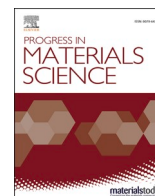


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## Printing of flexible light emitting devices: A review on different technologies and devices, printing technologies and state-of-the-art applications and future prospects

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### ABSTRACT

The primary purposes of lighting remain visibility and safety, but our quality of life can be improved by creating a complete visual environment that includes needs like health and communication. Therefore, it is indispensable to go past rigid, planar lighting towards flexible lighting. This also implies the use of inexpensive application techniques qualified for continuous manufacturing like printing techniques. Suited to fulfil these lighting needs are the alternating current powder electroluminescent (ACPEL) device, the organic light emitting diode (OLED) and the light emitting electrochemical cell (LEC) thanks to thin flexible and printable layers. ACPEL devices can be screen printed establishing economic, simple manufacturing. Their high AC-driving voltage makes them more fitted for non-wearable applications. Well-matched for wearable lighting are OLEDs thanks to low DC-driving voltages. The high sensitivity to oxygen and water of the OLED materials however necessitates a high barrier encapsulation. Unlike OLEDs, LECs realize a balanced charge injection using mobile ions in their active layer broadening the choice in electrodes, but the improvement in lifetime is limited because of incomplete knowledge of the working mechanisms. This review gives an overview of these devices and their working principles, materials, deposition techniques, applications, state of the art and an outlook.

## 1. Introduction

Light is one of the most important necessities of life. Besides enabling vision it forms the physical stimulus behind many biological, chemical and physical changes in life forms. The first notion of controlled fire was found about 250 000 years ago with wood, animal grease and pitch as primary fuels. It had multiple purposes going from lighting and heat to preparing meals and protection. The first lamps, where the focus was mainly on light, used animal fat as fuel. More than 4500 years ago the first oil lamps made their appearance, followed by gas lamps and kerosene lamps. For thousands of years light was created exclusively by the combustion of fuels. That changed during the 1800s, when electricity was made visible by arc lighting and the more practical incandescent lighting. The latter was truly a breakthrough in lighting and is basically a filament heated up to the point where it starts to glow contained in a vessel to control the atmosphere [1]. Science did not stop there and electrical lighting was widely researched and developed leading to the invention of the following major light sources: fluorescent lamps, high pressure sodium (HPS) lamps, mercury lamps, metal halide

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(MH) lamps, tungsten halogen (TH) lamps and light emitting diodes (LEDs) [2].

While electrical lighting evolved rapidly over the years, so did our needs. Although the primary purposes of lighting are still visibility and safety, our quality of life can be largely improved by creating a complete visual environment that includes other needs such as health, communication, productivity, comfort, aesthetics and mood. To meet all of these needs it is necessary to think beyond conventional rigid and planar lighting and pave the way for bendable lighting, such as light emitting devices on substrates that can be bended, flexed or even stretched like paper, polyethylene terephthalate (PET), other plastic-like materials and even textiles. This opens the door for a wide variety of applications. Indoor and outdoor applications such as wallpaper, banners, flags, safety indications and advertisement can be realised and also wearable applications such as safety and protection clothing, fashionable clothing and even light therapy and photobiomodulation are within reach.

To preserve the flexibility of the substrate the light emitting devices should consist out of very thin and flexible layers that can preferably be coated or printed directly onto the substrate to minimise the need for additional adhesion layers. By using these printing and coating techniques, different designs can be applied that enhance the functionality of the final light emitting devices and/or maintain the properties of the substrate. Within this respect, the electroluminescent device (EL), the organic light emitting diode (OLED) and the light emitting electrochemical cell (LEC) are the perfect candidates for future lighting applications, as they consist out of thin layers that can be directly printed or coated on flexible substrates.

The history of electroluminescence commences more than 100 years ago, when in 1907 Henry Joseph Round was the first to witness the phenomena when a crystal of carborundum lit up when he applied a DC voltage to it [3]. The earliest report of alternating current (AC) electroluminescence was given by George Destriau in 1936 when he conducted experiments with powder phosphors suspended in oil [4]. In general electroluminescence is the non-thermal generation of light as a consequence of applying an electric field to a material. In other words, it is the conversion of electrical energy into optical energy [5,7,280]. During the 1940s transparent conducting electrodes based on tin oxide (SnO<sub>2</sub>) were discovered which captured the interest of both scientists and industry [8]. It was not until 1952 that engineers at Sylvania Electrical Products, Inc. used the phenomenon to create a practical EL device, the 'inorganic powder type' or the 'dispersion type' EL panel [9]. The following decade, the device was researched worldwide thanks to its advantages such as a multicolour emission, a low power dissipation and the lack of catastrophic failure. The first EL devices were constructed out of a doped ZnS powder suspended in a dielectric placed between two electrodes. This set-up is still popular today and known as the high field AC powder EL device (ACPEL). Other types of EL devices do exist [10], but use expensive manufacturing techniques and are not suited for the applications mentioned in this article due to higher required electric fields [11]. Due to their low brightness, lifetime and efficiency research slowed down during the 1960s and 1970s, but the devices regained interest in the 1980s and were commercialized mainly to be used in advertisement and displays [9,13,281]. Nowadays the ACPEL device is the most popular commercially available high-field EL device due to its high potential for illumination and display applications which mainly lies in the advantages of using flexible substrates and low cost roll-to-roll fabrication processes. The utilisation of novel materials, not only improved the device performances, but also created the possibility of highly flexible and even stretchable devices [14,15] that are mostly applied using the screen printing technique [282]. Noteworthy is that recent research is often showing the application of ACPEL devices on textile substrates [17–19], but the devices are however not suitable for wearable clothing because of their AC driving voltage of around 100 V.

Organic electroluminescence was introduced by André Bernanose et al. in 1953 by exciting organic films at a very high AC voltage of 500–2000 V [20]. In 1963 Martin Pope et al. however demonstrated electroluminescence from a single crystal anthracene by applying a DC voltage [21]. The publication of Ching Tang and Steven Van Slyke introduced the first small molecules OLED (SM-OLED) in 1987 [22]. In its most basic form, an organic light emitting diode is constructed out of an emissive layer (EML) sandwiched between two electrodes. Small molecules were the first building blocks of the EML and could only be applied using expensive and non roll-to-roll compatible vacuum deposition techniques, but nowadays they are also applicable using wet chemical deposition techniques [283]. Their material properties are however relatively easy adaptable. In 1990 a group at the University of Cambridge replaced the small molecules with a conjugated polymer poly(paraphenylenevinylene) (PPV) and therefore developed the first polymer OLED (P-OLED) [24]. Contrary to small molecules, polymers have a good chemical stability and can be deposited using low-cost solution processing techniques [25]. In 1992 GörGustafsson et al. fabricated the first flexible OLED on polyethylene terephthalate (PET) using poly [2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) as an active emissive layer [26]. Five years later Gu et al. demonstrated a SM-OLED on a thin plastic substrate precoated with indium tin oxide (ITO) [27]. During the following decades these first publications gave rise to a worldwide research resulting in highly flexible and efficient OLEDs fabricated with innovative materials [28–56] mostly on plastic-like materials and metal foils [13,15,53–70] applied by a variety of both vacuum deposition and solution processing techniques. OLEDs are often referred to as the future in solid state lighting due to their light weight and thin layers, flexibility, high efficiencies and non-expensive and environment friendly fabrication techniques. In addition they just require an applied direct current (DC) voltage of around 3–5 V to ensure light emission, which makes them eligible for many applications like wearables amongst others. Although a lot of progress has been made, there are some remaining challenges to be dealt with, for example their limited lifetime and fast degradation under ambient conditions [71].

In 1995 Qibing Pei et al. discovered a new and simple luminescent device with two metallic electrodes and an active layer that is composed of a conjugated polymer, a solid electrolyte that consists of a high concentration of mobile ions and ionically conductive material [72]. The working mechanism of this device is based on the injection of electronic charges also described as electrochemical oxidation and reduction of the conjugated polymer, hence the name of the device, the Light Emitting Electrochemical Cell. A few months later J. K. Lee et al. used a ruthenium polypyridyl complex, an ionic transition-metal complex (ITMC), as an emitter material in the same device [73]. Contrary to OLEDs, LECs accomplish a well-balanced charge injection of electrons and holes into the emissive layer by the usage of mobile ions instead of a built-in electrical potential created by the difference in work function of the electrodes

which makes the material choice for the electrodes a lot easier [74].

This implies that stable materials that do not degrade rapidly under ambient conditions qualify as electrodes. The optimal operation of OLEDs depends strongly on the correct nm-thickness of the organic layers, which is of less importance for LECs and which also implies the ease to apply the devices with low-cost printing and coating techniques. The latter leads to the most appealing advantage of the technology being the low production cost, that would be one order of magnitude lower than that of LEDs and OLEDs [75]. Even though a lot of research has gone into it, the mechanism at work behind the operation of the LECs is however highly complex and not yet fully understood. This complexity brings forth several degradation mechanisms, side reactions and over time a loss in efficiency and luminescence quenching of the recombination zone [76]. Compared to the other light emitting devices, the LEC also shows a relatively slow turn-on-time.

Since their discovery these solid state lighting devices have been constantly improved by gaining more insight into the mechanisms at work and by the introduction of innovative materials and application techniques. A major advantage lies in the application of printing and coating techniques to fabricate the devices which paves the way for a total freedom-of-design and an improved integration on the substrate of choice. Furthermore these techniques make a roll-to-roll and more economical and ecological production possible.

This review emphasizes an elaborate understanding of the devices with a detailed explication of the light emission mechanisms, an overview of the stack build-up and deposited materials, an insight into the printing and coating techniques, a synopsis of the devices performances and the current state of the art.

## 2. Devices

### 2.1. Working mechanism

The knowledge and understanding of the mechanisms responsible for light emission in the different devices are of high importance to define the materials to be used for the stack build-up and the techniques to apply them. Although the working principles behind the OLED are well known and defined, this is not completely the case for the ACPEL device and the LEC. Several theories were proposed to explain the processes at work for light emission of the ACPEL devices, with the Fischer model being most common the working mechanisms behind the LEC are only partially known due to their complexity and there are still some unexplained processes at work.

#### 2.1.1. ACPEL device

No consensus has been reached on a general theory of the processes at work inside the ACPEL device, but the bipolar field-emission model or the Fischer model is widely accepted [11,77,78]. Albrecht Fischer observed that light is emitted in the form of lines or striations extending through most of the diameter of the phosphor particles when a voltage above a certain threshold is applied. Typically the luminescent lines consist of two collinear halves that give the impression of two comets fleeing each other (Fig. 1). A halve lights up alternately each time the adjacent cell electrode becomes positive. These observations led Fischer to propose the following model explaining the EL-mechanism.

When ZnS EL powders are produced, a firing process with high temperatures (1100–1200 °C) takes place during which the hexagonal phase (wurtzite) of the ZnS crystal is dominant. Cooling the powders down however causes a phase transition to the cubic crystal structure (zinc blende), leaving the particles with many defects like imperfection lines and screw dislocations. Excessive and insoluble copper (Cu), used as a dopant, precipitates on these defects forming thin high conducting copper sulfide ( $\text{Cu}_{2x}\text{S}$ ) lines or needles in the crystal matrix. These needles are conductors meaning that the conduction bands and valence bands are not separated and there is no energy gap leaving the electric charge carriers, electrons in this case, free to move within the needles. ZnS, on the other hand, is a wide bandgap semiconductor meaning it has a (band)gap between the valence band and the conduction band that can be

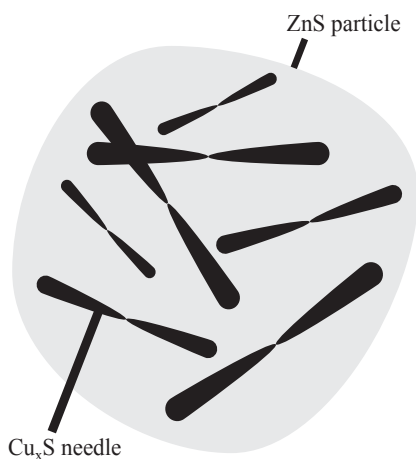


Fig. 1. EL lines of ACPEL device as observed by Fischer.

overcome when electrons obtain sufficient energy to move from the valence band into the conduction band leaving a hole in the valence band. The electric field lines and band model before applying an external electric field to the device are illustrated in Fig. 2.

By applying an external electric field (Fig. 3), a relaxation takes place within the  $\text{Cu}_{2-x}\text{S}$  needle causing a field intensification from the centre towards the tips of the needle, with a zero-field in the centre and a relatively high electric field concentrated at the needle's tips. This field intensification results in a bending of the energy bands which makes tunnelling of holes from the end of the needle and electrons from the other end through the barrier into the ZnS possible. Electrons are attracted by the positive electrode, while they are repelled by the negative electrode. Electrons are then captured in the conduction band on the positive side, while the holes are trapped in the valence band on the negative side.

When the electric field is reversed (Fig. 4), the trapped electrons want to return to their ground state and recombine with the trapped holes giving rise to the typical recombination radiation (primary light peak). A secondary light peak may take place caused by electrons returning from the opposite end of the crystal that recombine with the trapped holes.

### 2.1.2. OLED

An OLED emits light when a low DC-voltage signal is applied across the device. The basic principles behind the light emission of an OLED are well established and the following processes take place sequentially: charge injections at the cathode and anode, charge transport, recombination and exciton formation, and finally relaxation and light emission [79–81]. This working mechanism requires a well-balanced charge injection by a built-in electric field across the device that is realised by using different work function materials for the anode (high work function) and cathode (low work function). The work function is the minimum energy necessary to extract an electron from a material to the vacuum immediately outside of that material. It is however not a material constant and can be altered by carrier-doping, modification of the surface dipole or a combination of both [82].

Fig. 5 shows the operating mechanism of an OLED. By applying a DC-voltage, electrons are injected from the cathode into the LUMO of the electron injection or transport layer and the holes from the anode into the HOMO of the hole injection or transport layer. Subsequently the electrons hop into the lowest unoccupied molecular orbital (LUMO) and the holes into the highest occupied molecular orbital (HOMO) of the neighbouring organic semiconductor. To achieve this, an energy difference between Fermi level and LUMO or HOMO level has to be overcome by applying a certain threshold voltage or by aligning the different energy levels. Electrons and holes then continue their path through the LUMO or HOMO level of the other layers with a similar charge carrier mobility, meaning that they move with an equal speed through the layers when pulled by an electric field. During this displacement through the device they have to traverse several interfaces with possible energy barriers to overcome, causing a rise in the required driving voltage.

The recombination of electrons and holes takes place in the emissive layer according to the Langevin theory of the recombination process [80]. If a distance between an electron and a hole is reached where their mutual binding Coulomb energy is greater than the thermal energy, it is inevitable that they recombine and form an exciton. This exciton then decays to its electronic ground state while emitting a photon called recombination radiation.

### 2.1.3. LEC

The underlying physics of the LEC is highly complex and its understanding is still limited. Two competing theories have been brought forward: the electrodynamic (ED) model [83–87] and the electrochemical doping (ECD) model [74,87–91]. van Reenen et al. showed that both models are actually limits of one unifying model and that they depend on the rate of carrier injection [92]. The ECD model applies to non-limited carrier injection when a dynamic p-n junction is formed, for example for ohmic contacts. The ED model on the other hand addresses the injection-limited devices.

Unlike OLEDs, LECs do not achieve the requirement of a balanced charge injection by electrodes with a difference in work function. A possible significant energy barrier between the Fermi level of the electrodes and the HOMO and LUMO level can be overcome by the mobile ions in the active layer. By applying a low bias, these mobile ions can migrate towards the electrodes, decreasing the energy barrier for carriers. Depending on the working model, the reasons for this decrease are different [92,93].

In the ED model (Fig. 6), anions and cations drift respectively towards anode and cathode and start to build up causing an enhancement of the electric field at the electrodes. This results in the formation of very thin electric double layers (EDL) of approximately 1 nm. The presence of these EDL causes a substantial drop of the electric potential at the electrodes and facilitates charge injections into the active layer. When the applied voltage is high enough electrons and holes travel through respectively the LUMO and HOMO level of the semiconductor towards each other and recombine to form an exciton in the field-free region between the EDL and emit light.

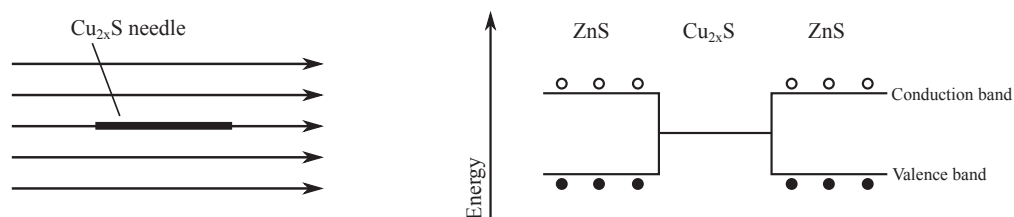
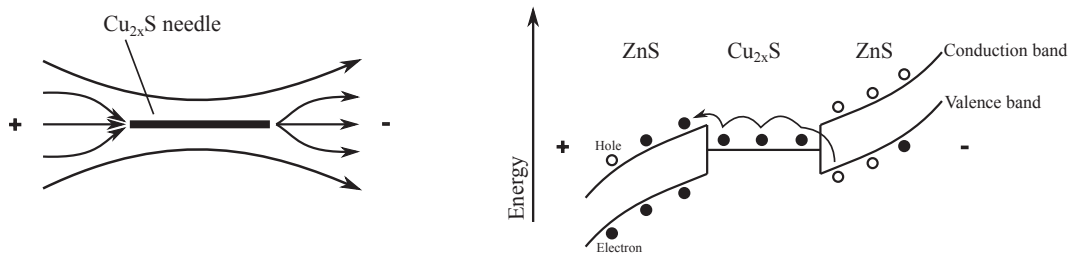
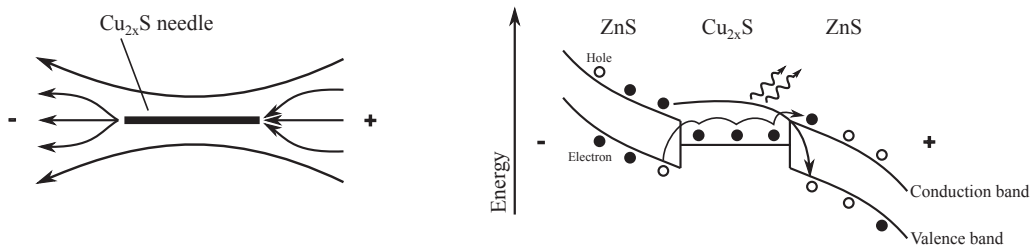


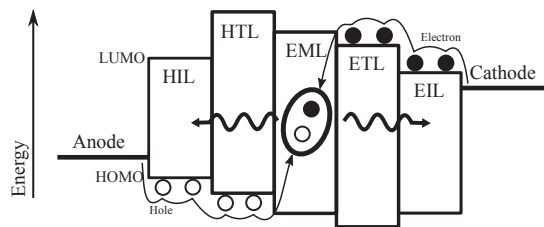
Fig. 2. Electric field lines and band model of ACPEL device before applying an external electric field. Electrons can freely move within the  $\text{Cu}_{2-x}\text{S}$  needles.



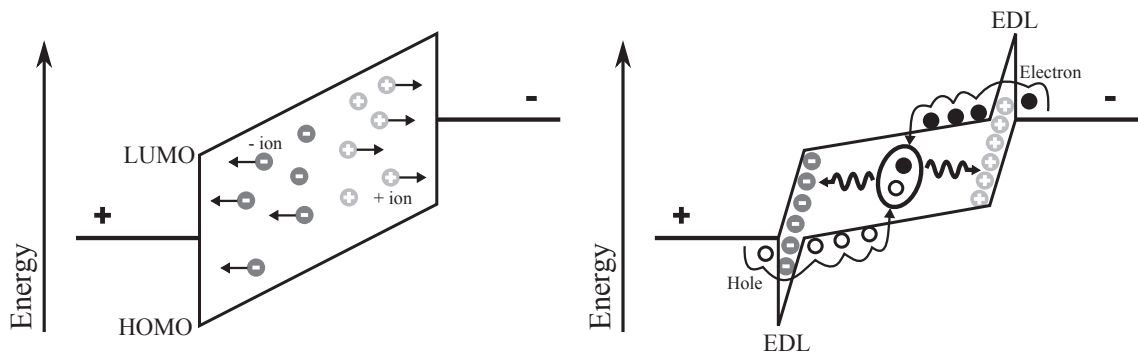
**Fig. 3.** Electric field lines and band model of ACPEL device after applying an external electric field. This enables tunnelling of holes and electrons from the  $\text{Cu}_{2x}\text{S}$  needles to the ZnS. Electrons are captured in the conduction band on the positive side, while holes are trapped in the valence band on the negative side.



**Fig. 4.** Electric field lines and band model of ACPEL device after electric field reversal. Trapped electrons want to return to their ground state and recombine with the trapped holes resulting in recombination radiation.



**Fig. 5.** Working mechanism OLED. By applying a DC voltage electrons and holes are injected into respectively the anode and the cathode. They traverse through the LUMO or HOMO level of the different OLED layers to recombine in the emissive layer and form excitons. These excitons decay to their electronic ground state while emitting photons.



**Fig. 6.** Working mechanism LEC EDL model. After applying a DC voltage anions and cations drift towards anode and cathode forming electric double layers (EDL) and facilitating charge injections into the active layer. Electrons and holes travel through the LUMO and HOMO level of the semiconductor and recombine to form excitons in the field-free region between the EDL and emit light.

In the ECD model (Fig. 7), charge injection is also made possible by the formation of the EDL like in the ED model, but after non-injection limited ohmic contacts are formed, the initial electronic charge injection is compensated by the migration of ions. At the cathode the injection of electrons is compensated by positive ions or cations which results in the formation of an n-type doped region. On the opposite side the extraction of electrons at the anode attract negative ions or anions and form a p-type doped region. This electrochemical doping therefore facilitates charge injection into the active layer. The doped regions grow towards each other into the active layer and form a p-n-junction when they meet. The electric potential drops significantly so recombination of electrons and holes becomes possible to form excitons and to finally emit light.

## 2.2. Stack build-up and materials

The layer stack of each of the three light emitting devices differs considerable, hence the dissimilar working mechanisms. While the ACEPEL device and the LEC require typically a simple set-up with only four and three layers respectively, the OLED usually demands more interlayers to enhance the efficiencies. In this paragraph the possible structures and build-ups for each device will be discussed together with a non-exhaustive summary of materials for every layer.

### 2.2.1. ACEPEL device

An ACEPEL device can be considered as a light emitting capacitor which contains phosphor particles embedded in a dielectric and packed between two electrodes [11]. By applying a high voltage alternating current (AC) signal ( $\pm 100$  V/m) with a high frequency the phosphor particles emit light [10].

Typically an ACEPEL device is constructed either in the forward structure or in the reverse structure [11]. Light emission in a forward structured ACEPEL device (Fig. 8) occurs through the transparent substrate, often PET or polyethylene naphthalate (PEN). The transparent electrode is either a transparent conductive oxide (TCO) like indium tin oxide (ITO), which is however not solution processable, or a transparent conductive polymer (TCP) such as poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS). Subsequently a phosphor layer is applied on top of the TCO or TCP. The name of this layer might be confusing since it refers to the general term for the materials used for the active light emitting phosphorescent layer and not to the element phosphor (P). This layer consists of an inorganic compound such as doped zinc sulphide (ZnS) powders with a particle size of 5–20  $\mu\text{m}$  and a layer thickness of about 20  $\mu\text{m}$ , an activator material which acts as an acceptor such as copper (Cu), a donor like chlorine (Cl) and a binder. The choice and the concentration of dopant(s) and EL phosphors are responsible for the emitted wavelength and thus the colour of the emission. EL phosphors can be found in multiple colours being blue (450–485 nm), blue-green (485–500 nm), green (500–565 nm) and yellow-orange (565–625 nm). Other colours, including white, can be obtained by blending two or three phosphors in various ratios or by cascading the emitted light. White light can be created by mixing blue, green and orange phosphors. However, over time a colour shift takes place due to the different rates of luminance decay of the phosphors. To overcome this issue mixtures of phosphors exhibiting a similar luminance decay were developed. Another possibility is to cascade the emitted light by adding fluorescent dyes or pigments to the dielectric layer, the phosphor layer or on top of the device. This layer will act as a filter, absorbing certain wavelengths and transmitting others [94]. The phosphor layer is added on top of a 5  $\mu\text{m}$  thick dielectric layer with a high dielectric constant ( $\epsilon_r$ ) and a high breakdown voltage like ferroelectric barium titanate ( $\text{BaTiO}_3$ ) dispersed in a polymer binder and plasticizer. The values of the dielectric constant and breakdown voltage are highly dependent on temperature and layer thickness (at room temperature:  $\epsilon_r \approx 500$  [95] and breakdown electric field  $\approx 35$  kV/cm [96]). The purpose of this layer is to prevent the ions traveling to the other electrode, to avoid electrical shorts and to act as a reflector. The stack periphery is completed by a back electrode, mostly Ag or Al, with a thickness of approximately 10  $\mu\text{m}$ . Often a bus-bar is printed around the circumference of the device simultaneously with the back electrode to prevent power losses and equally distribute the power.

For a non-transparent substrate such as textiles and paper the structure of the ACEPEL device is reversed so the light passes through the top electrode (Fig. 9).

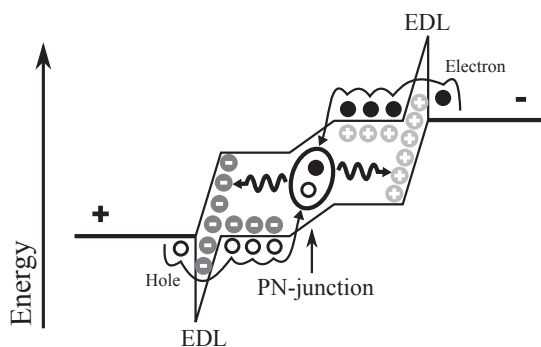


Fig. 7. Working mechanism LEC ECD model. After formation of EDL, the electronic charge injection is initially compensated by the migration of ions that form n-type and p-type doped regions. This electrochemical doping facilitates charge injection into the active layer and forms a p-n-junction making recombination of electrons and holes possible to form excitons and finally emit light.

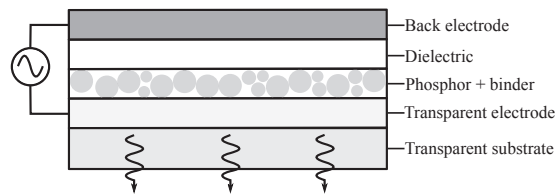


Fig. 8. Forward structure ACPEL device with a transparent electrode, a phosphor and binder, a dielectric and a back electrode.

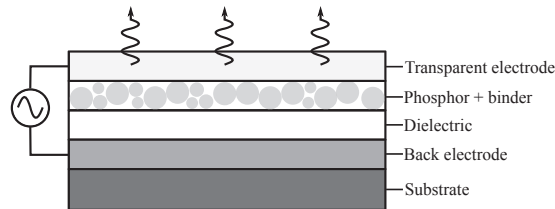


Fig. 9. Reverse structure ACPEL device with a back electrode, a dielectric, a phosphor and binder and a transparent electrode.

Besides the two most commonly used structures the floating electrode structure provides an alternative to create a two-tone ACPEL device (Fig. 10) [11]. Two highly conductive electrodes are mounted alongside each other on a substrate followed by the dielectric and emissive layer and completed by a floating transparent electrode. When operated the device acts basically like two ACPEL devices in series, which means that the applied voltage is equally divided over the two. Therefore to obtain the same brightness as a conventional ACPEL device, twice the voltage must be applied and the gap between the two electrodes does not illuminate. Differing the size of each half results in a lower capacitance for the smallest half and therefore in a voltage drop across it. Consequently the smaller half will be brighter than the larger half generating a two-tone ACPEL device.

A fourth and final alternative way to configure an ACPEL device is the interdigitated electrode structure (Fig. 11) [11]. This configuration requires no transparent electrode since the two interdigitated electrodes are printed in one run with the same material. The gap between both electrodes has to be very narrow and may not pass 100–200  $\mu\text{m}$ , otherwise the lateral field between the electrodes will not be strong enough to energize the phosphor that is mounted on top of both electrodes. This structure can be printed in only two runs, requires two different materials and can be mounted on top of any substrate. There are however considerable downsides to this structure. These devices only emit light in the gaps between the electrodes which results in a significant lower brightness than conventional devices. Practically there is also the additional difficulty of printing the electrodes as close together as possible. It is hence no surprise that this structure is not very popular.

An overview of the utilized materials for the ACPEL layers can be found in Table 1.

### 2.2.2.2. OLED

An OLED has a multi-layered structure including a bottom electrode, a hole injection layer (HIL), a hole transport layer (HTL), an electron blocking layer (EBL), an emissive layer (EML), a hole blocking layer (HBL), an electron transport layer (ETL), an electron injection layer (EIL) and finally a top electrode. All layers have a thickness between 1 and 200 nm, depending on the selected material [99].

Two main configurations can be distinguished (Fig. 12): the bottom emitting OLED (BE-OLED), where the light is emitted through a transparent anode and substrate and the top-emitting OLED (TE-OLED), where the light leaves the device through a transparent cathode. For these two configurations the structure can be completely inverted, hence the inverted BE-OLED and inverted TE-OLED [99,100]. Indispensable for OLEDs is an adequate high barrier encapsulation to protect the OLED stack materials against degradation in ambient conditions.

Similarly to the ACPEL devices, OLED structures can be deposited onto several flexible substrates. The thin nanometre layers bring however the additional requirement of an extremely flat substrate surface. This demand for a smooth surface morphology can be

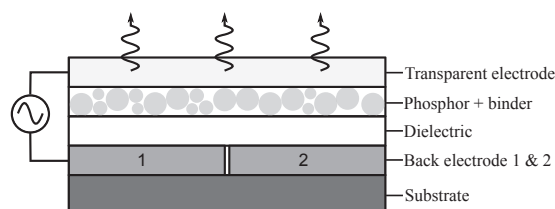


Fig. 10. Floating electrode structure ACPEL device with two back electrodes, a dielectric, a phosphor and binder and a transparent electrode.

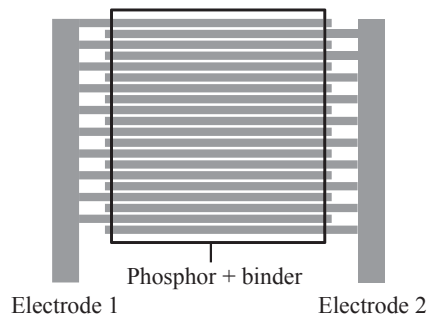


Fig. 11. Interdigitated electrode ACPEL structure with two electrodes and phosphor and binder.

Table 1  
Overview of the materials used for flexible ACPEL devices [11,97,98].

ACPEL layer	Materials
Transparent electrode	TCO ITO, SnO <sub>2</sub> , GZO, AZO, FTO
	TCP PEDOT:PSS
	Other CNT
Phosphor	ZnS:Cu,Cl(Br, I) (blue); ZnS:Cu,Cl(Br, I) (green); ZnS:Mn, Cl (yellow); ZnS:Mn, Cu, Cl (yellow); ZnSe:Cu, Cl (yellow); ZnSSe:Cu, Cl (yellow); ZnCdS:Mn, Cl (Cu) (yellow); ZnCdS:Ag,Cl (Au) (blue); ZnS:Cu,Al (blue)
Dielectric	BaTiO <sub>3</sub> , BaTiO <sub>3</sub> ceramic sheet
Binder material (dielectric and emissive layer)	Cyano-resin, PVDF
Back electrode	Ag, Al, carbon

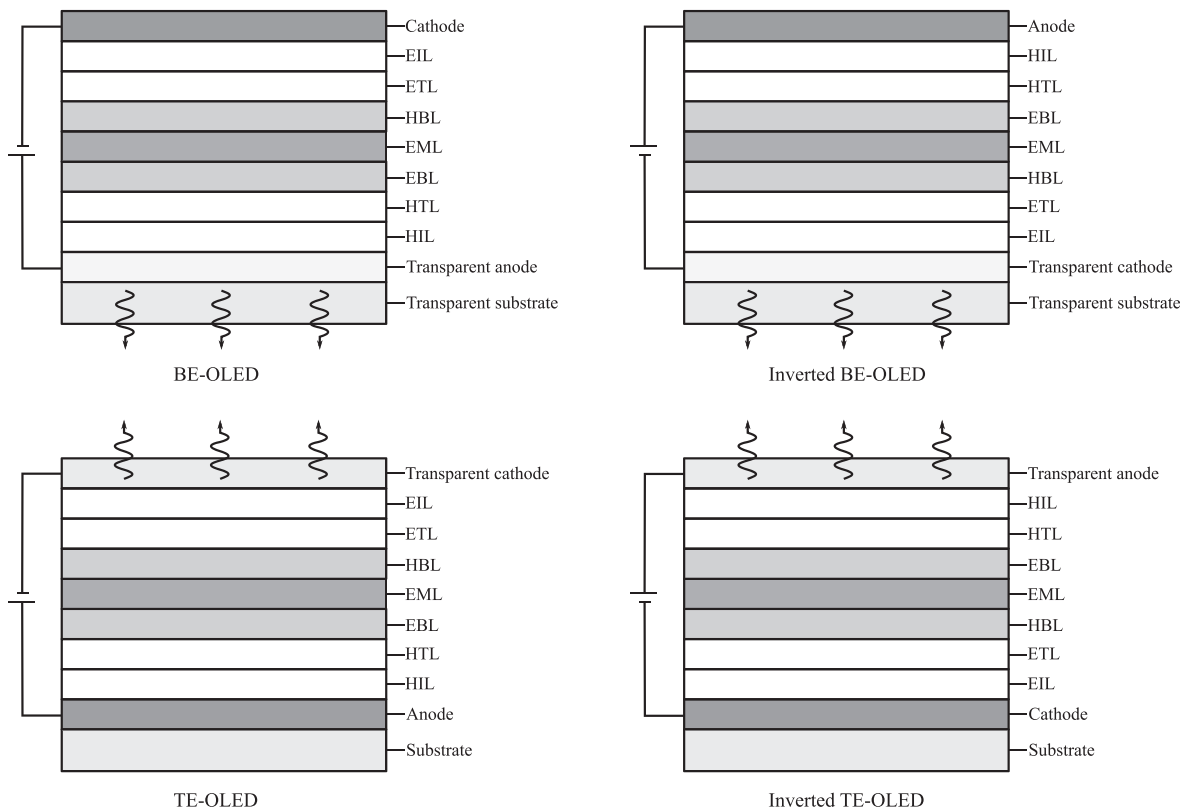


Fig. 12. (Inverted) BE-OLED and (inverted) TE-OLED with (transparent) anode, HIL, HTL, EBL, EML, HBL, ETL, EIL and (transparent) cathode.



expressed in a root mean square roughness ( $R_{\text{RMS}}$ ) lower than 2 nm and a maximum roughness of the highest asperity ( $R_{\text{max}}$ ) lower than 20 nm [81]. If these roughness numbers are exceeded, a planarization layer to smoothen the surface is necessary.

At least one of the electrodes has to be transparent for the light to be able to pass through. For this purpose transparent conducting materials (TCMs), a collective name for all transparent and conductive materials applied in optoelectronic devices, are used. To be considered as a suitable electrode the conductivity has to be higher than  $2000 \Omega^{-1} \text{cm}^{-1}$ , the transmission across the visible spectrum has to be higher than 85%, the  $R_{\text{RMS}}$  cannot pass 4 nm and the peak to valley value ( $Z_{\text{max}}$ ) has to be lower than 30 nm [81]. Typically the layers on top of the electrode planarize the surface. Frequently used as a transparent electrode are TCOs like ITO. Other possible candidates are transparent materials with a metallic conductivity. These materials however have a high absorption and reflection that have to be overcome. Examples are thin metal layers, semiconductor-metal-semiconductor (SMS) structures, graphene/carbon nanotubes (CNTs) and metal grids [71,81,101–103].

The purpose of the HIL and the HTL is to improve the amount of injected holes that migrate from the anode towards the EML. Often one layer that combines both hole injection and transport functionalities is used. The EIL and ETL have a comparable objective for the injection and transport of electrons from the cathode towards the EML. The EBL and HBL have to prevent respectively the electrons and the holes leaving the EML and moving towards the opposite electrode. Between every layer the charge carriers have to overcome an energy barrier that increases the driving voltage of the OLED and the probability of non-radiative recombination. The most challenging barrier to overcome is the one between the electrodes and the organic layers. For this reason buffer layers are often inserted between the electrodes and the following organic layers. These layers are very thin (1–10 nm) and ensure the energy alignment across the interface [71,81,101–103].

In the EML the electrons and holes recombine, form excitons, radiatively relax and emit photons. This layer either consists of conjugated polymers (P-OLED) or small molecule organic compounds (SM-OLED). Polymers are chains of covalently bonded molecular units called monomers. These monomers are interconnected by incomplete electron orbitals at the opposite ends. The semiconducting character of the polymers is a result of the alternating sequence of single and double bonds (delocalized  $\pi$  orbitals formed in carbon-containing compounds). The conjugated polymers are very flexible and their mechanical and processing properties can be modified. Luminescent or photoluminescent functional units can be attached to the backbone of the polymer. Small molecules are semi-crystalline or crystalline materials and in contrast to polymers, small molecules have defined structures and molecular weight. Thin films of these molecules can have various morphologies that have an impact on film properties like density, hardness, carrier transport and transparency. Another difference between the polymer and small molecule layer was the deposition method. Polymers can be solution processed and small molecules had to be evaporated or sublimated in vacuum resulting in higher production costs. This though is somewhat superseded since some small molecules can also be solution processed these days [71,81,101–103].

The final layer of an OLED is the top electrode. Similar as the other electrode, a high conductivity is necessary, but instead of being transparent the material has to be preferably reflective to enhance the light output at the other side (transparent electrode). Therefore mostly metals qualify as excellent back electrodes [71,81,101–103]. Table 2 summarizes the materials used for each OLED layer.

**Table 2**  
Overview of materials used for flexible OLEDs [71,81,101–104,284].

OLED layer		Materials
Transparent electrode (TCM)	TCO	ITO, IZO, $\text{SnO}_2/\text{F}$ , $\text{SnO}_2/\text{Sb}$ , $\text{Mo}/\text{In}_2\text{O}_3$ , $\text{F}/\text{In}_2\text{O}_3$ , $\text{Nb}/\text{TiO}_2$ , $\text{B}/\text{ZnO}$ , $\text{Al}/\text{ZnO}$ , $\text{Ga}/\text{ZnO}$ , $\text{In}/\text{ZnO}$ [71,81], FTO, AZO, GIO, GITO, ZIO, ZITO [102]
	Materials metal conductivity	SMS structures: $\text{ZnO}/\text{Ag}/\text{ZnO}$ , $\text{ITO}/\text{Ag}/\text{Cu}/\text{ITO}$ , $\text{ZnS}/\text{Ag}/\text{ZnS}$ , $\text{ITO}/\text{Ag}/\text{ITO}$ [81] Graphene and CNTs [71,81,101] Metal grids and nanostructures: Ag NW and NM [71,81] Dielectric-metal-dielectric: $\text{ZnS}$ , $\text{ZnO}$ , $\text{WO}_3$ (dielectric) Au, Ag (metal) [71,101] Metal-dielectric-metal [71]
Buffer layer	Anode	Small molecules: $\text{CuPc}$ , TPD, $\alpha$ -NPD, TCTA, m-MTDATA, 2-TNATA [81] Polymers: PEDOT:PSS, PANI:CSA [81] Metal-oxides: $\text{MoO}_3$ , $\text{WO}_3$ , $\text{CuO}$ , $\text{RuO}_2$ , $\text{V}_2\text{O}_5$ , $\text{Fe}_3\text{O}_4$ , $\text{Ag}_2\text{O}$ , $\text{ZnO}$ [81]
	Cathode	Inorganic salts: $\text{LiF}$ , $\text{CsF}$ , $\text{NaF}$ , $\text{MgF}_2$ , $\text{CaF}_2$ , $\text{Cs}_2\text{CO}_3$ [81] Metal-oxides: $\text{TiO}_2$ , $\text{ZnO}$ , and $\text{ZrO}_2$ [81] Organic molecules: Alq, oxadiazole-, triazole-, pyridine-, triazine-, and quinoline-based molecules [81] Polymers: PEIE, PFN-OH [81]
Hole transport layer	Small molecules	TPD, NPD, $\alpha$ -NPD, $\text{CuPc}$ , TAPC, TPA1, PDA TPD17, TPD12, TPD15, m-MTDATA, TPT9, TPT1, TPT2, TPT7, TCTA, DTASi, DTASi, MPMP [81,102], NPB, Spiro-NPB, TTP, Spiro-TPD, Spiro-TAD, $\text{CuPc}$ , PtPC, H2PC, DBTPB, PPDN, MeO-TPD [102]
Emissive layer	Polymers	PEDOT: PSS [101]
	Small molecules	Ir(III) complexes, Pt(II) complexes, Pt(II) porphyrins, Cationic Cu(I) Complexes, Neutral Cu(I) Complexes, Three-Coordinate Cu(I) Complexes, Alq <sub>3</sub> [81,101,102], TPB, PBD, bathocuproine, TPBi [102]
Electron transport layer	Polymers	PPV, MEH-PPV, $\text{OC}_1\text{C}_{10}$ -PPV [284], P Ani, PPy, PT, PPP, PA [102], P3AT, PITN, PEDOT, LPPP, PPS, PHT, P3HT, PANI, PFO [103]
	Small molecules	TPBi, PyPySPyPy, BMB-TB, $\text{POPy}_2$ , TQB, B3T, BBTB, DBPSB, Flrpic, TAZ, BCP, B3PyPB, B3PyPPM, TmPyPB [81], TAPC, LiQ, BPD, Bphen, BAlq, TPBi, TPYMB, BeBq <sub>2</sub> [102]
Back electrode	Polymers	Oxadiazole polymers [104]
	Metals	Ag, Al, Au, Mg, Ca, In [284] Mg:Ag, LiF, Mg:Al, CsF:Al, $\text{Al}_2\text{O}_3$ :Al [102]

### 2.2.3. LEC

The fundamental form of a LEC is constructed out of two electrodes, at least one of them transparent, with in-between an active layer that consists of a mixture of an organic semiconductor and a solid electrolyte that contains mobile ions. Some of the challenges faced by OLEDs can be overcome by these mobile ions since they establish proper charge injection from essentially any electrode material. As a result ambient-stable and high work function materials can be utilized for the electrodes. It also facilitates electrochemical doping of the active layer increasing the conductivity which makes it possible to use thicker active layers than OLEDs ( $\gg 100$  nm) [106]. These benefits lead to a rather simple device architecture that can easily be applied using solution processing techniques [107].

Generally two device architectures of the LEC can be distinguished: the stacked type and the planar or surface type as seen on Fig. 13. The layers of the stacked LEC are all applied on top of each other creating a large light emitting region. Research indicates that it is quite challenging to obtain a substantial light output with the planar LEC [108–110]. Because of the alongside placing of the layers, the planar structure does not require a transparent electrode and the open and large inter-electrode gives an easier insight into the electrochemical process. Nevertheless in practical applications this structure is not favourable because of the small stripe-like emissive zone [76,108], which is comparable to the interdigital ACPEL device.

LECs can be classified into three classes according to the type of ionic luminescent material used in the active layer: (1) conjugated polymers, (2) ionic transition metal complexes (iTMCs) and (3) the third generation spanning small molecules, quantum dots and luminescent nanoparticles [76]. Polymer LECs (P-LECs) have an active layer consisting of a conjugated polymer, often MEH-PPV, meshed with a salt and an ion-solvating polymer such as poly(ethylene oxide) (PEO) [72]. Conjugated polymers combine both electronic and optical properties of semiconductors with the processability of conventional polymers. The second type of LEC, the iTMC-LEC, does not require supplementary charges to drive the device since the iTMCs are intrinsically ionic [111,112]. These iTMCs consist of a metal centre generally surrounded by three organic ligands, are easily solvable in benign solvents and tend to be phosphorescent triplet emitters resulting in higher electroluminescence efficiencies than single emitters. Usually ionic bis(cyclometalated Iridium (Ir)(III) complexes are used as iTMC. Currently novel, for the third generation, electroluminescent materials are being researched including small molecules [113,114], sustainable copper(I) complexes [115], quantum dots [116] and luminescent nanoparticles [76].

To improve the performance of the device, multiple layers can be added to the three layer build-up, for example hole and electron transport layers such as PEDOT:PSS can be added to smoothen the surface of the electrode and facilitate the injection and transport of holes and electrons.

A summarization of the most frequently used materials to complete the LEC stack can be found in Table 3 [108,118,285].

### 2.3. Device performance

The performance of the light emitting devices can be quantified by their luminous output and their efficiencies. Although there is still no standardisation in the measurement and the characterisation of the light emitting devices, the luminous output is usually quantified using radiometric and photometric quantities and units [119–122], while different methods are in place to measure the reliability and efficiency of the devices [123–125,127,278]. The following paragraphs elucidate these figures of merit and also give an overview of the latest published devices performances .

#### 2.3.1. Radiometric and photometric quantities and units

Radiometry is a measurement science studying the physical measurement of the properties of electromagnetic radiation, while photometry could be considered as the field of study that describes the effects of visible light on the human eye. The radiometric and photometric quantities and units are defined by the International Commission on Illumination (CIE, 2016a) and adopted by the Consultative Committee for Photometry and Radiometry of the International Committee for Weights and Measures (CIPM) in the International System of Units (Système International d'Unités, SI) [119–122]. Table 4 shows the commonly used radiometric and photometric quantities and units. The conversion between the photometric and radiometric quantities is given by the photopic spectral luminous efficiency function  $V(\lambda)$  with a peak luminous power efficiency of 683 lm/W at a frequency  $\lambda$  of 555 nm. This function gives the spectral response of the human eye to various wavelengths of light.

Generally the above mentioned photometric quantities are used to describe the luminous performance of the light emitting devices, therefore in this review the radiometric quantities will not be discussed in detail. The most fundamental quantity in photometry is the

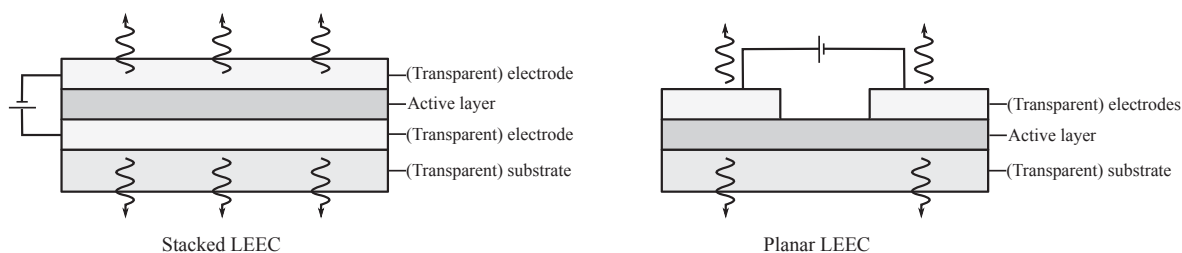


Fig. 13. Stacked and planar structure LEC with an active layer sandwiched between two (transparent) electrodes.

**Table 3**  
Overview of materials used for flexible LECs [108,118,285].

LEC layer		Materials
Transparent electrode	TCO	ITO
	Materials metal conductivity	CNTs
Hole transport layer	PEDOT:PSS	
Active layer	iTMC	Ionic Ir <sup>III</sup> complexes: f.e. Ir(pppy) <sub>2</sub> (bpy)] <sup>+</sup>
		Ionic Ru <sup>II</sup> complexes: f.e. poly-[Ru(vbpy) <sub>3</sub> ] <sup>2n+</sup>
		Ionic Os <sup>II</sup> complexes
		Ionic Cu <sup>I</sup> complexes
		Ionic Re <sup>I</sup> complexes
		MEH-PPV, PPP, PFO, PT, PF-B
Electron transport layer	Conjugated polymer	ETT-15
	Ionic conductive polymer	
	Electrolyte	PEO (Li), Crown ether/Li salt complexes, ionic liquids, LiTf
Back electrode	ZnO	
	Au, Al, Ag	

**Table 4**  
Overview of mostly used radiometric and photometric quantities and units.

Radiometric		Photometric	
Quantity	Unit	Quantity	Unit
Radiant power ( $\Phi_e$ )	W	Luminous flux ( $\Phi_v$ )	lm
Radiant intensity ( $I_e$ )	W/sr	Luminous intensity ( $I_v$ )	lm/sr = cd
Irradiance ( $E_e$ )	W/m <sup>2</sup>	Illuminance ( $E_v$ )	lm/m <sup>2</sup> = lux
Radiance ( $L_e$ )	W/(m <sup>2</sup> .sr)	Luminance ( $L_v$ )	lm/(m <sup>2</sup> .sr) = cd/m <sup>2</sup>

luminous flux, since its simplest relationship with radiant power.

The **luminous flux** ( $\Phi_v$ ) is defined as *the change of luminous energy with time*

$$\Phi_v = \frac{dQ_v}{dt}$$

where  $Q_v$  is the luminous energy emitted, transferred or received and  $t$  is time. The luminous flux is expressed in lumen (lm). One **lumen** is defined as the luminous flux emitted in unit solid angle (steradian) by a uniform point source having a luminous intensity of 1 candela.

The **luminous intensity** ( $I_v$ ) is defined as the density of luminous flux with respect to solid angle in a specified direction

$$I_v = \frac{d\Phi_v}{d\Omega}$$

where  $\Phi_v$  is the luminous flux emitted in the given direction, and  $\Omega$  is the solid angle containing that direction. The luminous intensity is expressed in candela ( $cd = lm/sr$ ). The candela is one of the seven base units of the SI, meaning that all other units can be derived from them. One **candela** is defined as the luminous intensity, in a given direction, of a source that emits monochromatic radiation of frequency  $540 \times 10^{12}$  Hz and has a radiant intensity in that direction of  $1/683$  W/sr.

The **luminance** ( $L_v$ ) is defined as the density of luminous intensity with respect to projected area in a specified direction at a specified point on a real or imaginary surface

$$L_v = \frac{dI_v}{dA} \frac{1}{\cos\alpha}$$

where  $I_v$  is luminous intensity,  $A$  is area and  $\alpha$  is the angle between the normal to the surface at the specified point and the given direction. The luminance is expressed in candela per square metre ( $cd/m^2 = lm/m^2sr$ ).

The **illuminance** ( $E_v$ ) is defined as the density of incident luminous flux with respect to area at a point on a real or imaginary surface

$$E_v = \frac{d\Phi_v}{dA}$$

where  $\Phi_v$  is luminous flux and  $A$  is the area on which the luminous flux is incident. The illuminance is expressed in lux ( $lx = lm/m^2$ ).

### 2.3.2. Device efficiency and reliability

To accurately quantify the efficiency and reliability of the light emitting devices several aspects must be taken into account such as the eye response and the viewing angle [123]. The normalized photopic human eye response is described by the spectral shape  $V(\lambda)$  mentioned in the previous paragraph. The light emitting devices comprise both organic and inorganic layers with for each layer a

specific set of optical properties, like the index of refraction, forming complex optical cavities themselves where photons are scattered in random directions [128]. Therefore the emission pattern of the devices differ from that of an ideal Lambertian emitter, also resulting in a modulated emission spectrum and device efficiency.

Also here the distinction can be made between the radiometric quantities and their eye response-weighted equivalents or in other words the photometric quantities as seen in Table 5.

An often used photometric efficacy is the **luminous power efficiency** of a source ( $\eta_P$ ) that is defined as the quotient of the luminous flux emitted by the power consumed by the source

$$\eta_P = \frac{\varphi_{tot}}{I \cdot V}$$

where  $\varphi_{tot} = \int_0^\infty K(\lambda) \cdot P_{OLED}(\lambda) \cdot d(\lambda)$  and  $K(\lambda) = 683 \cdot V(\lambda)$ . The luminous power efficiency is expressed in lumen per Watt (lm/W).

Another photometric efficacy is the **current efficiency** ( $\eta_L$ ) can be seen as the equivalent to  $\eta_{ext}$ , which is discussed below, with that difference that it takes into account all incident photons according to the response of the eye, while  $\eta_{ext}$  also weighs the photons emitted in the invisible spectrum. The current efficiency ( $\eta_L$ ) is defined as the ratio between the emitted light and the electric flux

$$\eta_L = \frac{AL}{I}$$

where L is the luminance of the OLED (in cd/m<sup>2</sup>) and A is the device active area, which is not necessarily equal to the area of light emission. The current efficiency is expressed in candela per ampere (cd/A).

The radiometric power efficiency or **wall plug efficiency** ( $\eta_{W,W}$ ) is wavelength-independent and is defined as the ratio of the total optical power emitted to the electrical power injected.

$$\eta_{W,W} = \frac{P}{I \cdot V}$$

Another distinction can be made between the internal and external quantum efficiency.

The **internal quantum efficiency** (IQE or  $\eta_{int}$ ) is defined as the ratio of the total number of photons generated within the structure to the number of electrons injected.

The **external quantum efficiency** (EQE or  $\eta_{ext}$ ) is defined as the ratio of the number of photons emitted by the light emitting device into the viewing direction or into all directions (depending on the measurement set-up with or without integrating sphere) to the number of electrons injected.

$$\eta_{ext,quantum} = \frac{N_{photon}}{N_{carriers}}$$

The difference between the internal and external quantum efficiencies is the fraction of light coupled out of the structure into the viewing direction or into all directions ( $\eta_c$ ).

$$\eta_{ext,quantum}(\lambda) = \eta_{int,quantum}(\lambda) \cdot \eta_c(\lambda)$$

### 2.3.3. Recent published performances

#### ACPEL device

Shuai Zhang et al. produced colour tuneable hybrid ACPEL devices with organic fluorescent materials [129]. After screen printing the ACPEL devices on ITO coated PET substrates a mixture of F8BT and DCJTb was drop casted over the devices. A luminance of 110 cd/m<sup>2</sup> at a frequency of 2000 Hz and an operating voltage of 110 VAC was acquired for a reversed device with a F8BT and DCJTb mixture of 1.20 mg/mL and a F8BT:DCJTb mass ratio of 3:1.

A screen printed flexible ACPEL device on a PET substrate based on an Ag nanowire electrode was demonstrated by K.W. Park et al. [130]. A luminous power efficiency of 1.53 lm/W at 200 VAC and 400 Hz was measured.

#### OLED

Maria Gioti et al. conducted a study on the optical properties of emitting polymers used in efficient flexible OLED devices [131]. OLEDs using different emitting polymers were gravure printed on an ITO coated PET substrate. A device with lab synthesized yellow-light emitting polyether containing bis(styryl) anthracene units showed the highest luminance of approximately 365 cd/m<sup>2</sup> at an operating voltage of 10 VDC and a current density of 350 mA/cm<sup>2</sup>. Of the OLEDs produced with commercially available light emitting polymers the PFO OLEDs scored the best with a luminance of about 200 cd/m<sup>2</sup> at 10 VDC and 750 mA/cm<sup>2</sup>.

Multi-solution process flexible white SM-OLEDs fabricated with a stamping technique were presented by Yu-Sheng Tsai et al. [132].

**Table 5**

Overview of mostly used radiometric and photometric efficiencies.

Radiometric		Photometric	
Quantity	Unit	Quantity	Unit
Radiant efficiency ( $\eta_E$ )	W/(sr.A)	Current efficiency ( $\eta_L$ )	cd/A
Power efficiency (wall plug efficiency) ( $\eta_{W,W}$ )	W/W	Luminous power efficiency (luminous efficacy, luminosity) ( $\eta_P$ )	lm/W

These devices reached a luminance of 1062 cd/m<sup>2</sup> and a current efficiency of 5.57 cd/A at a current density of 20 mA/cm<sup>2</sup>.

Tobias Bocksrocker et al. presented a simple method to fabricate highly conductive and flexible metal/polymer hybrid anodes for efficient OLEDs on a PET substrate [59]. A device with an optimized Au grid as anode showed a luminance of 10000 cd/m<sup>2</sup> at an operating voltage of 8 VDC and a current density of 200 mA/m<sup>2</sup> and reached a luminous power efficiency of 7.2 lm/W at 3.5 VDC.

Deepak Kumar Dubey et al. built a solution processed highly efficient flexible white OLED on a polyethylene naphthalate (PEN) substrate with a current efficiency of 36 cd/A, a power efficiency of 18.7 lm/W and a 12.5% external quantum efficiency at 1000 cd/m<sup>2</sup> with a maximum luminance of 9586 cd/m<sup>2</sup> [133].

To obtain an improved hole injection ability, a lower driving voltage and an augmented device performance, Nan Zhang et al., inserted the composite hole injection layers (c-HILS) PTAA/AgNWs/PTAA into a solution processed blue flexible OLED on PET substrates [134]. Their devices showed a maximum luminance of 6051 cd/m<sup>2</sup> at 9 V, a maximum current efficiency of 1.76 cd/A, a low turn-on voltage of 2.7 VDC at 1 cd/m<sup>2</sup> and a driving voltage of 6.0 VDC at 1000 cd/m<sup>2</sup>.

Various desired designs of a patterned insulating layer in conductive AgNW bottom electrodes for flexible OLEDs were investigated by Sunho Kim and Byungil Hwang using a pre-patterned dry film photoresist (DFR) followed by a single hot-pressing step [135]. The fabricated devices demonstrated a maximum luminance of 27.341 cd/m<sup>2</sup> at 6.5 VDC and a turn-on voltage of 2.8 VDC. Moreover a paper-based OLED using the DFR insulator as a planarization layer was made presenting a turn-on voltage of 3.5 VDC and a maximum luminance of 157 cd/m<sup>2</sup> at 7.5 VDC.

#### LEC

Piotr Matyba et al. reported fully solution processed flexible and metal-free LECs with transparent electrodes [136] with a turn-on voltage of 2.8 VDC, an efficiency of 2.4 (4.0) cd/A at a brightness of 100 (50) cd/m<sup>2</sup>.

By optimizing the composition of the active layer and installing a specific operational protocol Junfeng Fang et al. improved the lifetime of a LEC to more than one month at a brightness exceeding 100 cd/m<sup>2</sup> and a power conversion efficiency of 2 lm/W [137].

Solution fabricated flexible blue-green and white LECs based on a cationic iridium complex were fabricated by Jiaqi Wu et al and showed an efficiency of 10.71 cd/A at 5 VDC and 9.8 cd/A at 7 VDC [138].

Fang-Cheng Liang et al. demonstrated a flexible textile-based LEC with a transfer printed AgNW-PDMS electrode [139]. The devices reached a luminance of 58 cd/m<sup>2</sup> at 7 VDC under 10% linear strain without any change to the electroluminescence properties.

Flexible LECs were fabricated on two types of paper (copy paper and specialty paper) substrates by Amir Asadpoordarvish et al. using a handheld airbrush in ambient conditions [140]. The specialty paper device showed a luminance of 200 cd/m<sup>2</sup> at 11 VDC, a current efficacy of 1.4 cd/A and a power conversion efficacy of 0.4 lm/W during a long-term operation at a current density of 14 mA/cm<sup>2</sup>. The copy paper device displayed a luminance of 35 cd/m<sup>2</sup> at 20 VDC, a current efficacy of 1.2 cd/A and a power conversion efficacy of 0.2 lm/W at a current density of 3 mA/cm<sup>2</sup>.

## 2.4. Conclusion

A resume of all the devices performances listed above can be found in Table 6. An important note is that these numbers apply to

**Table 6**

Overview of device performances of flexible light emitting devices.

Device performance	ACPEL device	OLED	LEC
Luminance	110 cd/m <sup>2</sup> [129]	365 cd/m <sup>2</sup> [131] 200 cd/m <sup>2</sup> [131] 1062 cd/m <sup>2</sup> [132] 10000 cd/m <sup>2</sup> [59] 9586 cd/m <sup>2</sup> [133] 6051 cd/m <sup>2</sup> [134] 27.341 cd/m <sup>2</sup> [135] 157 cd/m <sup>2</sup> [135]	100 (50) cd/m <sup>2</sup> [136] 100 cd/m <sup>2</sup> [137] 58 cd/m <sup>2</sup> [139] 200 cd/m <sup>2</sup> [140] 35 cd/m <sup>2</sup> [140]
External quantum efficiency		12.5% [133]	
Luminous Power Efficiency	1.53 lm/W [130]	7.2 lm/W [59] 18.7 lm/W [133]	2 lm/W [137] 0.4 lm/W [140] 0.2 lm/W [140]
Current efficiency		5.57 cd/A [132] 36 cd/A [133] 1.76 cd/A [134]	2.4 (4.0) cd/A [136] 10.71 cd/A [138] 9.8 cd/A [138]
Current density		350 mA/cm <sup>2</sup> [131] 750 mA/cm <sup>2</sup> [131] 20 mA/cm <sup>2</sup> [132] 200 mA/m <sup>2</sup> [59]	1.4 cd/A 1.2 cd/A
Driving voltage	110 VAC [129] 200 VAC [130]	10 VDC [131] 8 VDC [59] 9 VDC [134] 6.5 VDC [135] 7.5 VDC [135]	5 VDC [138] 7 VDC [138] 7 VDC [139] 11 VDC [140] 20 VDC [140]

devices on flexible substrates. Higher performances can be reached for devices on rigid substrates, but are not within the scope of this article.

All the above explains the preference of one of the devices above the other for a certain application. The mechanisms at work for the ACPEL device are fully understood, making it easier to find innovative or alternative materials and techniques to enhance the device performance. The device on the other hand, requires an AC driving voltage of about 100 V, which is hazardous for the human body, excluding its use in wearable applications. Its simplicity in structure and deposition, for example by the screen coating technique, makes it however popular for commercial indoor and outdoor applications or simple displays. The table shows that its luminance and luminous power efficiency are significantly lower than the OLED, but for its suitable applications this does not form an obstacle. The OLED demonstrates by far the highest luminance and luminous power efficiency and the lowest driving DC voltage. Especially the latter opens a range of applicable options in smart textiles going from lighting integrated in clothing for aesthetic or safety purposes to wound dressing for light or biomodulation therapy. Additionally, thanks to its high luminance, it is very popular as surface lighting on a variety of flexible or bendable substrates of all kind of shapes and sizes such as in the automobile sector. The OLED's luminance mechanisms are well understood, but its complexity and the obligatory use of a high barrier encapsulation add substantial difficulties to the equation keeping the lifetime relatively low. This is where the LEC comes in with its uncomplicated three layer structure and the possibility to use more chemically stable materials. Its luminance and luminous power efficiency are comparable to that of the ACPEL device, but it functions with similar DC voltages as that of the OLED. However the working principles of the LEC are still not completely identified which enhances the difficulty of finding techniques or materials to improve the lifetime, keeping the luminous performances also relatively low.

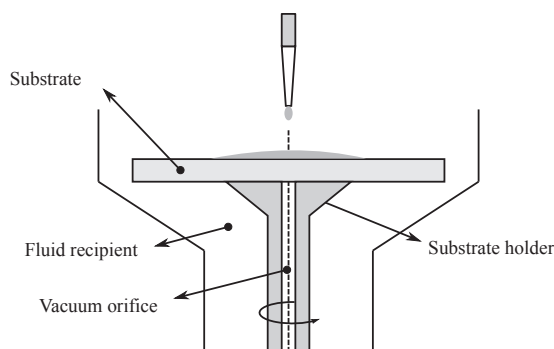
### 3. Printing and coating techniques

Several printing and coating techniques can be applied to deposit the layers of the light emitting devices. Some of these techniques, such as spin coating and vacuum deposition, are suited for lab scaled fabrication. Upscaling to mass production on the other hand, requires the use of roll-to-roll printing techniques as spray coating and inkjet printing. The techniques that are frequently used to apply light emitting devices are discussed more extensively.

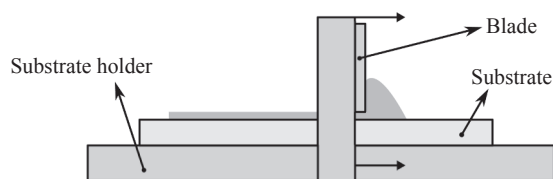
#### 3.1. Spin coating

The most commonly used thin film deposition technique is spin coating [141–143] (Fig. 14). Layers with a thickness from a few nm up to a few  $\mu\text{m}$  can be deposited, but the technique is widely used for layer thicknesses of 10–100 nm. Firstly a flat substrate is placed in the centre of the horizontal rotating sample holder and held in position by a small vacuum underneath the sample. The solution is dispensed in the centre of the substrate until the entire surface is covered. The sample holder is then rotated at a high speed and acceleration. Sometimes it is preferred to first rotate the sample and then dispense the solution. Under influence of the centrifugal force a smooth and uniform thin film is formed. Usually a volatile solvent is used that evaporates when the sample is rotated. The thickness of the layer depends on the spin coated solution (concentration, viscosity, surface tension, ...), the speed, the acceleration and the time of spinning.

Spin coating is a widely spread and fast lab-scale technique, but it is not suited for roll-to-roll production. Only 10 % of the spin coated solution forms the actual coated layer, which makes the other 90 % waste. Furthermore the restricted choice of substrate (small, flat, rigid and squared) and materials limits the use of the technique for industrial purposes. When a flexible substrate is spin coated, it has to be attached to a rigid backplane to prevent bending. For research purposes the spin coating technique is often applied to deposit the polymer layers of OLEDs and LECs.



**Fig. 14.** Spin coating. A flat substrate is placed on a sample holder and rotated at a high speed and acceleration. Under influence of the centrifugal force a smooth and uniform thin layer is formed.



**Fig. 15.** Blade coating. An ink is applied in front of a sharp blade that is placed at a certain distance of a substrate. The blade is then linearly moved across the substrate resulting in a wet film behind the blade.

### 3.2. Blade coating

Blade coating [132,144–151] (Fig. 15), also known as doctor blading or knife coating, is a roll-to-roll compatible coating technique to deposit layers with a well-defined thickness (typically 10–500  $\mu\text{m}$  [151]) on large area substrates. The set-up is quite simple and consists of a sharp knife blade that is placed at a certain distance from the substrate. An ink is then applied right in front of the blade. For research purposes or lab-scaled processing the blade is linearly moved across the substrate resulting in a wet film behind the blade and removing all excess ink. For continuous processing however, the blade is kept stationary and the substrate passes through a slit between the blade and a support roller. The final thickness of the layer depends on the distance between the knife and the substrate (roughly half of that distance [151]), the speed of the knife, the viscosity of the ink, the applied amount of ink, the surface energy of the substrate and the surface tension of the ink.

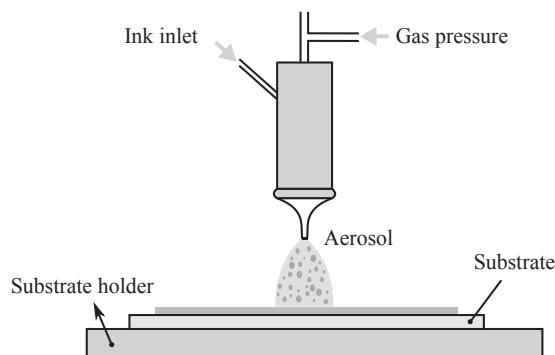
Compared to spin coating this technique is less accurate, but it has a very low coating waste since almost all excess ink can be recycled. It does take some practice by the operator to optimize the parameters and obtain the wanted layer thickness. There are many variants of this process making it more suitable to apply thin organic films. With the blade spin technique [153] the substrate is spun around after deposition by blade coating to remove excess coating and dry the wet film. Another variant is the continuous blade coating method developed for multilayer large area application of OLEDs and solar cells [153]. The substrate is heated from the bottom by a hotplate and hot wind is applied from the top after blade coating the surface. Consequently the solvent evaporates immediately and dissolution of the coating is prevented. The blade coating technique is often employed to apply the relatively thick layers ( $\mu\text{m}$  range) of the ACEPL and LEC devices.

### 3.3. Spray coating

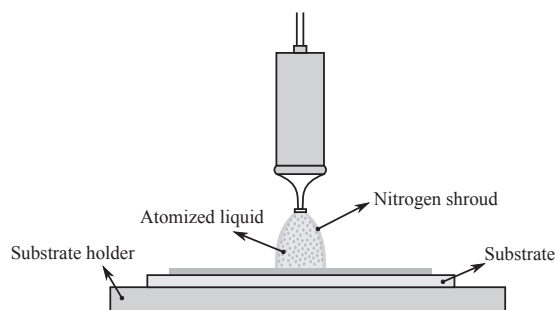
Spray coating is a high-throughput large-area deposition technique that is often used for industrial coating purposes. Both solutions and dispersions (further referred to as inks) can be deposited into layers with nano- or microscale thickness. These layers are formed by atomizing the ink at the nozzle of the spray head into a continuous flow of micro-sized spherical droplets. Within this coating technology there are three commonly available methods: pneumatic-based spray coating [154,155], ultrasonic spray coating [156–158] and electro spray coating [159,160].

In the pneumatic-based method (Fig. 16), the liquid bulk is impacted with a high-velocity gas, creating high frictional forces over the liquid surface and breaking it down into small droplets with a size of about 10  $\mu\text{m}$  [155]. A well-known example of this technique is the airbrush gun [161].

A more advanced method is ultrasonic spray coating (Fig. 17) where the atomization is realized by the vibrations produced by the mechanical expansion and contraction of piezoelectric transducers inside the nozzle. A liquid that is fed into the device will form capillary waves due to this vibrational energy. These waves emerge onto the atomizing surface where the kinetic energy is dissipated and the liquid is broken down into a spray of micrometre sized droplets. This approach has many advantages over the conventional spray deposition including smaller and more consistent droplets, a more directed spray by usage of an inert gas, a uniform coverage



**Fig. 16.** Pneumatic spray coating. A liquid bulk is impacted with a high-velocity gas, creating high frictional forces over the liquid surface and breaking it down into small droplets.



**Fig. 17.** Ultrasonic spray coating. The atomization is realized by vibrations produced by the mechanical expansion and contraction of piezoelectric transducers inside the nozzle. A liquid that is fed into the devices forms capillary waves due to this vibrational energy. These waves emerge onto the atomizing surface where the kinetic energy is dissipated and the liquid is broken down into a spray of micrometre sized droplets.

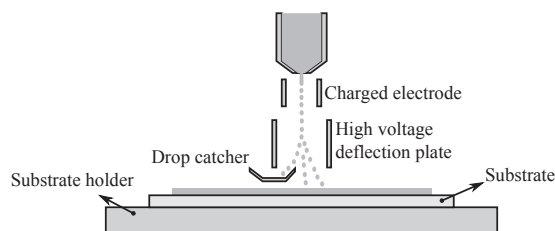
over large areas even for very thin films down to 5 nm and the possibility of using multiple spray nozzles for simultaneous deposition. A world leader in ultrasonic spray coating is the Sono-Tek Corporation [162], since the company's founder was the inventor of the ultrasonic nozzle. An alternative can be found in the aerosol jet technology produced by Optomec [163]. Here the ink is already ultrasonically atomized in a reservoir and the resulting aerosol with droplets of about 15  $\mu\text{m}$  in size is then transported towards the nozzle where it is driven to the substrate by a carrier gas. The extra benefit of this technique is the high lateral resolution of less than 10  $\mu\text{m}$  to create any micro-sized pattern.

Another state-of-the-art spraying method is electro spraying or electrohydrodynamic spraying where atomization of the ink is done using electrical forces. A capillary nozzle is kept at a high electrical potential so the ink flowing out of the nozzle is dispersed into small droplets under influence of the electrical field. This technique has some advantages over mechanical atomizers, such as the mono-disperse size distribution of the droplets that can be as low as nanometre scaled size and the possibility to control droplets size by adjusting the flow rate and the voltage of the electrical potential. Because the droplets are electrically charged, their motion (deflection and focus) can be guided by means of an electric field. Furthermore they are self-dispersing in space resulting in a higher deposition efficiency.

The atomization process has a lot of parameters to adjust: viscosity and density of the ink, flow rate, power, coating speed, droplet size, gas flow properties, nozzle design, working distance, hotplate temperature, surface energy of the substrate and the amount of spray coated layers on top of each other. By tuning all of these parameters a homogenous layer with the desired thickness can be obtained. Once the optimal spray coat parameters are set, it is a fast process with a minimum of ink waste which is easily upscalable for roll-to-roll applications. It is also suitable for all kind and shapes of substrates. Spray coating can be performed with a wide variety of inks – both nanoparticle and suspensions – spanning a very broad viscosity range. However, it is mainly used for coating and, apart from the aerosol jet printing technique, cannot achieve very narrow resolutions. The complete stacks of the LECs [140] and ACEP devices can be applied by spray coating. It however remains a challenge to spray coat an entire OLED stack, mainly due to the high requirements of roughness and thickness of the layers.

### 3.4. Inkjet printing

Inkjet printing [164,165,167–169] is a contactless printing technique in which a digital image is recreated by the ejection of ink droplets onto a substrate. This technique is mainly situated in the publishing industry, but its popularity is growing in the thin film device industry thanks to its many advantages such as fast printing, cost-effectiveness and low temperature fabrication. Furthermore it has a first-rate compatibility with both organic and inorganic materials and is often used to print highly conductive paths and patterns. Two methods of operation for inkjet printing can be distinguished: continuous inkjet printing (CIJ) and drop-on-demand inkjet printing (DoD).



**Fig. 18.** CIJ method inkjet printing. Piezoelectric vibrations create a continuous flow of ink that is broken down into droplets at the print head nozzle due to harmonic modulation.



In CIJ systems (Fig. 18) piezoelectric vibrations create a continuous flow of ink that is broken down into droplets at the print head nozzle due to harmonic modulation. To create a pattern individual drops are mostly navigated by means of binary or variable electrostatic deflection. Excessive droplets are collected and recycled which makes the system more complex because of the need for pumps and ink purification. The droplets have a size between 80 and 100  $\mu\text{m}$  and travel towards the substrate with a speed of about 20 m/s [169]. On a substrate the droplets diameter is about twice the size of that of a released droplet. The CIJ method has some advantages over the DoD method such as the possibility of using inks with a lower viscosity, a higher drop velocity and thus higher print speeds, but it is less interesting economically since more ink has to be used and it is difficult to control individual droplets.

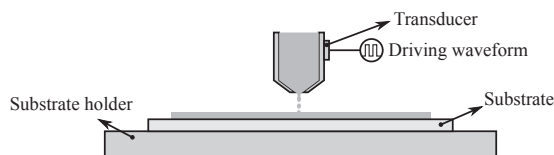
In the DoD method (Fig. 19) droplets are thermally, piezoelectrically, electrostatically or acoustically actuated and are only jetted on demand by applying a (digital) signal or waveform, eliminating ink waste. It is possible to control the ejection of ink droplets through each nozzle [76]. This method is widely spread because of its high accuracy, controllability and cost-effectiveness. With thermal activation a pulse of current passes through a heating element that is inside each chamber of the cartridge causing a rapid vaporization of the ink. Bubbles of ink vapour are formed that force the ink drops to leave the nozzle. The droplets can be as small as 0.5  $\mu\text{m}$  for water and 1  $\mu\text{m}$  for methanol and toluene [167]. The process reaches temperatures up to 300  $^{\circ}\text{C}$  causing the evaporation of water at the nozzle's surface and consequently clogging of the nozzle, making it unsuitable to print biological inks and polymers. This limits the use of thermal DoD to applications such as desktop printing with mostly aqueous inks. The piezoelectric activation utilizes piezoelectric elements placed at the back of each ink reservoir. By applying a voltage the elements change shape, resulting in a pressure wave forcing smaller droplets of 10–20 pL to eject the nozzle. Since printing of both aqueous- and UV-based inks on a variety of substrates is amongst the possibilities, piezoelectric DoD is often applied for research and industrial purposes. It is more suited for printing aqueous inks with conjugated polymers since the process does not require high temperatures. Both thermal and piezoelectric actuated systems however display some limitations in the density (and size for the thermal actuated systems) of the nozzle array and in the ejection frequency due to respectively thermal problems and physical limitations. A possible answer to these limitations are the electrostatically actuated systems where the liquid is directly manipulated by an electric field [170–173,286]. A liquid is placed inside an electric field and an electric charge is induced at the surface of the liquid's meniscus. This will cause an electric stress that will stretch the meniscus in the direction of the electric field and into a sharp cone under influence of an electrostatic field. When the electrostatic forces surpass the surface tension forces, charged liquid uniform droplets are ejected from the nozzle. The ejected droplets can be smaller than the nozzle size resulting in a higher resolution than that of the piezoelectric inkjet printers and more viscous inks can be printed. Currently however this method is not widely commercially employed. The most recently developed method is the acoustic inkjet printing method in which droplets are generated using acoustic energy [173,175,286]. An ink droplet is generated by focussing an ultrasound beam with an acoustic lens on the fluid surface. The size of a droplet can be dynamically varied by adapting the fluid-to-transducer distance, clogging of the nozzles is not possible and the viscosity range is broader.

Commercially available printers can print lines with a width of 50  $\mu\text{m}$  and a minimum spacing of 20  $\mu\text{m}$ . However more advanced inkjet technologies can go down to line widths and spacing between 3 and 20  $\mu\text{m}$  [176]. This immediately shows one of the most appealing features of this technology namely the possibility to print very precise patterns and images with high precision. Other advantages are a high printing speed, a very low ink waste (mainly for DoD) and a roll-to-roll compatibility. The inkjet printing technology is however limited to flat substrates and to a certain film thickness. Further it requires a rather complex ink formulation with a certain evaporation rate of the solvents and a well-tuned dynamic viscosity. The inkjet printing technique is thus highly suitable to print almost every OLED [177–179], LEC [180–182] or ACPEL device [17,183] layer depending on the required thickness. A remaining challenge lies in the inkjet printing of transparent layers, but this can be surpassed by printing very narrow arrays or grids.

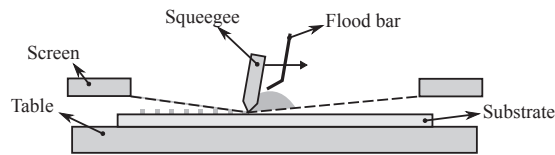
### 3.5. Screen printing

The screen printing technique (Fig. 20) dates all the way back to the cave paintings in the Stone Age and is basically a highly developed form of stencilling [185,283,287,288]. This relatively simple and low-cost technique uses a screen formed by a woven mesh of synthetic fabric or metal wires. The screen is placed above the substrate and the mesh of the screen is brought into line contact with the substrate before applying the ink on top of the screen. The distance between the screen and the substrate or the snap-off needs to be perfectly adjusted to obtain clear prints; too low and the ink will already touch the substrate before printing, too high and the ink release will not be optimal. A fill blade or squeegee is moved across the screen, pushing ink through the open area of the mesh forming the pattern. The surplus is removed by the edge of the squeegee or the flood bar leaving a thick, micrometre range layer on the substrate. Although relatively simple the technique requires some practice to optimize the many variables such as the mesh-size of the screen, the angle and pressure of the squeegee, the printing speed and the viscosity of the ink.

Due to its limitations in resolution, screen printing is often used in the graphic arts world for low-information-content graphics like



**Fig. 19.** DoD method inkjet printing. Droplets are thermally, piezoelectrically, electrostatically or acoustically actuated and are only jetted on demand by applying a (digital) signal or waveform, eliminating ink waste.



**Fig. 20.** Screen printing. A screen formed by a woven mesh of synthetic fabric or metal wires is placed above a substrate and an ink is applied on top of the screen. A fill blade or squeegee is moved across the screen, pushing ink through the open area of the mesh forming the pattern.

signage, textiles and promotional work. Sporadically this technique is used to deposit OLED layers [187,188], but it is mainly known as the signature application technique for EL devices [11,189,190].

### 3.6. Pad printing

While most printing or coating techniques are limited to rigid and planar substrates, pad printing (Fig. 21) is not restricted by the shape of a substrate and can deposit layers on convex and concave surfaces [152,191]. It is an off-set printing technique where a pattern or layer of ink is transferred from a gravure plate to a substrate using a silicone rubber stamp. An ink reservoir is employed to apply the ink on the gravure table, making it possible to use relatively volatile solvents since the ink is only momentarily exposed to the atmosphere. Similar to screen printing this technique also demands a lot of experience to optimize or control the different variables such as the surface energy of the substrate, the stamp and surface tension of the ink, the hardness of the silicone rubber, the shape of the pad, the depth of the gravure, the positioning and speed of the pad and the applied force. Depending on the viscosity, quite a large amount of ink is necessary to acquire a good quality print. The pad printing technique has been used to apply polymer layers in solar cells [152,191].

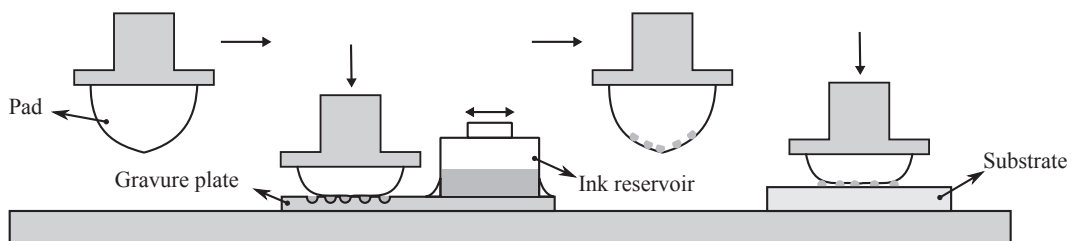
The Karlsruhe Institute of Technology (KIT) developed a variant of this pad printing process in cooperation with the company Franz Binder GmbH & Co to print electroluminescent layers on three-dimensional objects [192].

### 3.7. Slot-die coating

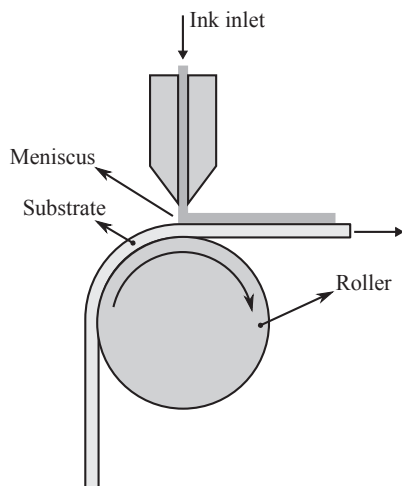
Slot-die coating (Fig. 22) is a non-contact premetered coating technique for large-area coating of thin and uniform films in web and cross direction [151,194]. The coating process itself is rather simple, but the slot-die head is complex and precision engineered. A uniform flow of coating is provided by a metering system into the slot-die head that distributes the coating onto a substrate. The head actually approaches the substrate that is moving across a roller or a linear stage until a meniscus is formed between them. The distribution of the coating, and consequently the coating thickness and uniformity, is controlled by the slot-die head more specifically by the dimensions of the internal cavity and opening and the process parameters. For roll-to-roll, non-patterned and large-area devices, this technique is suitable to apply the layers of all three light emitting devices [18,76,150,151,194–196].

### 3.8. Gravure coating

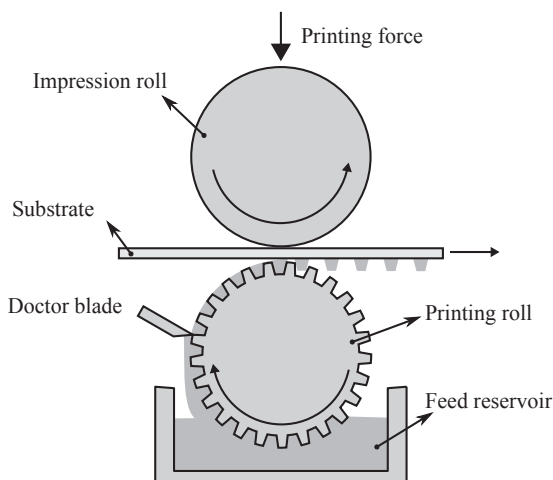
A roll-to-roll technique that does allow the printing of patterns is gravure coating [151,197] (Fig. 23). More complex variations of this technique are being developed but in its simplest form it includes a two roller system. A coating roller with an engraved pattern (or gravure roller) is submerged in an ink reservoir, filling up the gravure with ink. It is then turned until the gravure reaches a web or substrate that is clamped between the gravure roller and a support roller. Before reaching the web, the roller passes by a doctor blade that removes excess ink. Consequently the ink is transferred from the gravure roller to the web. This gravure technique is quite fast, the web speed can be up to 15 m/s [151,197]. Furthermore many types of solvents can be used. Changing patterns however requires engraving of a new roller, which is a time consuming and expensive process. Another possible limitation is the high viscosity of the inks (between 50 and 200 mPa [198]) that can be used for this technique. Fortunately the surface tension of the inks also plays an important part in the process of obtaining uniform layers, allowing the usage of lower viscosity inks. Mostly gravure coating is used in the world of magazines and packaging, but it is a very interesting technique for upscaling the production of the light emitting devices [198–203].



**Fig. 21.** Pad printing. A pattern or layer of ink is transferred from a gravure plate to a substrate using a silicone rubber stamp.



**Fig. 22.** Slot-die coating. A uniform flow of coating is provided by a metering system into the slot-die head that distributes the coating onto a substrate. The head actually approaches the substrate that is moving across a roller or a linear stage until a meniscus is formed between them.



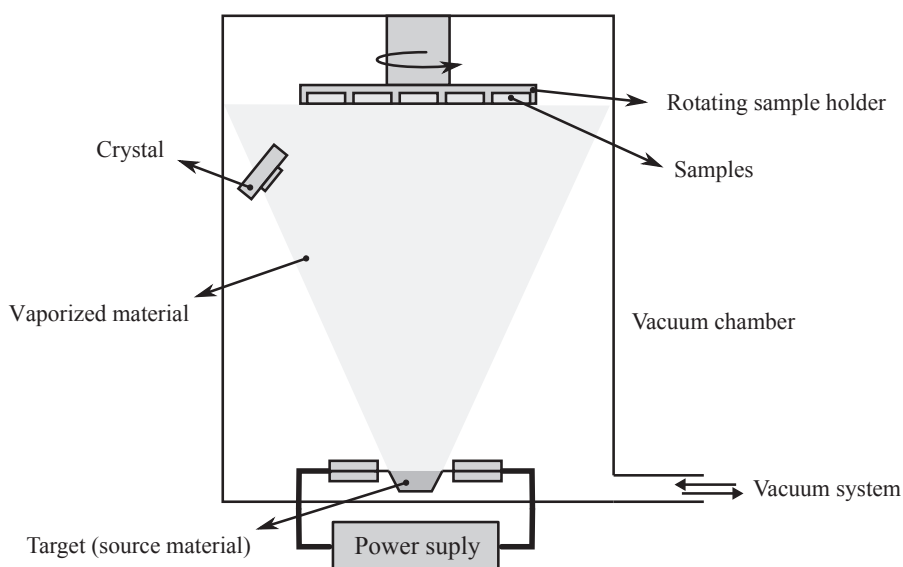
**Fig. 23.** Gravure printing. A coating roller with an engraved pattern (or gravure roller) is submerged in an ink reservoir, filling up the gravure with ink. It is then turned until the gravure reaches a web or substrate that is clamped between the gravure roller and a support roller. Before reaching the web, the roller passes by a doctor blade that removes excess ink. Consequently the ink is transferred from the gravure roller to the web.

### 3.9. Vapour deposition

There are two general vapor deposition techniques: physical vapour deposition (PVD) and chemical vapour deposition (CVD). This technology is not roll-to-roll nor is it a wet chemical deposition method and is quite expensive, but it is at the moment indispensable to apply certain metallic electrodes of the OLEDs. PVD includes a variety of vapour-phase technologies. It describes the vacuum deposition methods which use physical processes to deposit thin solid films by the condensation of a vaporized form of a solid material onto a substrate. Most commonly applied PVD processes are thermal evaporation and sputtering [141,204,279]. Thermal evaporation (Fig. 24) involves resistance heated intermetallic boats that are used as a coating source. Consequently only metals with a relatively low melting point, for example aluminium and silver, can be used for this technique.

During the sputtering process, on the other hand, a solid target of the to be coated material is bombarded with ions extracted from a concentrated plasma cloud. This causes the ejection of high-energy atoms from the target surface towards the substrate. This technique can be used to deposit all pure metals and alloys and most oxides and nitrides.

CVD is also used to deposit thin films, but in contrast to PVD, the deposition of a layer is the result of a chemical reaction in the vapour phase on the heated substrate surface [205,279]. A mixture of gases interacts with the substrates surface at a relatively high temperature. This results in the decomposition of some of the constituents of the gas mixture forming a thin solid film on the substrate.



**Fig. 24.** Thermal evaporation. Resistance intermetallic boats are used as a coating source to deposit thin solid films by condensation of a vaporized form of a solid material onto a substrate.

### 3.10. Conclusion

All printing techniques are eligible for continuous manufacturing, are economically viable and can be environment friendly. They differ in the following specifications: ink formulation, resolution and size of the printed features, the possible layer thickness and roughness and manufacturing time. An overview of all mentioned printing techniques and their deposition of the different layers of the light emitting devices can be found in [Table 7](#).

The ACEPL device requires a simple application technique that has the possibility to obtain micrometre thick smooth layers. The device is almost exclusively deposited using the screen printing technique. This fast and simple technique is perfectly fit to deposit the micrometre thickness layer stack of the device. Other well-matched printing techniques are blade coating, pad printing, slot-die coating and gravure coating, but since the screen printing technique is so widely spread they are less used. Other mentioned printing techniques are within the possibility to apply the devices, but are too precise or more appropriate to deposit thin nanometre range

**Table 7**

Overview of printing and coating techniques used to deposit layers of the light emitting devices.

Device layer	Spin coating	Blade coating	Spray coating	Inkjet printing	Screen printing	Pad printing	Slot-die coating	Gravure coating	Vapour deposition
ACEPL device									
Transparent electrode					[206]	[207]	[18]		[208]
Phosphor	[208–211]	[212]			[206,213]	[207]	[18]		
Dielectric	[208–210,214]	[212]			[206,213]	[207]	[18]		
Binder material (dielectric and emissive layer)	[209]				[213]	[207]			
Back electrode				[215]	[206]	[207]			
OLED									
Transparent electrode (TCM)		[216]	[217]	[218]		[219,220]			[39,46,51,206]
Buffer layer	[221]								[39,46,51]
Hole transport layer	[39,67,206]		[46,217]	[178]				[199]	[33,39,46,51]
Emissive layer	[206]		[158]					[199]	[33,39,46,51]
Electron transport layer									[33,39,46,51]
Back electrode				[206]					[33,39,46,51]
LEC									
Transparent electrode			[140]				[195]		
Hole transport layer	[137]		[140]		[222]		[195]		
Active layer	[137,222]		[140]				[195]		
Electron transport layer			[140]				[195]		
Back electrode			[140]				[195]		[137,222]

layers.

At lab scale, spin coating is often the go-to technique to apply the polymer layers of the OLED and vapour deposition to deposit the metal and small molecule layers. Both techniques are not roll-to-roll applicable, nor ecologic (spin coating has a lot of ink waste) nor economically viable (vapour deposition is a very expensive technique), hence the need for printing techniques. OLED layers however lie in the nanometre range thickness and demand a very low roughness of less than 5 nm to avoid electrical shorts or preliminary degradation. Therefore only spray coating, inkjet printing and gravure coating are acceptable to apply the thin polymer layers. The electrodes, which have a thickness of about ten times the polymer layers, can be also deposited using blade coating, screen printing, pad printing or slot-die coating.

The LEC also provides an answer to the extremely thickness and roughness requirements of the OLED. The device only consists of two electrodes and an active layer that can be thicker than the active layer of the OLED. All suggested printing techniques are qualified to deposit the complete layer stack of the LEC.

## 4. Discussion and future prospects

### 4.1. Issues and challenges

Even though the advantages are numerous, the light emitting devices still struggle with significant challenges that might limit their use in certain applications. A concise overview and explication of these hurdles is given in this paragraph.

#### 4.1.1. ACPEL device

Over time a reduced electroluminescent performance of the ACPEL devices is observed that is generally assumed to be caused by two types of material degradation: the absorption of water vapour or moisture by the emissive layer and the internal structural change within the ZnS grains [282].

Water vapour or moisture infiltrates the emitting layer and causes the electrolytic decomposition of ZnS and the generation of free zinc metal which results in a grey discoloration of the grains. By applying an adequate barrier layer this problem can be solved. For example R. Withnall hermetically sealed every grain with a SiO or Al<sub>2</sub>O<sub>3</sub> coating leading to an extent of the useful half-brightness lifetimes of the devices up to 3000 h [11].

The understanding of the second degradation mechanism on the structural change within the ZnS grains is very incomplete and requires more investigation. Under continuous AC excitation it is noticed that the electroluminescence of the devices decreases over time and that this decline occurs even faster at elevated temperatures. There is a consensus that it could comprise some type of diffusion process of Cu in and around Cu<sub>x</sub>S, moreover two different models are suggested. The first model is derived from the Fischer based mechanisms behind the luminescence and indicates the diffusion of Cu in the ZnS lattice due to the applied AC electric field [223]. During device operation, diffusion of copper ions or attraction of copper ions of the adjacent host crystal rounds the sharp tips of the copper sulphide needles. These tips provide a highly localized electric field in the phosphor region that excites luminescence. When they get blunted too much they reach a state where it is not possible to induce sufficient electric field for luminescence anymore. Alternatively Warkentin et al. suggest another model using the extended x-ray-absorption fine-structure (EXAFS) technique [224]. With this technique they investigated the mechanisms behind the electroluminescence and the eventual degradation of the electroluminescent materials ZnS:Cu,Cl and ZnS:Cu,Mn,Cl. The resulting EXAFS spectra identified the Cu<sub>x</sub>S precipitates as CuS instead of Cu<sub>2</sub>S and showed that these remain unchanged between new and degraded samples. Both isolated Cu ions and CuS clusters are located in the ZnS lattice. The Cu ions serve as effective recombination centres by trapping electrons, but the CuS clusters do not contribute directly to the luminescence. It is suggested that isolated EL-active Cu ions diffuse to CuS clusters or to the ZnS particle surface, resulting in less recombination centres and consequently less luminescence. This model is supported by the degradation and rejuvenation studies performed by Stanley et al. where they demonstrate almost complete rejuvenation of a degraded device by heating [225].

Besides the material degradation, the operating AC-voltage of an ACPEL device is quite high and varies between 50 and 200 V. This makes the device dangerous and not suitable for wearable applications [97].

#### 4.1.2. OLED

OLEDs struggle with some issues and challenges in the form of light-output losses and degradation mechanisms.

**4.1.2.1. Light-output losses.** The working mechanism of an OLED is characterized by light outcoupling losses that limit the external quantum efficiency to just 20% or even less, while the internal quantum efficiency is almost 100%. The causes of these losses can be found in the surface plasmon polaritons (SPPs) modes, waveguide modes, the substrate modes and electrode absorption [226–233].

When free charges at the surface of a metal, couple with electromagnetic radiation they form electromagnetic waves, or SPPs, that travel along the metal–organic (metal–dielectric) interface and that reach a maximum intensity at the interface and fade out exponentially into both surrounding media. On a planar surface they have a non-radiative nature indicating that the energy coupled to SPPs dissipates in the form of heat and quenches optical emissions. This decrease in light-output caused by SPPs can be reduced either by preventing the coupling of energy to SPPs or by partially recuperating the lost energy. Since the coupling occurs through the optical near field, an approach to avoid the coupling to SPPs is to increase the distance between the emissive layer and the cathode by increasing the layer thickness. This will however give rise to extra waveguided modes replacing the losses due to SPPs. Another and

more beneficial method is to adjust the structure of the device in such a way that the emitters are oriented so that their transition dipole moment is parallel to the substrate plane. This decreases the coupling of the horizontally oriented dipoles to SPPs considerably compared to the perpendicular dipoles and thus increases the outcoupling efficiency. To partially recuperate the energy coupled to SPPs several tactics have been suggested such as index coupling [234], prism coupling [235] and grating coupling [227,236,237]. The latter method was proven to be the most effective to be applied in OLEDs because of its high extraction efficiency of SPPs and because the recuperated power is outcoupled in the same direction as the light emission [238]. For this grating coupling technique periodic microstructures are applied to convert SPPs into photons.

The waveguide and substrate modes are related to Snell's law that gives the relationship between the angles of incidence and refraction for a wave passing through an interface between two media with different indices of refraction. Due to a large difference in the refractive index ( $n$ ) most of the light is captured (or in other words internally reflected) within the transparent electrode (waveguide modes) and the substrate (substrate modes) and finally lost due to absorption [226]. Various light extraction technologies and highly transparent electrodes offer a solution to improve the out-coupling efficiency of OLEDs. In their review Hong and Lee give an overview of developments in these extraction technologies and transparent electrodes [232].

**4.1.2.2. Degradation mechanisms.** In order to improve the lifetime, numerous researches on the degradation of OLEDs were conducted. In 2015 Sebastian Scholz et al. published an extended review paper on the degradation mechanisms and reactions in mostly SM- OLEDs [239]. This publication focuses on the visible appearances of the mechanisms and individual reactions behind them. With this in mind the mechanisms are divided into intrinsic and extrinsic degradation mechanisms depending on the factors that caused them. Device degradation gives rise to five main visual effects: dark spot growth, catastrophic failure, exponential short-term decay, exponential long term decay and initial luminance increase.

A dark spot is a small non-emissive region of a (nominally) active area of OLEDs. These spots can already arise during the production process and grow further throughout storage of the device in both ambient and nitrogen environment. Several degradation mechanisms are suggested to lead to dark spot formation, but mostly delamination of the cathode material seems to be the main cause. This was proven by Yoon-Fei Liew et al. by peeling off the existing cathode and replacing it with a new one, eliminating all dark spots [240]. During their study they also noticed that the nucleation of dark spots occurs during the deposition of the cathode at the organic/cathode interface and that changes in the organic layers at that interface also play a role in the growth of dark spots. The growth of dark spots might originate from grains clusters and stains, protrusion, gaps in the cathode, a broken cathode, pinholes and particles [241].

Catastrophic failure or the complete breakdown of a device is mostly the consequence of electrical short circuits caused by contamination or imperfections of the substrate surface, local heating effects or electromigration of the cathode metal during the operation of the device.

The classification of the degradation mechanisms indicates if the degradation is caused by factors outside or inside the device. Since extrinsic degradation mechanisms depend on factors outside the device, they are easier to investigate and often the result of impurities, light, oxygen, water, temperature and process parameters. Contamination by oxygen and water can cause numerous reactions in OLEDs: chemical reactions resulting in oxidation, hydrolysis of chelate molecules initiating various reactions of hydrolytic products, oxidation of organic materials (photo- and electrochemical reactions), charge transfer doping leading to the formation of filled traps and/or free charge carriers and morphological changes. To protect the OLEDs against water and oxygen a high barrier encapsulation is absolutely indispensable. An effective encapsulation requires a water vapour transmission rate (WVTR) and an oxygen transmission rate (OTR) lower than respectively  $10^{-6}$  g m<sup>-2</sup> per day and  $10^{-3}$  cm<sup>3</sup> m<sup>-2</sup> per day [49]. Especially for flexible OLEDs this is challenging since conventional encapsulation techniques are still not up to the task. Very important is also the exclusion of contaminations and impurities on the surface of the substrate and in the materials used. That is why proper cleaning of the substrates and filtering of the materials is indispensable. Furthermore the process parameters play a significant role in the characteristics of the device and the morphology of the different layers. Over time an operating OLED tends to demonstrate an increase in the drive voltage and a decrease in the luminance generally caused by an increased injection barrier at an interface or a decreased conductivity of a transport layer [242]. High temperatures cause a faster device degradation (Joule heating) and at a certain critical temperature even device failure. In general most of these external factors can be managed or regulated by improving the production conditions, for example working in a clean room environment and by applying a high barrier encapsulation.

More difficult to determine are the intrinsic degradation mechanisms. Following processes are known to cause intrinsic degradation: migration of ionic and other entities, charge accumulation, changes in the electric field profile caused by molecular reorientation, charge carrier reactions, exciton reactions, charge balance and position of the recombination zone, electrochemical reactions. Migration processes such as drift and diffusion contribute to the degradation of OLEDs. Diffusion of electrically neutral species through the layers of the device can cause exciton quenching or non-radiative recombination. The applied or built-in electrical field can initiate the drift of ions. A significant difference in charge injection barriers at the interfaces of the HTL or the ETL can lead to accumulation of excessive charges at those interfaces. The internal electric field formed during OLED operation is in the order of 1 MV/cm and can initiate the reorientation of molecular dipoles which alters the electric polarization of the layer and even possibly the devices' electric field profile [239]. This can have an effect on the recombination efficiency, the outcoupling efficiency and the excited-state quenching by charge carriers. When charge carriers come in the proximity of the recombination zone at densities in the order of  $10^{11}$ - $10^{12}$  cm<sup>-2</sup> they tend to quench excitons affecting the device efficiency. Another possibility is the interaction of the charges and specific materials that induce chemical reactions. Electrochemical processes near the electrodes, for example corrosion, can initiate numerous chemical reactions inside the device. Generally these reactions occur in the anode, the cathode and the organic layers. Comparable to the charge carriers also the excitons are of importance for the degradation of the device. The charge balance and the

position where the recombination takes place are critical to obtain high efficiencies and lifetimes.

#### 4.1.3. LEC

In general the proper working of the LEC is influenced by degradation mechanisms, side reactions, electrochemical stability and optimization of the recombination zone [76]. Usually the LECs display a loss in efficiency and luminescence quenching over time. In some cases these reactions can be partially reversed by relaxation of the cell as published by Sebastian B. Meier et al. [243]. They could obtain a partial recovery of the photoluminescence of the cells by turning off the bias voltage at different times.

Irreversible degradation of the LECs is caused by several processes, mostly dark spot formation [244,245] and side-reactions with the ion transport material PEO [109,246]. The dark spot formation is similar to that of the OLEDs. The spots diminish the emitting area and therefore also cause a reduction of the luminescence. Their growth can occur during operation time and during storage and they are believed to be the result of heavy doping and the presence of oxygen and moisture, although the precise cause has not been found yet. Side-reactions with the ion transport material PEO are another example of irreversible degradation. These reactions generally arise at the cathode interface and may block the injection of electrons causing an imbalance of charge carriers through the active layer. Such a hampering of the electrons can finally lead to micro-short, since the opposite doped region can grow from one electrode to the other. This can be avoided by using different materials to lower the conduction band of the conjugated polymers [247]. Different papers demonstrate the improvement of the electrochemical stability of the electrolyte and therefore the lifetime of the LECs [248–250].

To obtain a high recombination efficiency optimization of the recombination zone is crucial. Injected electrons and holes have to recombine in the active layer and should not pass that layer. To achieve this, the n-type and p-type doping near the electrodes has to be far enough from that recombination zone.

#### 4.2. State of the art and future prospects

The last paragraph discussed the issues and challenges that the light emitting devices are facing. In the past decades solutions have been brought forward and the devices have constantly been improved using innovative materials and new printing techniques suited for continuous manufacturing, being more environmental friendly and more economical. The devices also arouse interest of new and intriguing markets such as the smart textiles field where different functionalities are added to textiles to make them smart and intelligent. Nonetheless, a lot of the initial issues are still standing today and the increasing complexity of the devices (the shape, the size, the purpose, the operating environment, etc.) also brings forth new challenges and hurdles to be taken. This paragraph will argue the development of the light emitting devices starting from the invention, but with focus on the state of the art and the future prospects.

##### 4.2.1. ACEPEL device

The ACEPEL device is the most popular commercially available high-field EL device due to its high potential for illumination and display applications which mainly lies in the advantages of using flexible substrates and low cost roll-to-roll fabrications processes.

The first ACEPEL device was fabricated on a metal plate that also acted as an electrode, followed by an inorganic phosphor layer in an insulating medium, insulated between a thin rubber layer and an oil layer, and finished off with a transparent electrode consisting of a metal ring and NaCl/glycerin solution [4]. This first Destriau-model of an ACEPEL device was later converted to a four layer device built up with a thin metal electrode, a dielectric, a phosphor layer and a transparent conducting electrode. At current time even more simplified versions of the ACEPEL device are available where the phosphor is embedded in an insulating dielectric flexible binder.

In literature three eras of ACEPEL development are mentioned [282]. During the first era, that took place between the late 1950s and the first half of the 1960s, basic studies on the device were carried out where the focus lied on the fabrication of flat light sources. The interest in the devices was lost however due to their low brightness and low lifetime of powder which would last only a hundred to a thousand hours. The second era, between 1986 and the early 1990s was mostly dominated by progression in the DC powder EL and thin film EL research, however also the luminance and lifetime of the ACEPEL devices was improved. During this time the ACEPEL devices were often used as backlights for liquid crystal displays (LCDs). The third era commenced in the late 1990s and is mainly ruled by the flexible ACEPEL devices that are generally developed for backlight and lamp applications and architectural and decorative lighting [8, 17,282].

Researchers successfully applied the ACEPEL device on a variety of substrates. Often PET is used as a substrate, K. Park et al. established a flexible ACEPEL device on a Ag NW PET substrate [130]. T Bin Hu et al. inkjet-printed the transparent conducting polymers PEDOT:PSS onto a PET open-mesh fabric and used a precision dispensing system and Al foil to finish the rest of the ACEPEL device [17]. ACEPEL devices were fabricated on four different paper substrates, glossy paper, sticker paper, magazine paper, and newspaper, by Jin-Young Kim et al. [213] to protect the paper against the production process and increase the conductivity of the metal electrode a spin-on-glass layer was applied before depositing the other layers. ACEPEL devices might not be suited for wearable applications because of their high operation AC-voltage, but do fall into the scope of other smart textile application, such as interior and exterior design and visual merchandising. W. Deferme et al. screen printed and analysed different smart designs of the ACEPEL devices to maintain typical textile properties such as flexibility, breathability and drapability [206].

Progress in the performance of the devices is still made by incorporating innovative materials. Often carbon nano tubes (CNTs) are used to improve the device performances. By inserting shortened single wall CNTs (SWCNTs) between the phosphor and dielectric layers the EL performance, both efficiency and brightness were increased by Jin-Young Kim et al. [251]. Fang-Hsing Wang et al. added CNTs in the dielectric layer which resulted in a device with a low power consumption, a high brightness and an improved luminous efficiency [252]. Furthermore these ACEPEL devices on PET showed a superior flexibility and luminous reliability.

Mostly ACEPEL devices are fabricated using screen printing, doctor blading or spray coating, all low cost, easy to apply and roll-to-

roll compatible techniques. To simplify the process even more, Jin-Young Kim et al. applied the emitting and dielectric layer at the same time by spin coating a mixture of powder phosphor and an organic binder, that forms a dielectric layer on the phosphor particles during the process [209]. Marc de Vos et al. demonstrated flexible EL lamps applied directly onto a woven fabric using a novel dispenser printing process, where pressured air pushes ink through a syringe and nozzle onto a substrate. The advantage of this printing technique lies in the easily changeable design [253].

ACPEL devices mostly find their application as flexible, very thin, low brightness, inexpensive, large area background lighting displays for example in car radios, remote controls and electronic gadgets and minorly in the advertisement as lighting panels [254]. Despite the recent progress it still struggles with a number of issues that prevent further use of the device. An important constraint is the limited brightness that makes it impossible to use as a sunlight readable device. Another limitation lies in the restricted lifetime of the device, which can be augmented by using an encapsulation. The used materials are less oxygen and moisture sensitive than those of the other light emitting devices, which makes the barrier layer requirements less strict. A major advantage of the ACPEL device over the other devices lies in the application of the screen printing technique. The ACPEL devices can be fully printed with this inexpensive, fast, easily upscalable technique that is perfectly suited for continuous manufacturing. Furthermore the recent progress made on textiles, might open a new application field for the ACPEL devices in interior and exterior lighting design such as banners, flags, wallpaper and tiles.

#### 4.2.2. OLED

OLED technology has advanced rapidly the last decades because of the wide interest in the devices for both display and solid state lighting applications. The devices advantages are numerous: high brightness and efficiencies, low power consumption, wide viewing angle, colour tenability, high resolution, fast switching speed, very thin nanometre range layers, and thus lightweight and flexible, and surface light emitting. However, the devices also come with important challenges such as the necessity of a high-barrier layer.

The race for organic electronics was started by Ching Tang and Steven Van Slyke who fabricated the first SM-OLED in 1987 [22]. This device was composed of an ITO coated glass substrate, an aromatic diamine, a luminescent fill of fluorescent metal chelate complexes and a Mg:Ag electrode. All layers were applied using vacuum deposition techniques. Three years later in 1990 Jeremy Henley Burroughes et al. replaced the small molecules with a spin coated conjugated polymer polyparaphenylenevinylene (PPV) as emissive layer and the first P-OLED was a fact [24]. The main advantage of using polymers over small molecules was that the polymers can be used to form useful and robust structures by using solution processing techniques. Nowadays the small molecules can also be deposited using solution processing techniques. Görgustafsson et al. employed the advantage of the mechanical robustness of the emissive polymer and fabricated the first fully flexible P-OLED on PET in 1992 with following structure: soluble polyaniline (electrode and HIL), poly(1,4-phenylene-vinylene) (EML) and Ca (EIL and electrode) [26]. This publication opened the door for a wide variety of flexible electronics applications in both the solid state lighting and the display market. The latter will not be discussed in this paper. Innovative designs of the flexible devices can serve as surface light sources in automotive, clothing, decorative lighting, signage, advertisement and healthcare.

M-H Park et al. published a review paper in which the recent progress in the field of flexible OLEDs for solid state lighting was discussed [101]. This paper states that in order to obtain working practical flexible devices three technologies must be improved: flexible substrates, flexible electrodes, and flexible high-barrier encapsulations.

Numerous flexible materials can be used as an OLED substrate, there are however some physical properties that need to be taken into account such as the Young's modulus, the thermal expansion coefficient, the minimum bending radius, the glass transition temperature, the WVTR and the OTR. If the substrate possesses a WVTR and an OTR higher than respectively  $10^{-6}$  g m<sup>-2</sup> per day and  $10^{-3}$  cm<sup>3</sup> m<sup>-2</sup> per day, the substrate itself has to be provided with a high barrier encapsulation. Often plastic materials such as PET, polycarbonate (PC) and metal foils are used as flexible substrate. Another interesting and expanding field that has been entered by OLED technology is that of the smart textiles. In contrast to ACPEL devices OLEDs operate at a low DC-voltage, making them suitable for wearable applications. The integration of the devices onto the textiles remains a challenge, mostly due to the relative high roughness of the substrate. In general OLEDs are fabricated on other uniform substrates and consequently incorporated onto the textile surface. Researchers at the Fraunhofer Institute focused during the TexOLED project on the integration of miniaturized foil-based OLEDs onto fabrics by means of conductive adhesives or embroidery [255]. Multiple research groups produced fibre based OLEDs where the layers are applied using vacuum deposition techniques or simply dipping [61,63,256,257]. To preserve the characteristic softness of the fabrics even with integration of electronics, Choi et al. fabricated a highly efficient OLED on fabric woven from PEN fibres [258]. The fibre was thermally laminated with a planarizing layer to reduce the roughness, the complete OLED stack was applied using thermal evaporation and a transparent encapsulation was added to protect the device against degradation.

Highly important for a proper electric charge carrier injection is the choice of the electrode materials [35,103]. Possible suitable materials are graphene, conducting polymers such as PEDOT:PSS, Ag nanowires and dielectric/metal/dielectric multilayers. For the latter generally zinc sulphide (ZnS), zinc oxide (ZnO) and tungsten trioxide (WO<sub>3</sub>) are used as dielectric layers and Ag an Au as metal layers.

Due to the fast degradation of OLEDs caused by water vapour and oxygen an adequate high barrier encapsulation is absolutely necessary [62,102,259–262]. This is one of the major problems when dealing with flexible devices. Different techniques have been developed to encapsulate flexible OLEDs. Among these techniques thin film encapsulation (TFE) is the most known technique where a multilayer of films is applied using sputtering, evaporation or plasma vapour deposition of high-density film such as Sinitride, Al, Ta and SiO. For the encapsulation to be effective however, the devices have to operate at a lower temperature than glass-encapsulated ones. Another technique is Barix encapsulation or Vitex where an organic–inorganic multilayer, that consists of an alternation between Al<sub>2</sub>O<sub>3</sub> and polyacrylate, is applied using sputtering and vacuum deposition techniques. The polyacrylate is used to planarize the



surface and forms a protection against mechanical damage, while the  $\text{Al}_2\text{O}_3$  makes up the actual barrier layers. These techniques are quite expensive, have a long processing time and a low production throughput. Two other techniques are practiced: atomic layer deposition (ALD) and molecular layer deposition (MLD). Both techniques deploy self-limiting reactions to deposit films of various oxides, metal nitrides, metals and metal-sulfides (ALD) and organic polymers (MLD). With these techniques it is possible to control the film thickness and to obtain uniform reproducible films. Pinholes in the layer can be avoided because they use self-limiting reactions, which stop the growth process after reaction of the complete surface. However ALD and MLD are also expensive and have an inadequate throughput. Therefore other techniques are being developed. LG utilises a face-sealing encapsulation technique using inorganic/organic/inorganic layers. A flexible lamination technique was presented by Lee et al. where a metal foil/elastic polymer bilayer is being applied [101]. This Flex Lami-encapsulation demonstrates similar properties as a glass encapsulation.

OLEDs are often considered as the future in solid state lighting, replacing the traditional incandescent and fluorescent lamps [263]. Additionally to the already above listed advantages they do not contain any mercury, emit light across the visible wavelengths and show a lot of promise as energy efficient solutions [264,265]. Their thin nanometre range layers can be applied with a variety of techniques to almost any type of flexible substrate. Furthermore the devices can be considered ecological because they do not emit any UV light. Nevertheless some quite difficult challenges are still to be tackled for OLEDs as solid state lighting, such as improving the device stability and lifetime, the electricity to light conversion efficiency, the contrast, the pixel switching and colour saturation and the high production costs [263]. The first issues require the application of innovative manufacturing and encapsulation materials, while a solution for the latter can be found in utilizing low cost printing techniques. A fully printed OLED has not been achieved yet, but research is ongoing and it is expected to be done in the near future.

Another not to neglect competitor in the world of solid state light is the light emitting diode (LED), which is not part of this article, but will be mentioned here to give a more complete discussion. OLEDs have an infinite contrast ratio, thinner flexible layers, a lower weight, a superior power consumption and are area emitters [263].

OLEDs are often employed in the display and lighting industry, but new fields are being explored to employ these devices. The field of smart textiles is one of these new markets, where OLEDs can be applied directly onto textile materials for protective, esthetical or commercial purposes [266–268]. Interior designers are inspired by the many possibilities of OLEDs in lighting on every possible shape and substrate for example on curtains, windows, walls and the automotive sector tries to incorporate them onto car interiors. Another non-explored field is that of the biomedical applications, where OLEDs can be a valuable addition to light therapy as a surface emitter [269]. The existing difficulties do however need to be solved before OLEDs can be truly the future of solid state lighting.

#### 4.2.3. LEC

An answer to the encounters faced by OLED technology is the LEC technology. This device has a simple layout and does not need the built-in electrical field to insure proper charge injection and transport, thanks to the mobile ions present in the active layer. Nonetheless the exact physics behind the device is highly complex and still not fully understood.

In 1995 Qibing Pei et al. presented their innovative light emitting device, the LEC [72]. These devices were fabricated on top of a glass substrate and consisted of three layers, a sputtered ITO layer as a bottom and transparent electrode, a spin coated conjugated polymer with added electrolyte and a thermal vacuum deposited metal electrode. One year later J. K. Lee et al. demonstrated that the conjugated polymer can be replaced by an ionic transition-metal complex (ionic small molecular-weight complex) [73].

The operation time of the device is limited by the exciton-polaron quenching caused by the growth of the doped regions towards the emission zone. To counter this limitation several physical or chemical strategies were developed to stabilize the p-i-n junction. Zhibin Yu et al. fabricated P-LECs using a poly(ethylene oxide) (PEO) oligomer capped with methacrylate end groups that conduct ionic species to form the p-i-n junction [270]. During the junction formation the methacrylate polymerizes and stabilizes the p-i-n junction and decreases the non-uniformity of the doped regions. Another approach, demonstrated by Leger et al., is to chemically fix the p-i-n junction using polymerizable ions creating a stable and permanent configuration of compensating ions [271]. Shi Tang et al. developed a fast and flexible method to photochemically stabilize the doping structures of the conjugated polymer by adding a photo-initiator compound and a cross-linkable ion-pair monomer to the active layer and then shortly expose it to UV-light [272]. By adding copolymers of poly(ethylene glycol) methyl ether methacrylate to the active layer Wantz et al. increased the glass transition temperature above room temperature. The p-i-n junctions are frozen when the devices under applied voltage bias are cooled down to room temperature and thus stabilized [273].

Because of their relatively slow turn-on time, high-end display applications are not within the scope of the LEC technology. The devices simple structure and production process however makes them highly suited for lighting applications on a variety of flexible substrates [274]. White light with a high luminance and efficiency is necessary when looking at general lighting applications, which remains a challenge. Asadpooravish et al. used an ambient mobile airbrush to apply a complete LEC device on a specialty-paper and low-end copy paper [140]. This resulted in a highly flexible, low weight and low cost device. Orti and Bolink proved that light emitting fibres fabricated by dip-coating a steel wire with the LEC device are suited to be integrated in textile [275]. LECs do have an answer to some of the difficulties that OLEDs are struggling with [276]. They realise a well-balanced charge injection by using mobile ions instead of a built-in electrical potential, so the choice in materials is more free and air-stable metals and thick emissive layers can be employed. Also a high electrical efficiency is amongst the advantages of the devices. Despite those advantages over OLEDs, LECs are not ready to take their place in solid state lighting due to a lower operational time and efficiency. More research is required to fully uncover the complex processes and electronic structures behind the luminescence and chemically stabilize the LEC junction [277].

## 5. Conclusion

To improve our quality of life we can look beyond the use of lighting solely for visibility and safety purposes, we can create a complete visual environment including other needs like health, communication, productivity, comfort, aesthetics and mood. This leap into this brighter future requires that we move beyond rigid and planer lighting and pave the way for lighting on flexible and bendable substrates. It implies also light emitting devices that can be flexed and bended without losing their luminous performance. Three light emitting devices embrace all the necessary requirements, namely the alternating current powder electroluminescent device (ACPEL device), the organic light emitting diode (OLED) and the light emitting electrochemical cell (LEC). Moreover these devices possess uniform surface light emission combined with high efficiencies. Additionally the production techniques are of high importance and aim to be cost-effective, high throughput and environmental friendly. All these qualities can be found in printing techniques as there are blade coating, spray coating, ink jet printing, slot-die coating, screen printing, pad printing and gravure printing.

ACPEL devices are driven by a high AC voltage ( $\sim 100$  V) and composed of  $\mu\text{m}$  thick simple layer structure generally entirely screen printed. The latter is a high-throughput and very cost effective technique, making these devices easily producible at a very economical price. Furthermore screen print pastes are increasingly water based, making the production process also environmentally-safe. In addition the materials used are chemically stable and do not require a high-end barrier layer. Other printing techniques like blade coating can be applied, but are less common. The devices' AC driving voltage makes them less popular to incorporate in wearables. At present ACPEL devices are mostly used in commercial advertisement and elementary displays. However they caught the interest of the world of interior and exterior design to be applied as large lighting panels or simple displays in wall paper, floors, flags, etc... Furthermore they are also highly suited to be used as safety indication lighting. Considering the limited resolution and turn on time they can reach, they will likely not be used for high precision applications. Thanks to the complete knowledge of the operational capacitive principle of the device, research can be focussed on integrating innovative materials like graphene and silver nanowires and on applying different compositions and designs of the device on any type of substrate.

The structure of OLEDs is more complex consisting out of numerous nm thick layers to improve the light outcoupling and increase the efficacy, but with a low DC driving voltage ( $\sim 4$  V). This complexity ensures that the choice in materials is rather limited for the proper working of the devices, narrowing also the possible deposition techniques. A fully printed OLED is therefore yet to be realised. Most used application techniques are spin coating (for polymers) and PVD (for metal and small molecules), keeping currently the production cost of the devices quite high. Spray coating, more specifically ultrasonic spray coating, shows a lot of promise to replace both techniques as it is capable to deposit both polymers and metals without changing their internal structure. Moreover this technique is roll-to-roll compatible, economic and ecologic (if the used ink is water or alcohol based). In a way the OLEDs' low driving voltage forms an answer to the unsuitability for ACPEL devices to be incorporated in wearable textiles. In other words OLEDs are highly fit to be merged into wearables. The roughness of the textiles combined with the nm thick layers of OLEDs adds another difficulty to the equation in the form of a planarizing layer. The application field for light emitting wearables is very broad, for instance for esthetical purposes (light emitting clothing of all shapes and forms) or safety purposes (safety clothing). Flexible OLEDs are in fact eligible for a high variety of applications. Besides the obvious ones like lighting of all designs and shapes, both indoor and outdoor, they also appeal to the automotive sector as indoor lighting and even to the medical sector as light therapy. This alluring device however also comes with the necessary shortcomings of which the most important is its sensitivity to water vapour and oxygen reducing its life time and making an high barrier encapsulation crucial. To obtain an aforesaid encapsulation for flexible devices forms even a greater challenge than that for rigid devices, especially when it has to be printed. Research is ongoing and progress in the use of innovative or alternative materials, in the type of flexible substrates and in the application of high throughput printing techniques is often made. Hopefully researchers continue this exploration and do not trip over the many hurdles to be taken.

Initially the LEC formed an alternative for the complex OLED with its simple three layered stack, also emitting light driven by a low DC voltage, but it does not accomplish the well-balanced charge injection by using an intrinsic electrical potential thanks to the addition of mobile ions in the active layer.

This also makes the choice in electrode materials more liberate thus chemically stable materials can be selected that are not in need of an high-end encapsulation and that can be applied by several printing techniques, keeping the production costs within limits. However the devices employ the same active materials as OLEDs (with additives and mobile ions), using the same harmful solvents. Nevertheless the largest obstacle for this devices progress is the lack of fully comprehension of the mechanisms at work for the initiation of light emission. Consequently the lifetime of the devices stays relatively low. At the moment the application of the LEC can be found in lighting, signage and basic displays, but is rather limited. This can be enlarged and broadened once the working principles are fully discovered and understood, and accordingly solutions can be brought forward to improve the lifetime of the devices.

To summarize, these three light emitting devices all have their advantages and limitations and qualify for different applications and fields. As different their working mechanisms are, large similarities can be found in their surface light emission, their liberty to be applied on flexible and bendable substrates and their ability to be deposited using high-end throughput printing techniques. With research into this three devices still ongoing the future for flexible and bendable lighting looks promising.

## Declaration of Competing Interest

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

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