scale processing methods are approximately 2% efficient; thus indicating a factor of up to 6 reduction in efficiency during scale-up [38,39,109]. Consequently, assuming a similar reduction in the luminous efficacy for the top-emitting, inverted P-OLED, luminous efficacy would be reduced to 1.3 lm/W for a device fabricated using large-scale production methods.

Furthermore, we can assume a worst-case operational lifetime of 1 year based on prior studies of OPV devices fabricated by large-scale production methods [29,110-112]. This assumption would reduce the top-emitting inverted P-OLED operational lifetime by a factor of 9.6 (i.e., to 2920 h). To illustrate the effect of assuming such significant reductions in luminous efficacy and operational lifetime under a “worst-case” large-scale production scenario, we carried out a scenario analysis using the Monte-Carlo

method for a top-emitting P-OLED where we calculate probability distributions for C_{op} , E_{use} and $GHG-CO_{2,op}$ under three different scenarios (Fig. 6), assuming relative standard deviations of the luminous efficacy and operational lifetime of 25% [29,112-118].

Fig. 6. Histograms showing the probability distribution of (a) C_{op} , (b) E_{use} and (c) $GHG-CO_{2,op}$ for top-emitting inverted P-OLEDs (1 m²) generated using the Monte-Carlo method [29] for three different scenarios: small-scale, which assumes luminous efficacy (B_f) and operational lifetime (L) values of 7.9 lm/W and 28000 h (as reported in Table 3); large-scale (worst case), which assumes a factor of 6 reduction in B_f compared to the small-scale scenario and a 2920 h operational lifetime (i.e., 1 year); and large-scale (best case), which assumes a future “best case” large-scale production scenario that results in P-OLEDs with B_f and L values of 7.9 lm/W and 50000 h, respectively. Normal distributions for B_f and L were generated as inputs for the Monte-Carlo analysis, assuming a relative standard deviation of 25% in B_f and L to represent typical performance parameter variations for polymer optoelectronic technologies. The Monte-Carlo analysis was carried out using Microsoft Excel with 10000 random sampling iterations of the input distributions employed to calculate the probability distribution for C_{op} , E_{use} and $GHG-CO_{2,op}$.

The first scenario, assumes average luminous efficacy and operational lifetime values achievable using current small-scale production approaches, as reported in Table 3. The second scenario assumes “worst-case” luminous efficacy and operational lifetime values reported above due to large-scale production methods. The third scenario assumes a future “best case” scenario in which the luminous efficacy and operational lifetime values at large-scale production are 7.9 lm/W (same as currently-achievable small-scale production value) and 50000 h (the operational lifetime of a commercial inorganic LED). Figure 6a shows that C_{op} was similar for small-scale and best-case, large-scale production at 94\$/a ± 33 \$/a and was significantly lower than C_{op} for the worst-case, large-scale production (562\$/a ± 196 \$/a), because C_{op} is inversely proportional to luminous efficacy and is insensitive to operational lifetime

(as it is calculated on a yearly basis). However, E_{use} , is both inversely proportional to luminous efficacy and directly proportional to operational lifetime. Therefore, the larger operational lifetime values for best-case, large-scale production resulted in larger E_{use} values (58 GJ $\pm$ 26 GJ) compared to the small-scale (33 GJ $\pm$ 14 GJ) and worst-case, large-scale production (21 GJ $\pm$ 9 GJ). In other words, a given device produced under the best-case, large-scale production scenario consumes significantly more energy than a device produced at small-scale or for the worst-case, large-scale production scenario, simply because it operates for longer. However, $GHG-CO_{2,op}$ exhibited a similar trend to the C_{op} data as it is also calculated on a yearly basis (and, therefore, is independent of operational lifetime) with small-scale and best-case, large-scale production scenarios exhibiting the lowest emissions.

Based on these scenarios, it is hypothesized that P-OLEDs would have to be mass-produced with luminous efficacy and operational lifetime values reported for small-scale devices (Table 3) in order for them to be viable in terms of the metrics C_{op} and $GHG-CO_{2,op}$ and competitive with commercial inorganic LED counterparts. Luminous efficacy, in particular, is the more critical performance parameter to maintain upon scale-up since the yearly cost and energy to operate P-OLED devices and the yearly $GHG-CO_2$ emissions during operation are significantly greater than for the production of P-OLEDs even under the best-case, large-scale production scenario. For example, the projected best-case C_{op} (94 \$/a) and $GHG-CO_{2,op}$ (~1800 kg/a of CO_2) for top-emitting, inverted P-OLEDs fabricated using large-scale production are greater than the projected C_{dev} (~55 \$/m²) and $GHG-CO_{2,dev}$ (~50 kg of CO_2) for large-scale production. Therefore, we expect that regular replacement of a P-OLED device would be relatively inexpensive and would have low greenhouse gas impacts - particularly in comparison to a commercial inorganic LED with similar luminous efficacy (C_{dev} of 844 \$/m² and $GHG-CO_{2,dev}$ of ~5000 kg of CO_2) - thereby making operational lifetime less critical. However, since processing techniques and manufacturing methods have not been standardized for P-OLEDs, it is difficult to draw definitive conclusions on what the projected device performance parameters should be for P-OLEDs upon scale-up. Therefore, studies such as ours would have to obtain performance parameter data from optimized large-

scale production processes for P-OLEDs (which are still under development) and draw comparisons with performance parameter data from existing optimized laboratory-scale or pilot-scale processes, which may result in currently unforeseen benchmarks [5]. Finally, it should be noted that for our study we focused only on blue light-emitting P-OLEDs and the metrics would most likely be improved for red and green P-OLEDs due to their higher efficiencies and longer operational lifetimes.

5.3. Conclusions

In conclusion, from a comparison of various P-OLEDs device architectures it was found that the top-emitting inverted P-OLED architecture is likely to be the most promising device architecture to pursue in terms of achieving operational lifetimes and efficiencies that are competitive with commercially-available blue inorganic LEDs and to achieving fully-solution processed large-scale production. Additionally, the device costs per area and embodied energies for the top-emitting P-OLEDs were significantly lower than those for the blue inorganic LED, making P-OLEDs already competitive in terms of up-front cost and energy expenditures. Given these factors and the performance parameters (luminous efficacy and operational lifetime) currently-achievable at lab-/prototype-scale, top-emitting P-OLEDs could be adopted for portable optoelectronic technologies (e.g., cell phone displays; indicator lights) due to the relatively short use stage of such technologies and inexpensive materials requirements that allow consumers to dispose of them after only 5-10 years. However, the performance parameters need to remain at current lab-/prototype-scale values during development and scale-up in order to ensure their performance is competitive with inorganic LEDs. Maintaining high luminous efficacy (i.e., electricity-to-light conversion efficiency) upon scale-up will be more important than maintaining long operational lifetimes, since the yearly cost and energy to operate P-OLED devices and the greenhouse gas emissions during operation are significantly greater than for the production of P-OLEDs. Therefore, regular replacement of a P-OLED device would be relatively inexpensive and would have low greenhouse gas impacts; particularly in comparison to an inorganic LED with similar luminous efficacy.

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