Recent developments on printed photodetectors for large area and flexible applications

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Abstract

New generation photodetectors processed by printed technologies will become a feature of our daily life in the near future. By combining printing technologies with tailored solution processable materials, high efficiency, large area and low-cost devices are achieved. Medical and health care, Internet-Of-Things, Industry 4.0 are strongly increasing fields taking advantage on the developments of printed technologies in general and printed photodetectors, in particular. Applications for consumer electronics such as smart phones, laptops and tables, fingerprint scanning for personal security are some of the most interesting areas to explore in printed photodetectors.

This work reviews the recent progress in the development of photodetectors by printing technologies, taking into account both materials and printed technologies.

1. Introduction

Optoelectronic materials are being explored for being implemented in sensors, unleashing their potential to transform our lifestyle. Video imaging, optical communications, biomedical imaging, security, night-vision, gas sensing and motion detection are some of the applications with high interest [1].

Nowadays, photodetectors, PD, have a large impact in some of the technologies used in our daily life. PDs are being used in applications such as radiation [2], smoke and flame detection [3] and security inspection [4], among others. Photodetector used for medical X-ray imaging [5] and spectroscopy [6] and also for security systems [7] are some other applications with attracting interest.

The development of novel materials and applications is essential to promote higher performance, sustainability and added value to optoelectronic devices and solutions. Thus, materials are being developed with improved properties, such as, chemical and radiation resistance, wavelength response range, flexibility and integrability, among others [8].

Printed photodetectors can overcome some limitations of current silicon based photodetectors, such as low absorption coefficient and the high temperature, complex and expensive fabrication processes, as well as expand their application range to flexible and large area devices [9]. Solution processable materials allow the fabrication of these devices by printing technologies, such as inkjet printing, spraying and screen printing. Thus, the processing of flexible and stretchable devices, improving their integration and allowing production scalability at low fabrication costs is a reality. On other hand, the main limitation of printed photodetectors are the reduced charge transport and stability. [8].

In this review the most relevant works on printed photodetectors and their applications are presented and discussed. Thus, an overview of the state of the art on the different types of printed photodetectors is provided and the main challenges and future needs addressed.

In the following, the state of the art on printed photodetectors is presented, together with the printing technologies used for their processing. The main applications are also summarized and the future challenges and trends outlined. Finally, the main conclusion on the current state of the art and future needs on printed photodetectors are presented.

2. Printed photodetectors

To improve flexibility, precision and reduce the fabrication cost in order to achieve new generation of PD, new methods based in printing technologies have been applied [10]. Printing technologies are additive processing techniques that involve a single deposition step per layer usually followed by a drying step [10, 11].

There are several printing techniques such as inkjet, screen printing and spray printing that can be used for the fabrication of photodetectors [8]. Printing technologies are significantly growing, as they present many advantages when compared to traditional lithography methods, such as low cost processing, low material waste and the possibility to print into flexible and large areas [12].

2.1. Main requirements and characteristics of printed photodetectors

Currently, there are a reasonable number of different printed PD for various applications and fabricated from a variety of materials. Nevetheless, there are still great challenges for developing the new generation PDs, when shrinking the size of conventionally used thin-films hinder their quality or when ultrathin materials become almost transparent losing the ability to gather or absorb light. Thus, a possible way for developing high performance photodetectors arise form the combination of the advantages of both organic (simple solution processability) and inorganic semiconductors (high charge carrier mobility) [13-15]. Depending on the application, a photodetector must fulfill specific requirements to present a good performance, some of these requirements and characteristics being difficult to combine in one type of photodetector, such as high sensibility and high detection bandwidth [16, 17].

One of the most important requirement is the sensitivity of the photodetector in a specific spectral region, as determined by the active material. In some cases, the responsivity should be constant or at least well defined within some wavelength range. It can also be important to have zero response in some other wavelength range, e.g., solar-blind detectors are sensitive only to ultraviolet light [17-19]. A PD should be suitable for some range of optical powers: the maximum detected power can be limited by damage issues or by a nonlinear response, whereas the minimum power is typically determined by noise. The magnitude of the dynamic range (specified as the ratio of maximum and minimum detectable power) is often the most important. Some detectors based on photodiodes can exhibit high linearity over a dynamic range of more than 70 dB [16, 17]. Concerning the photon detection, a high quantum efficiency is required in order to get a good efficiency conversion of photons into electrons [17, 20].

The active area of a detector plays an important role when working with strongly divergent beams from laser diodes. For light sources with very high and/or non-constant beam divergence, it is hardly possible to get all the light into the active area [17, 21, 22]. The detection bandwidth is typically large, beginning at low frequencies and the maximum frequency being limited by internal processes as the speed of electric carriers in the semiconductor material, or by the involved electronics [17, 23]. Time response strongly affects the work of a PD used for detecting pulses. Some detectors show a certain "dead time" after the detection of a pulse, where they are not sensitive [16, 17]. Another requirement of photodetectors is the high reliability and also the low noise for reduced errors. In summary, current density , fast responsivity, selected absorption bandwidth, and high quantum efficiency must be considered in order to process optimal PDs [8].

Formulated into functional inks, semiconductors can be processed by means of printing techniques, allowing thus large-area, scalable and flexible device fabrication, opening a complete new pathway for organic electronics and its integration into circuits [8]. An increasing number of printable organic semiconductors and nanoparticles employed as active materials in PD are being proposed in the literature, and this flourishing field is speeding toward scalability and industrial commercialization [8].

In contrast to lithographic methods, mostly employed in the fabrication of inorganic devices via a subtractive approach, printing technologies are additive processing techniques that involve a single deposition step per layer, eventually followed by a drying or sintering step [8].

There are some materials and inks that can be combined and used to produce new types of photodetectors depending to the application. Poly (3,4-ethylenedioxythiophne) poly(styrenesulfonate) (PEDOT:PSS) and poly (3-hexylthiophene)/phenyl-C61-butyric methyl ester (P3HT:PCBM) show good properties as an ink to apply in photodetectors with distinct applications and functions [12, 24].

In the majority of the research works silver and gold materials were used in the structure of printed photodetectors due to their stability and conductibility [25].

2.2. Photodetector structures

Among the diverse light detecting devices and concepts that have been developed over the years, the basic working principle of a PD consists in the process of light absorption which leads to the generation of separated holes and electrons, and their subsequent collection in opposite electrodes. Such mechanism is typical of inorganic p–n junctions as also of donor–acceptor organic semiconductors [17, 26].

The nature of the active materials leads to the charge excitation, dissociation and migration to the respective electrodes. Several studies and reviews have already been reported concerning these processes [8].

A simple sketch of the electronic structure of a PD is shown in **Figure 1**. In this PD, a semiconductor layer is sandwiched between two conductive layers, a p-layer on top and a n-layer underneath, denominated PIN-heterostructure. The photodiode structure is bonded on top of the interconnect layer where the optical signal propagates before entering the detector. Light is coupled first from the interconnect layer to an InP membrane waveguide, spaced by 300 nm (500 nm) when the photonic wire is fabricated in Si (Si_3N_4), then it is absorbed and detected in the PD [18, 25].

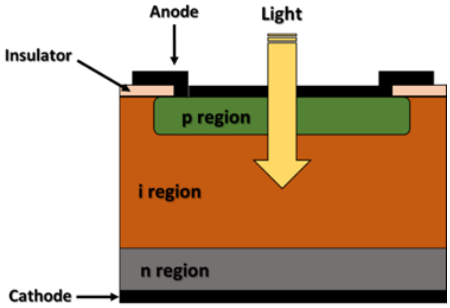


Figure 1. Basic structure of a PIN photodetector.

When a specific energy photon strikes the device, it generates an electron-hole pair. If this phenomenon occurs near the depletion region, the particles of the pair will be accelerated in opposite directions by the electric field. The electrons will migrate to the n side and the holes to the p side [27-29]. This movement gives rise to a reverse electric current in the photodiode. Even in the absence of light, there will always be a reverse current flowing in the photodiode, arising from electron-hole pairs formed spontaneously at the junction with enough energy to reach the terminals [17, 30].

Concerning printable PD, the same basic structure is used. However, printing technologies allow new and improved ways to develop PD. They allow the fabrication of well-defined structures as vertical or planar and more efficient three terminals with the addition of a gate electrode. A simple sketch of the electronic structure is shown in **Figure 2** [8]. Planar and vertical structures can operate as a photodiode or a photoconductor. Planar structures are easier to fabricate than the vertical ones and do not need a transparent electrode, showing a much larger distance between the electrodes. However, the length can drastically increase the carrier transit time, and its downscaling would require high resolution lithographic processes [8, 31]. Scaling of the distance is instead more easily achieved in a vertical structure, where the distance between the electrodes is defined by the thickness of a solution processed active layer, providing much faster charge collection. Three terminal devices such as field effect transistors (FETs) can be used as light sensors, typically with the scope of achieving very high responsivity. The main advantage of three-terminal devices has been proven as one of the best candidates for improving low power consumption and high-sensitivity ultraviolet photo- detectors (UV PDs) such as ZnO based photodetectors.

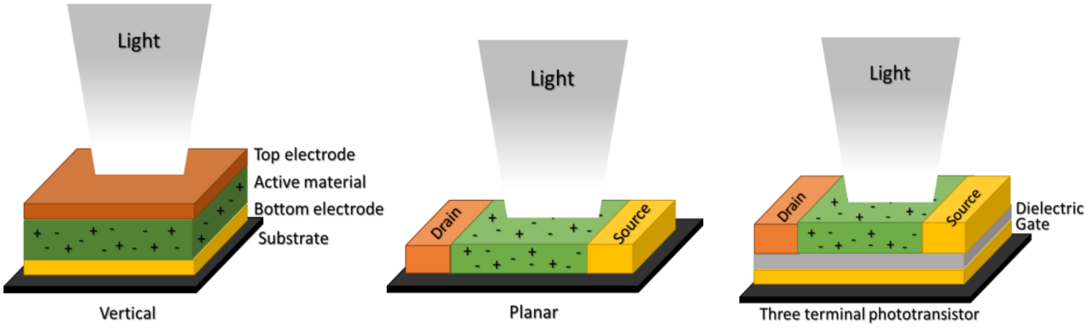


Figure 2. Most commonly printed photodetector structures: Vertical architecture, planar architecture and Three terminal phototransistor device.

Several layers can be printed subsequently resulting in fully printed devices with excellent optoelectronic performance [11, 31]. The most common printed PD structure is the vertical topology with the photoactive layer sandwiched between two conductive stripes. This structure is ideal to be processed by printing [24].

Table 1 summarizes the advantages/disadvantages and some generic applications of planar, vertical and three device structures for printed photodetectors.

Table 1. Advantages, disadvantages and aplications of planar, vertical and three device structures.

Structure	Advantages	Disadvantages	Aplications	Ref.
Planar	- Easy to construct - Do not need a transparent electrode on the top	- High distance between the electrodes (increment in the carrier transit time); - Downscaling require high resolution lithographic processes	-Photo-sensing elements for digital imaging; -Large-area shortwave infrared (SWIR) photodetector arrays - Optical fiber-based systems -Highly sensitive and applicable at night vision applications	[25, 32-36]
Vertical	- Higher integration density - Higher gain and fast response - Small distance between the electrodes	-Higher number of steps for patterning fabrication	- Imaging detectors - Position-sensitive detectors - IR sensor array	[30, 37-40]
Three device structure	- Very high responsivity - Low power consumption - Additional functions, mainly logic or nonlinear electronic characteristics	- Complex fabricaton with more components	- Optoelectronic devices - Interactive surfaces, flexible displays, and surveillance systems applications	[35, 40, 41]

3. Printed photodetectoers: technologies, materials and performance

The increasing interest on printing technologies is mainly related to the possibility of layer processing by using a non-complex and cost-effective technique compared to conventional electronics processing. Using additive fabrication processes it is possible to achieve a wide range of new functionalities which will allow new applications and/or the improvement of specific existing applications.

Traditional fabrication techniques such as lithography or thermal evaporation and spin coating (in the case of organic photodetectors), show drawback related to waste of material, energy and time-consuming processes, limitation to small areas or rigid substrates or being very expensive. Printing techniques instead, are additive processing techniques where just the required material amount is used and roll-to-roll compatible.

Among solution-processable semiconductors, organic conjugated materials are easily printable by different printing technologies, due to the possibility of tuning the formulations rheology and therefore of engineering inks for different printing techniques and applications. Their low-temperature processing (<150 °C) allows the device fabrication on a vast range of substrates, rigid and flat as well as flexible and curved, opening the way for the development of non-fragile, conformable electronics [8]. The selection of the printing technology is determined by requirements of the printed layers, by the properties of printed materials as well as cost effective and technical considerations of the final application of the printed samples [12].

Some of the most used and attractive printing techniques for printed photodetectors fabrication are inkjet printing, screen printing and spray coating.

Inkjet printing is a non-contact digital process [10, 42]. A pattern is designed in the computer software and the cartridge is moved all over the substrate following the pattern and depositing drops in the required places. This technology allows the design of complex patterns with high resolution customizing the material deposition volume and location, i.e. it provides the droplet-on-demand option (DOD). The material is deposited drop by drop, hence, for a continuous path drops need to overlap or touch each other. Multilayers can also be printed, and the thickness of the layer can be controlled either in this way or changing the amount of the material in the drop. **Figure 3a** shows a sketch of an inkjet printer working principle.

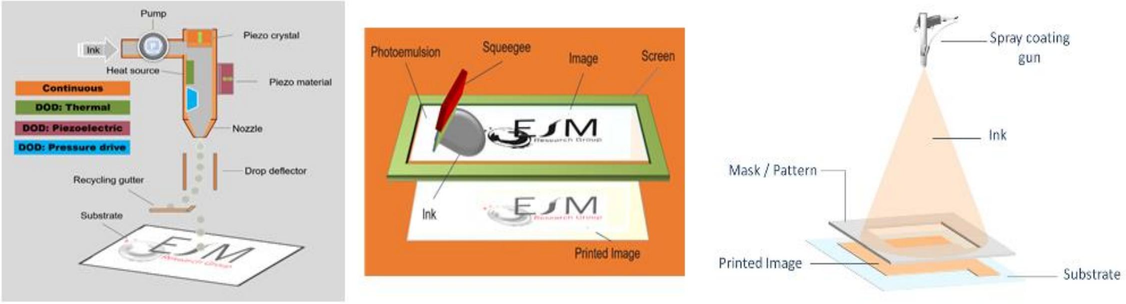


Figure 3. Schematic representation of: a) the inkjet printing process b) screen printing process.; c) spray coating deposition technique. Reproduced with permission [43].

One of the first inkjet printed layers on a photodetector was P3HT:PCBM [11]. The device structure consisted on a glass substrate covered by indium-tin-oxide (ITO), a spin coated PEDOT:PSS layer, an inkjet printed P3HT:PCBM blend and evaporated calcium (Ca) and silver (Ag) layers as the top electrode.

The first fully inkjet printed photodetector was fabricated based on a silver nanoparticles layer on top of the substrate as the bottom electrode, followed by poly(9,9'-dioctylfluorinene-co-bis-N,N'-(4-butylphenyl)-bis-N,N'-phenyl 1,4 phenylenediamine) (PFB) and poly(9, 9'-dioctylfluorene-co-benzothiadiazole) (F8BT) as the active layer and PEDOT:PSS as the top electrode [44].

The performance of the previously described fully inkjet printed photodetector was improved in terms of external quantum efficiency (EQE) by printing a thinner (~120 nm) P3HT:PCBM layer as the active material [12]. Moreover, by introducing a polyethylenimine (PEIE) layer (by spin coating) between the photoactive layer and the silver electrode, the cathode work function was reduced, and the wettability of the photoactive material was improved.

Further, a fully inkjet printed organic photodetector based on narrow band gap conjugated molecules [24] was demonstrated showing that the use of small molecules allows to improve the spectral responsivity of the detector. Recentely, an all-printed and flexible indirect X-ray detector was successsfully achieved by using a combination of screen and inkjet printing. Polymer based scintillator inks were used to print the photodetector TFT-based array with a promising performance for flexible optoelectronics with a low-cost and large-area [45].

Screen printing is a printing technique (**Figure 3b**) where a mesh is used in order to transfer the pattern to the substrate [46]. A squeegee is then moved all over the substrate along the mesh and the ink in transferred to the substrate though the apertures of the mesh. A screen printed CdS:CdO structure was reported as suitable to be used as a photodetector [47]. A porous CdS:CdO composite structure was developed by screen printing and sintering of CdS in air.

Also mercuric iodide (HgI_2) films were processed for X-ray detection [48-50]. The X-ray sensor with an oxide thin-film transistor backplane and HgI_2 sensing material displayed outstanding image quality under a low X-ray exposure and a low electric field [51].

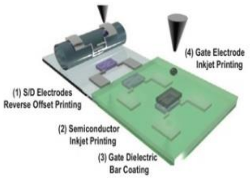
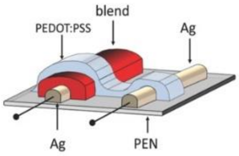
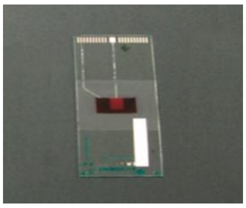
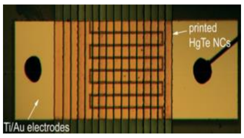
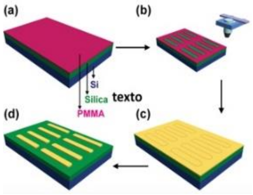
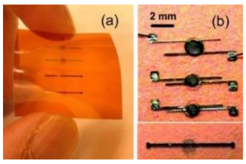
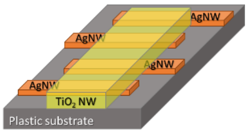
Similar to inkjet printing, spray coating technique (**Figure 3c**) is not in direct contact with the substrate and the material is deposited in micron size droplets. The liquid coming out from a nozzle is atomized by ultrasonic vibrations or pressure and the droplets are deposited randomly. An inert gas like nitrogen for example is used as a carrier gas. Depending on the solution flow rate and the moving speed of the nozzle, the thickness of the layer can be tunned. This deposition method does not allow complex patterns unless a mask with the desired pattern is used.

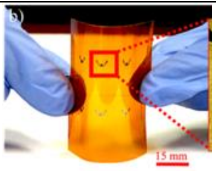
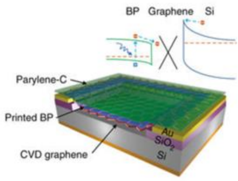
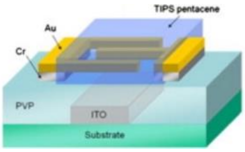
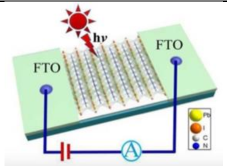
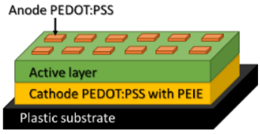
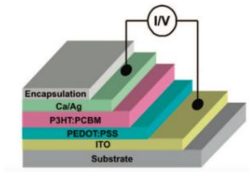
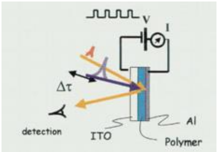
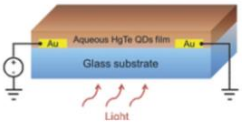
Fully spray coated photodiodes based on P3HT:PCBM has been presented [37] showing that despite the larger surface roughness with respect to films obtained by other techniques such as e spin coating and doctor blade, the performance was improved, with responsivities of 0.36 A/W and lifetime beyond 1 year.

Hybrid CMOS-imager with a solution processable polymer as photoactive layer was showed in [52]. A P3HT:PCBM layer was deposited by spray coating on top of a silicon based CMOS-pixel array. PEDOT:PSS, PEDOT:PSS/Graphene and carbon nanotubes have also been deposited by spray coating in organic photodetectors as transparent electrodes [53-58]. In this way P3HT:PCBM based photodetectors were fabricated on PET substrates by spray coating. Several groups have also investigated on spray coated ZnO, that was used as electrode and active layer [59, 60]. Finally, spray coated inorganic materials based on aqueous HgTe-QD have been used for the development of infrared photodetectors [34].

Together to the abovementioned representative examples, **Table 2** reports on the main printing technologies, the material used, device structure and mains characteristics and applications of the most significant printed PD.

Table 2. Printed photodetectors by printing technologies. All images reproduced with permission.

Printing Technology	Device material	Device Structure	Responsivity R (A/W)	Application	Ref.
Inkjet	Au/TIPS pentacene/Au phototransistor with ITO/PVP as back gate		0.11	Low-cost and high-performance photo-sensing element for digital imaging applications	[33]
Inkjet	PEN/Ag/P3HT:PCBM/PEDOT:PSS/Ag		0.39	Integration in pixels of future flexible and large-area digital imagers manufactured on plastic foils	[12]
Inkjet	ITO/PEDOT:PSS/ P3HT:PCBM /Al		0.25	Detection of bacterial superantigens as *Staphylococcal enterotoxin B* at low concentrations	[61]
Inkjet	Au/HgTe Nanocristals/ Au		0.65	Highly sensitive and applicable at night vision applications.	[25]
Inkjet	Au/PDPPTzBT/Au/ SiO_2/Si		10^6	Light triggered memory device	[62]
Inkjet	Ag/CNT/Ag photodetector		0.5	Low-cost fully integrated CNT based IR sensor array.	[63]
Inkjet	AgNW/TiO_2 NWs/AgNW photodetector		-	Photoelectric devices for transparent microelectronic applications.	[32]

Injekt	10 mg/ml graphene/MoS$_2$ in 10 ml C/T with 2.5 wt% EC)		0.30	Age-related macular degeneration and retine deseases.	[36]
Injekt	BP/graphene/ Si		0.164	High performance broadband for optoelectronic applications	[64]
Inkjet	PEN/PEDOT:PSS/PFN/T1:P3HT:PC$_{70}$BM/PEDOT:PSS		-	Applied in interactive surfaces and surveillance systems	[41]
Screen Printing	$CH_3NH_3PbI_3$		-	Can be applied in solar cells	[35]
Screen Printing	ITO/PEIE/PCDTBT:PC60BM/ PEDOT:PSS		0.34	Excellent performance in terms of dark current and quantum yield	[65]
Screen Printing	PEDOT:PSS:PEIE/PCDTBT:PC$_{71}$BM/PEDOT:PSSS		-	Can replace the silicon photodiodes used in image sensors	[66]
Spray	ITO/ PEDOT:PSS /P3HT:PCBM /Ca/Ag		0.36	Imaging applications or position-sensitive detectors	[37]
Spray	PEDOT:PSS/ Al/ P3HT:PCBM		0.01	Applied in optical fiber-based systems	[67, 68]
Spray	Au/ HgTe QD films/Au		0.5	Applied in large-area SWIR photodetector arrays	[34]

Some important aspects must be taken into consideration with respect to printable materials such as viscosity, surface tension, and solid content, among others.

In particular, each printing technology has important limitations in terms of viscosity, resolution and maximum size of the solid content, among others (Table 3).

Table 3. Printing technologies characteristics and ink/substrate requirements [43, 46].

Printing technology	**Advantages**	**Disadvantages**	**Viscosity (mPas)**	**Max. particle diameter (µm)**	**Film thickness (µm)**	**Resolution (µm)**
Inkjet (No contact)	-Low ink amount and viscosity	- Low throughput -Nozzle reliability	8-20	0.2	0.1-20	20-50
Screen printing (Contact)	-Robust -Simple -Thick layers	- Low throughput and resolution -High ink viscosity	200-10000	70	5-20	50-100
Spray (No contact)	- High throughput and thickness variation	-Low resolution	1-1000	10	7.5-10	1000

In conclusion, inkjet technology is suitable for high resolution printing patterns with the requirement of using low viscosity inks. For lower resolution patterns, simple printing technologies as screen printing and spray could be used. It is crucial that the ink parameters (viscosity/surface tension) match the printing technology to be used to acquire a good printing pattern.Thus, together to accomplish with the processing requirements, each printable layer of a photodetector has some specific characteristics and depending of the material used in the formulation of the inks, the final yield and the possible application of the device will be determined. **Table 4** shows this difference and the materials applied in the layers, as conductive, semiconductive or dielectric/insulator of some of the aforementioned photodetectors.

Table 4. Materials characteristics for printable photodetectors.

Layers (Inks)	Deposition technique	Material	Characteristics and performance	Ref.
Conductive	Spin-coating	Commercial Fluorine-doped tin oxide (FTO) glasses	Sheet resistance: 14 Ω/sq	[35]
Semiconductive (active material)	Screen printing	Synthesis of perovskite thin films by dipping PbO thin films, previously prepared, into CH_3NH_3I with 2-propanol. PbO thin films prepared by a solvent precipitation method ($Pb(NO_3)_2$ + $NaBH_4$ + Triton-X-100 + NaClO) and mixed with ethanol, ethyl cellulose and terpilenol.	Thickness of the film: 700-800 nm	
Dielectric		---	---	
Conductive	Photolithography, thermal deposition and lift-off	Gold interdigitated electrodes	Metal thickness: 30 nm	[34]
Semiconductive (active material)	Spray	HgTe quantum dots synthesized in aqueous solution via a reaction between $Hg(ClO_4)_2$ and H_2Te gas in the presence of 1-thioglycerol)	Layer thickness: 150-200 nm	
Dielectric		---	---	
Conductive	Spray	Anode: Indium tin oxide (ITO) coated glass with commercial PEDOT:PSS as a hole conductor	Anode sheet resistance: <15 Ω/sq	[37]

		Cathode: Ca and Ag	Cathode transmission: 65 % at 532 nm	
Semiconductive (active material)	Spray	Bulk heterejunction diode from a xylene solution of regioregular rr-P3HT and PCBM	rr-P3HT:PCBM ratio of 1:0.75	
Dielectric		---	---	
Conductive	Sputtering	ITO gate electrode For source/drain electrode a suspended top-contact electrode structure was fabricated with Cr and Au bi-layers	ITO thickness: 100 nm Cr and Au thickness: 50 nm each	[41]
Semiconductive (active material)	Inkjet	Organic semiconductor ink of 6,13-bis(triisopropylsilylethynyl) (TIPS)-pentacene prepared from anisole solution	---	
Dielectric	Spin-coating	Organic gate insulator of poly-4-vinylphenol (PVP) prepared with PVP powder mixed with poly(melamine-co-formaldehyde) methylated in propylene glycol monomethyl ether acetate	---	
Conductive	Inkjet	Graphene electrodes ink (graphene in cyclohexanone/terpineol (CT) with ethyl cellulose (EC))	Ink viscosity: ~12 cP	[36]
Semiconductive (active material)	Inkjet	MoS$_2$ in CT with EC	CT ratio: 7:3 Ink viscosity: ~12 cP	
Dielectric		---	---	
Conductive	Inkjet and vacuum deposition	Ag contact with PEDOT:PSS (*CLENOS P Jet N*) stripes	---	[61]
Semiconductive (active material)	Inkjet and spin-coating	P3HT:PC$_{61}$BM Commercial P3HT andcommercial PC$_{61}$BM dissolved in 1,2-dichlorobenzene and mesitylene	P3HT: RR = 96.6%, MW = 65500) PC$_{61}$BM purity > 99.5%	
Dielectric		---	---	
Conductive	Inkjet	Commercial PEDOT:PSS Ag S/D electrodes	Ag content: 39 wt%, viscosity: 1.5 cPs at 0.4 rpm, surface	[33]

			tension: 25.8 mN m1 ; ANP Co.)	
Semiconductive (active material)	Inkjet	poly[2,5-bis(2-octyldodecyl)pyrrolo[3,4-c] pyrrole-1,4(2H,5H)-dione-(E)-1,2-di(2,20 -bithiophen-5-yl) ethene] PDPP-DBTE	---	
Dielectric	Bar-coating	Commercial Poly(methyl methacrylate) (PMMA)	MW = 120 k, dielectric constant εr = 3.5)	
Conductive	Inkjet	PEDOT:PSS and Al	---	[12]
Semiconductive (active material)	Inkjet	P3HT:$PC_{61}BM$	---	
Dielectric		---	---	
Conductive	Optical lithography and evaporation	TI and Au electrodes	---	[25]
Semiconductive (active material)	Inkjet	Hydrophobic HgTe nanocrystals (NC) Synthesized in aqueous solution at room temperature via a reaction between $Hg(ClO_4)_2$ and H_2Te gas in the presence of short chain hydrophilic thiols such as thioglycerol or mercaptoethanola- mine as stabilizer	---	
Dielectric		---	---	
Conductive	Thermal evaporation	3 nm Ti and 30 nm Au		[62]
Semiconductive (active material)	Inkjet	PDPPTzBT (diketopyrrolopyrrole-thiazolothiazole copolymer)	Hole mobility of 1.80 cm^2 V^{-1} s^{-1} on/off ratio of 10^8	
Dielectric	Spin-coating and inkjet	3 nm of PMMA 300 nm silica (dielectric)	M_W (120 KDa, 350 KDa, 550 KDa and 996 KDa)	
Conductive	Inkjet	Ag electrodes	---	[63]

Semiconductive (active material)	Inkjet	Commercial multiwalled MW carbon nanotubes CVD-grown	Diameters in the range of 10–20 nm with a length between 1 and 10 mm	
Dielectric		---	---	
Conductive	Inkjet	AgNW electrodes	Five layers of AgNW to ensure the efficiency of photocurrent collection. (Sheet resistance: 10 Ω/sq)	[32]
Semiconductive (active material)	Inkjet	Titanium dioxide (TiO_2) nanowires	Good electron mobility	
Dielectric		---	---	
Conductive	Chemical vapour deposition	Graphene	---	[64]
Semiconductive (active material)	Inkjet	Black phosphorus ink	Low oxidation proportion (4.2%)	
Dielectric		---	---	

From **Table 4** it is concluded that the existing inks despite to show specific similar characteristics, differ in many key issues, such as funcionality, performance and materials composition that are more suitable for the specific application. Thus, a careful selection of the appropriated inks is critical by taking into acount the main function of the materials compositions for each specified layer that will be printed, as well as the compatibility of the different layers. By this state of art it can also be concluded that there are few fully-printed PD. With the use of printing technologies, low-cost and flexible PD with large scalability can be obtained.

4. Summary, final remarks and future trends

This review reports on the main materials, structures and techniques used for the development of printed photodetectors by inkjet, spray and screen printing technologies and their applications.

Printed photodetectors show increasing interest as printing is a low cost and roll to roll compatible process. The increased amount of research works and demonstration of the use of printed photodetectors in a wide-range of applications mirrors the high interest of the scientists and industries in their development and application. PD have a large range of technological applications in fast growing areas such as telecommunications, sensors, and medical devices, among others [17, 69]. These printed devices can be used in the automotive industry – for the detection of objects to avoid collisions- , in factories - for electrical arc detection, automatically cutting off the current where arcing occurs-, in a process control -as a position sensor -, in recycling plants, in non-destructive testing units and as detector systems that can endure extreme environmental conditions [70].

The well-known silicon-based photodetectors are manufactured using the time-consuming, expensive and complicated fabrication process that involve several steps [71]. Moreover, these sensors are rigid and brittle, with thick layers and limited to small areas.

Printing appears as the solution to these technological disadvantages, offering several advantages such as high speed, low cost, possibility of room temperature processing, and applicability to flexible substrates. Printing technologies also provide direct patterning of a surface eliminating the need of expensive masks and opening the door to complex patterns and the choice of deposited materials.

The key issues hindering further development are the improvement of novel materials and inks with tailored functional and processability characteristics, as well as to develop new and more flexible processing ways to directly print photodetectors.

It should also be noted that most of new products from printing technologies must be based on green solvents.

Despite all the advantages of printed photodetectors, the vast majority of the current commercially available optical devices are not based on printed technologies.

Applications such as flexible displays applied on medical imaging, diagnostics devices, identification tags and solar cells represent some consumer areas of higher and relevant interest for printed photodetectors. Photodetectors can be easily integrated into polymer films solutions which can be applied to flexible surfaces.

Medical and health care, Internet-Of-Things, Industry 4.0 are some of the examples of the areas where the printed photodetectors can have an important role, allowing the creation of high value services for these industries. New user interfaces based on gesture recognition for connected objects, consumer products, intuitive user interfaces are some examples to take into account. Concerning the medical applications, large area X-ray medical imaging for applied to digital imaging body and dental are one of the most investigated applications on this area.

Applications for consumer electronics such as smart phones, laptops and tablets, fingerprint scanning for personal security are one of the most interesting areas to explore in terms of printed photodetectors. Regarding to diagnostic industry, one of the most relevant application should pass for the link of a photodetectors array for detection associated with microfluidics and Lab-on-Chip technologies, which can show a very important role on the healthy science.

In this review we have attempted to summarize the last important works in printed optical devices with a special attention on printed photodetectors.

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