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Biodegradable all-polymer field-effect transistors printed on Mater-Bi

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ABSTRACT

The growing demand of disposable electronics raises serious concerns for the corresponding increase in the amount of electronic waste, with severe environmental impact. Organic and flexible electronics have been proposed long ago as a more sustainable and energy-efficient technological platform with respect to established ones. Yet, such technology is leading to a drastic increase of plastic waste if common approaches for flexible substrates are followed. In this scenario, biodegradable solutions can significantly limit the environmental impact, actively contributing to eliminate the waste streams (plastic or electronic) associated with disposal of devices. However, achieving suitably scalable processes to pattern mechanically robust organic electronics onto largely available biodegradable substrates is still an open challenge. In this work, all-organic and highly flexible field-effect transistors, inkjet printed onto the biodegradable and compostable commercial substrate Mater-Bi, are demonstrated. Because of the thermal instability of Mater-Bi, no annealing steps are applied, producing devices with limited carrier mobility, yet showing correct *n*-type behavior and robustness to bending and crumpling. The degradation behavior of the final system shows unaltered biodegradability level according to ISO 14851. These results represent a promising step toward sustainable flexible and large-area electronics, combining energy and materials efficient processes with largely available biodegradable substrates.

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Organic field-effect transistor; biodegradable electronics; Green electronics; flexible electronics; printed electronics

1. Main text

1.1. Introduction


Solution-processable organic electronic devices have recently experienced a steady increase in performances, show great versatility towards the design of applications, and combine cost-effective flexible substrates and easily scalable manufacturing techniques, such as printing [1–4]. Therefore, they have a great potential toward low-cost and low carbon footprint manufacturing processes for a wide variety of consumer products [5–7], ranging from photovoltaics to distributed electronics, such as internet-of-things (IoT) and bioelectronics [8–12].

One of the main targets of organic large-area electronics is the development of novel, short life-cycle and/or disposable applications. Therefore, in concrete terms, there is an inherent risk to produce a severe environmental impact by generating an increased plastic waste, mainly deriving from non-degradable plastic substrates, rather than offering a more sustainable technology. As a


consequence, given the very high target volumes, in order to guarantee a sustainable development, it is also necessary to address the issue of organic electronics end-of-life.

Plastic is in fact one of the peculiar elements that has characterized the last decade, with a continuously growing presence in our everyday life and consequently in the waste produced. Every year around 250 million tons of commodity plastics are produced [13], and most of these are long-lasting polymers, which will thus accumulate in the environment for decades if not properly disposed [14,15], and then will degrade into smaller fragments and migrate toward the oceans, ending up in the so-called garbage patches [16].

In the last decade, the European Union, among others, has promoted the adoption of biodegradable materials as a valid alternative for most of the non-degradable commodity polymers. Moving in this direction is a mandatory requirement also in the framework of organic electronics for a sustainable future pervasive technology, especially serving IoT. The predicted impact is wide,

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since biodegradable devices can be used for medical applications, during diagnosis, medication and treatment [17], for environmental sensors [18], ambient intelligence for daily-life assistance [19], soft robotics [20], food packaging [21], throw-away devices and security applications [22].

A field-effect transistor (FET), which is the most developed organic transistor architecture and a basic building block of active circuits, requires four main components, namely a substrate, a dielectric layer, a semi-conducting film, and conductive electrodes. The substrate is the layer upon which all other components are deposited on and generally amounts for the most of the device volume, and so of the plastic waste generated. In fact, flexible organic FETs (OFETs) and circuits typically employ slow degrading plastic foils, such as polyethylene naphthalate (PEN) or polyethylene terephthalate (PET). Replacing such plastics with biodegradable substrates is therefore an obvious strategy to turn flexible organic circuits into a biodegradable waste at their end-of-life, drastically reducing the potential amount of non-degradable plastic waste. While transitioning to biodegradable substrates, it is not only important to preserve the overall electronic properties, but also the robustness to harsh mechanical stresses, which should characterize OFETs to allow their integration in highly flexible and conformable applications.

Several biodegradable materials have been adopted and tested as substrates for OFETs. Among the natural biodegradable substrates, paper has been employed in several works because of its large availability and reduced cost [13–20]. Nanopaper has also been reported, thanks to its improved optical and mechanical properties [21–25]; only few works include a characterization of the biodegradation of nanopaper, yet not always complete or representative of the natural environment [26–29]. Other natural substrates, which are known to be biodegradable, but for which no actual assessment of the biodegradability after fabrication of the devices has been provided, include silk [30–34], gelatine [35,36], shellac [37–39], and polysaccharides [27,40], whose main example is Ecoflex© [35,36,41]. Other exotic substrates such as caramelized glucose [35,36], rice paper [42,43], wax [44], leather and fabric [19], collagen [45], agarose [46], chitosan and chitin have also been reported [47–51].

Next to these materials, synthetic polymers for biodegradable substrate applications have been presented in literature. The three main examples include polydimethylsiloxane, a well-known biocompatible and biodegradable polymer [52–54], polyvinyl alcohol [47,55–60], and polylactic acid [43,61]. Other synthetic polymers employed as substrates for biodegradable electronics are poly-lactic-go-glycolacid [62–64],

polycaprolactone [65], poly(1,8-octanediol-co-citrate) [66], polypropylene carbonate [67], polyvinylpyrrolidone [68] and polyhydroxyalkanoates [69], poly(3-hydroxybutyrate) and its copolymer with poly(3-hydroxyvalerate) [70–72].

Despite the large number of examples, in most cases no test on the actual degradation behavior of the final devices, which include the fabricated organic components, is performed. A proper evaluation of the effect of the electronic devices on the degradability of the substrate needs to be assessed case by case. On top, it is clear that the use of commercial biodegradable materials, largely present on the market at very low cost and mass volumes, would be inherently advantageous, with respect to custom-made substrates typical of previous works, in terms of more immediate application potential.

So far, the OFETs fabricated on biodegradable substrates comprise the use of metallic contacts and lithographic steps or other vacuum deposition processes. The demonstration of all-organic OFETs deposited at low temperature on the substrates by exploiting printing techniques for the semiconductor and electrodes would allow to combine sustainability of the substrates with the low carbon footprint of additive processes.

In this work, all-polymer *n*-type OFETs based on a model co-polymer has been fabricated on a widely adopted, commercial, certified biodegradable and compostable material, Mater-Bi [73,74], serving as substrate. Specifically, largely available compostable bags were adopted, as those currently sold in supermarkets for the transport of fruits and vegetables, conform to the UNI/EN 16640/2017 [75,76].

The OFETs have been fabricated on a $\sim 10\ \mu\text{m}$ thick Mater-Bi substrate, without any surface modification, achieving a good reproducibility thanks to the combination of two scalable techniques. We adopted inkjet printing for the patterning of the semiconductor and of the electrodes, while we made use of the characteristic chemical vapor deposition process of Parylene C to form a compact, pinhole-free dielectric with the aim of achieving mechanically robust devices [77,78]. Because of thermal instability of Mater-Bi, no annealing steps were applied during fabrication of OFETs, limiting the carrier mobility to $0.01\ \text{cm}^2/\text{Vs}$ in saturation regime. Indeed, a wide range of applications demands unconventional form factors and requires or foresees rolling, wrinkling, bending with sharp edges, and even crumpling upon handling [79]. To this purpose, the thickness of the substrate plays a key role, as the mechanical stress is inversely proportional to it [80,81]. Differently from most substrates commonly reported for the realization of OFETs, the adoption of a $10\ \mu\text{m}$ -thin Mater-Bi substrate enables the combination of mechanical robustness and

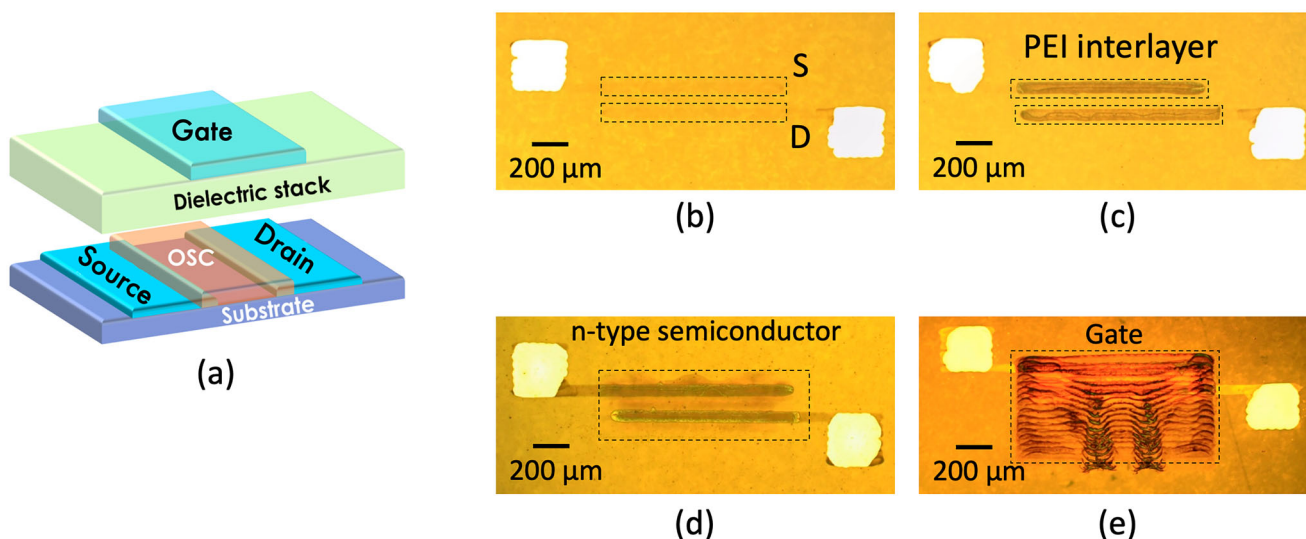


Figure 1. (a) Schematic of the device structure. Optical micrograph of the fabrication steps: (b) PEDOT:PSS source and drain electrodes, (c) PEI interlayer, (d) P(NDI2OD-T2) semiconducting area (indicated as OSC) and (e) PEDOT:PSS gate electrode. Silver pads for external electrical contacts can be seen as well; such pads are not necessary for the operation of the device and provide an optical guide for electrical probing.

biodegradability. Rolling and crumpling tests have been carried out and the printed organic transistors on Mater-Bi retain field-effect operation even upon the application of harsh mechanical tests, with a substantially unaffected carrier mobility in saturation after rolling with a bending radius of 1.5 mm, and only a twofold reduction after crumpling.

The results here reported are a first step towards mechanically robust, large-area organic electronics with drastically reduced environmental impact, fabricated with industrially viable additive technologies on commercial biodegradable substrates, contributing to the sustainable spreading and integration of intelligence and sensing in objects of daily use.

1.2. Results and discussion

Mater-Bi films obtained from commercial bags have been laminated on a rigid glass carrier to facilitate the handling during the processing. Mater-Bi presents a strong sensitivity to thermal treatments, already at temperatures as low as 50°C. As a consequence, the OFETs fabrication procedure has to respect a very limited thermal budget, where typical thermal annealing steps in OFETs fabrication are replaced with vacuum drying of the printed materials.

Solvent resistance tests have also been carried out, exposing the material to candidate solvents for printable formulations of the functional inks. In particular, Mater-Bi films show stability against water, allowing to adopt water based poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) formulations for

the patterning of the conductors, and mesitylene, which can be adopted for semiconductor ink formulations. As model semiconductor, we adopted the very well-known, good electron transporting poly[N,N'-bis(2octyldodecyl)naphthalene-1,4,5,8-bis(dicarboximide)-2,6-diyl]-alt5,5'-(2,2'-bithiophene) [82,83], indicated as P(NDI2OD-T2) in the following.

PEDOT:PSS has therefore been adopted to pattern by inkjet printing source and drain electrodes Figure 1(b) of a bottom-contact/top-gate OFET Figure 1(a) architecture by inkjet printing, with a characteristic channel length (L) of 65 μm and width (W) of 1000 μm (see Figure S1 in SI for the the average resistivity of printed PEDOT:PSS on Mater-Bi). Subsequently, a polyethyleneimine (PEI)-based injection layer was inkjet printed on top of the contacts Figure 1(c), with the aim of favoring the electrons injection into the semiconductor thanks to a reduction in the work-function [84]. The P(NDI2OD-T2) semiconducting layer Figure 1(d) was then inkjet printed to form the conductive channel. Afterwards, a Parylene C dielectric layer has been deposited via chemical vapor deposition, with a thickness of 240 nm, which leads to resulting in an areal capacitance of 10.7 nF cm⁻². Lastly, PEDOT:PSS gate electrodes have been inkjet printed Figure 1(e) on top of the dielectric layer. After each printing step, vacuum drying has been carried out, with the samples kept in vacuum for about 10 min to remove the solvent.

The electrical characterization of the printed n-type transistors has been carried out in a glove box with nitrogen atmosphere. Both transfer and output curves have been measured, as shown in Figure 2.

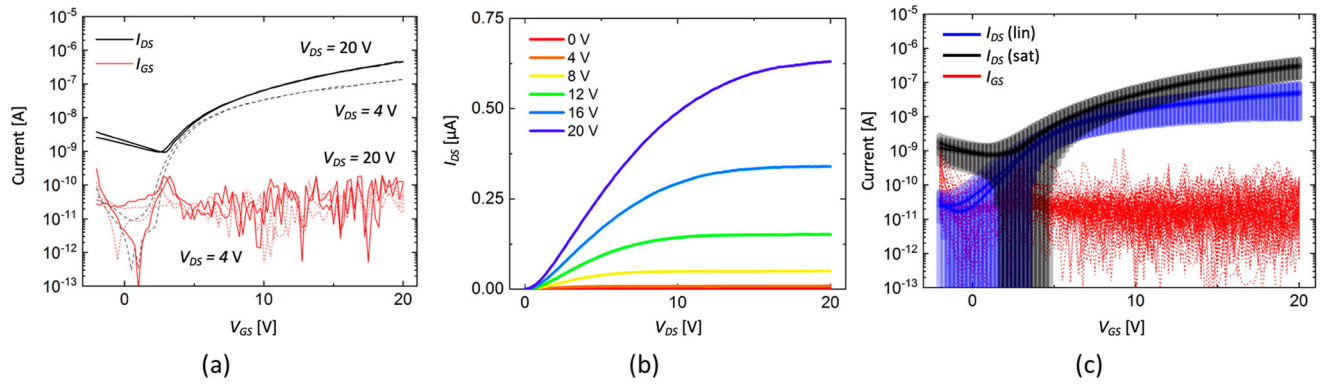


Figure 2. Electrical characterization of all-polymeric *n*-type OFETs printed on a biodegradable Mater-Bi substrate. (a) Transfer and (b) output characteristics for a single transistor. (c) Average transfer curve obtained from the characterization of 20 printed OFETs.

The supply voltages needed for the proper operation of these devices are in the range of 20 V. Figure 2(a) shows the proper field-effect modulation of the channel current and low leakage current, flat and in the 100 pA range. Focusing on the output curve, in Figure 2(b), small non-idealities related to charge injection can be highlighted, namely a mild S-shape of the curve is visible, typically indicating injection limitation. Nevertheless, the *n*-type OFETs printed on Mater-Bi substrates are properly functioning.

The reproducibility of these devices has been tested, and 20 OFETs, printed on a single (2×7) cm² substrate, have been characterized; the average transfer curve is shown in Figure 2(c). All the devices are properly working, with a yield of 100%, and they show an average maximum current equal to (330 ± 140) nA, and leakage curves that are laying below 100 pA on average. While both the standard deviation of the average transfer curve and the noise in the leakage current appears higher than for conventional, highly engineered substrates such as PEN [85], the overall yield and reproducibility show that the fabrication of all-polymer OFETs can be adapted onto a commercial Mater-Bi substrate.

The average field-effect mobility has been calculated, according to the gradual channel approximation [86], and it is equal to ~ 0.005 cm² V⁻¹ s⁻¹ and ~ 0.01 cm² V⁻¹ s⁻¹ in linear and saturation regimes respectively (Figure S3(a)). As reported in Figure 2(a), the average on-off ratio for these devices, calculated as the ratio between the on current ($V_{GS} = 20$ V) and the off current ($V_{GS} = 0$ V), is $> 10^5$ in linear regime (dashed line) and $\cong 300$ in saturation regime (solid line), due to a significant parasitic p-type current occurring at high V_{DS} voltages and low V_{GS} voltages. The subthreshold slope average value is equal to (685 ± 274) mV dec⁻¹, with the best performing device showing a subthreshold slope equal to 327 mV dec⁻¹. The average onset and threshold

voltages are equal to (2.36 ± 0.95) V and (2.74 ± 1.69) V, respectively.

While granting satisfying and reproducible performances, these OFETs are performing slightly worse than their counterparts on PEN [85]. There are two main reasons: (i) the absence of thermal annealing steps, known to improve the devices performances [87]; (ii) the dielectric structure employed, since the direct exposure of P(NDI2OD-T2) to Parylene C leads to a slight performance worsening, as shown in Figure S2. For the latter, thin interlayers, such as ultrathin PMMA [85], could be introduced to improve the electrical performances, if suitable processing solvents, compatible with Mater-Bi, are used.

The mechanical stability of the printed OFETs on Mater-Bi has been assessed through two main tests: rolling and crumpling, shown in Figure 3(a,b).

The devices have been rolled onto a thin plastic cylinder with a diameter of 1.5 mm, then brought back to a flat configuration, and subsequently characterized by measuring the transfer curves Figure 3(c). The same procedure has been followed with a different set of transistors also for crumpling, and the transfer curves measured afterwards are shown in Figure 3(d).

The average mobilities for rolled and crumpled devices have been extracted, and the curves are shown in Figure S3(b,c). Rolled devices have a mobility equal to ~ 0.007 and ~ 0.01 cm² V⁻¹ s⁻¹ in linear and saturation regimes, respectively, while crumpling leads to mobility equal to ~ 0.003 and ~ 0.005 cm² V⁻¹ s⁻¹ again for linear and saturation regimes, respectively. The subthreshold swing is equal to 747 and 627 mV dec⁻¹ for rolled and crumpled devices, respectively, showing no significant variation after the mechanical tests.

Overall, we observe that the drain current for devices is substantially unchanged upon stress caused by rolling, while crumpling causes a slight loss in performances. The latter is ascribable to a higher mechanical stress applied

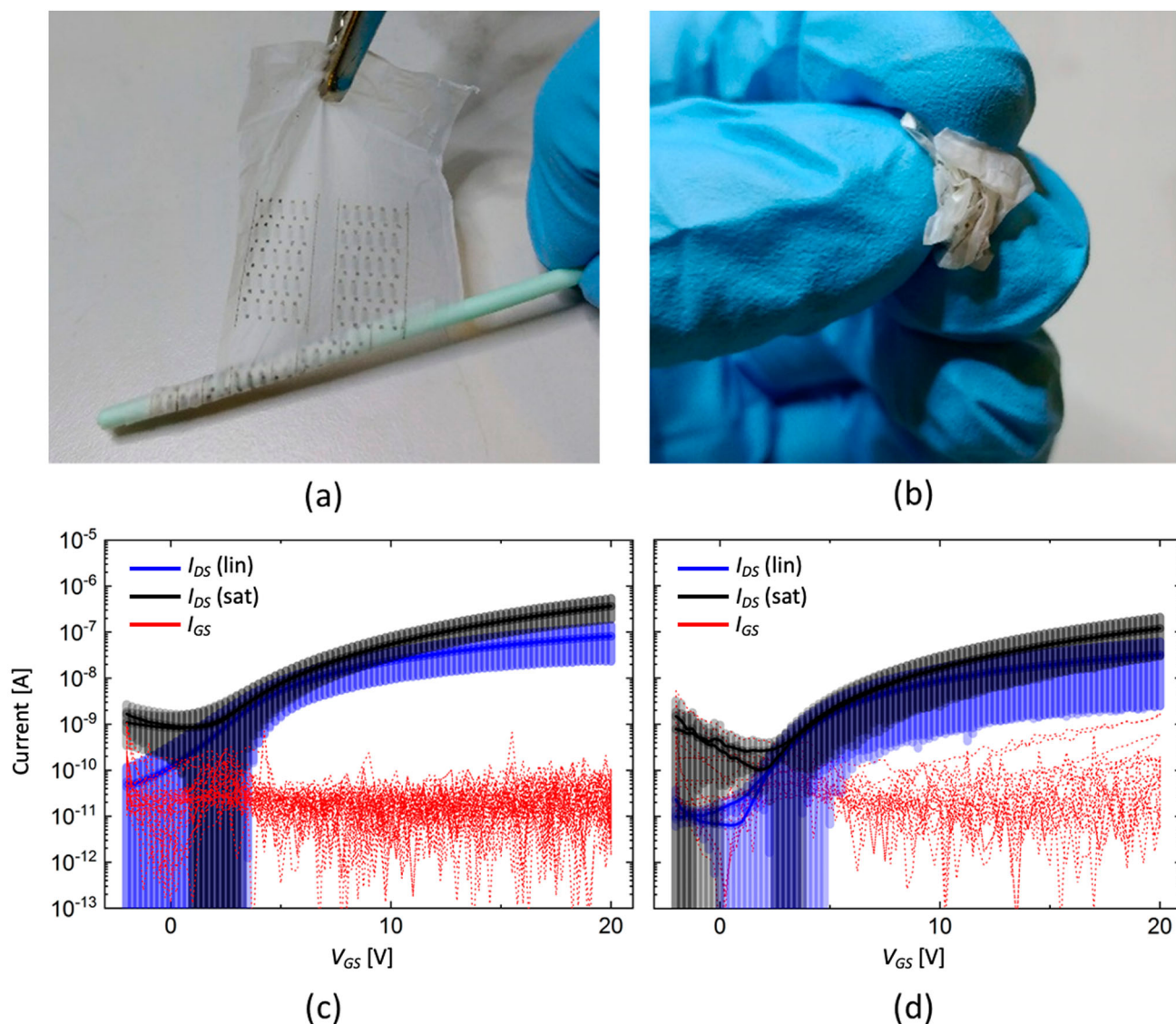


Figure 3. Photographs of the printed OFETs on Mater-Bi during the mechanical tests, (a) rolling and (b) crumpling, and average transfer curves, in linear (4 V) and saturation (20 V) regimes, after (c) rolling and (d) crumpling.

on the active layer deriving from smaller curvature radii produced upon crumpling, which can be as low as few microns [88]. In future, it will be possible to reduce this effect by using thinner substrates [79], or by placing the device in the so-called neutral plane position [80]. The standard deviations for both devices are close to that of as-fabricated OFETs, meaning that the mechanical tests do not harm the performances uniformity.

Having demonstrated the possibility to realize robust organic electronic devices on a widely available commercial substrate, it is important to assess the biodegradability of the final system, comprising the OFETs and the biodegradable Mater-Bi substrate.

In the last decades, multiple organizations have developed definitions and standardized tests for the evaluation of biodegradability in different environments [81,89,90]. Here the biodegradability of the printed devices has been

tested according to the ISO 14851 ('Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium – Method by measuring the oxygen demand in a closed respirometer') [91], which indicates the guidelines for the assessment of the biodegradability of plastic materials in either real or simulated seawater or fresh water. According to the general protocol, the sample is incubated with the chosen liquid medium within a hermetically closed system. In presence of a biodegradable material, microbial colonies, naturally occurring in real environmental water samples, start consuming it: the consequent biotic consumption of the oxygen (Biochemical Oxygen Demand – BOD) available in the system is measured as a function of the decrease in pressure, since a scavenger is added to sequestrate the evolved carbon dioxide. Plotting the detected values of BOD as a function of time results in sigmoidal curves describing the

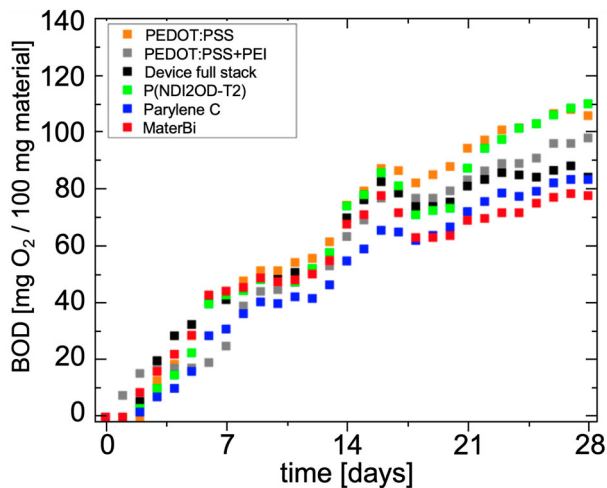


Figure 4. BOD [mg O₂/100 mg material] measured for the pristine Mater-Bi substrate (red), the full-stack device realized on Mater-Bi (black), PEDOT:PSS printed on Mater-Bi (orange), PEI-functionalized PEDOT:PSS on Mater-Bi (grey), P(NDI2OD-T2) printed on Mater-Bi (green) and Parylene C on Mater-Bi (blue).

progressive microbial growth, which directly depends on the biodegradability of the tested material. We performed this test on the pristine Mater-Bi substrate, on the substrate with each OFET component separately (PEDOT:PSS electrodes, PEDOT:PSS electrodes functionalized with PEI, P(NDI2OD-T2) semiconducting layers and Parylene C) and on the device full stack. Since Parylene C dielectric was deposited by means of chemical vapor deposition, it covered uniformly all the area of the Mater-Bi substrate. The other materials in the OFETs (PEDOT:PSS, PEI, P(NDI2OD-T2)) were printed only on specific parts of the substrate: the area of each sample was $(1.5 \times 3) \text{ cm}^2$ with an areal density of OFETs equal to 22 devices/cm². The results of the biodegradability tests are reported in Figure 4.

The curves of oxygen consumption for all the tested samples are almost superimposable. None of the samples shows a reduction in biodegradability compared to control Mater-Bi. On the other hand, small differences among the curves, suggesting apparent higher oxygen consumption for samples with PEDOT:PSS printed on Mater-Bi, PEDOT:PSS + PEI printed on Mater-Bi and P(NDI2OD-T2) printed on Mater-Bi, can be considered artifacts deriving from unavoidable variability of the experiment, due to random temporary folding of the thin films during the test. These results show that our printed, fully organic FETs do not substantially alter the degradation behavior of Mater-Bi.

1.3. Conclusions

Fully organic FETs have been fabricated at room temperature by means of additive manufacturing techniques,

namely inkjet printing and chemical vapor deposition, onto widely available, biodegradable Mater-Bi substrates.

This proof-of-principle demonstration of organic active electronic components on an already commercially widespread biodegradable material, rather than introducing a completely new one, indicates a potentially faster integration of electronic functionalities into everyday products, with minimum modifications and disruptions on the manufacturing processes currently in use.

The thermal budget for the OFET realization on Mater-Bi films has been eliminated, employing vacuum drying instead for a completely room-temperature processing. For such reason, the charge mobility obtained in OFETs based on P(NDI2OD-T2) is lower than what typically achieved on non-degradable plastic foils, such as PEN: nevertheless, the yield, level of reproducibility and mechanical robustness obtained with such all-organic arrays processed at room temperature is promising and provides a good basis for further developments.

Importantly, we directly tested the biodegradation behavior of Mater-Bi samples with fabricated OFETs arrays on top in order to assess the impact of the electronic components on the biodegradability of the Mater-Bi films. Our data indicate that Mater-Bi samples with full-stack OFETs show biodegradability levels comparable with those of the pristine substrate, and no significant variations in the degradation behavior are caused by the presence of a dense array of OFETs (22 devices/cm²).

The results reported here are the first example of fully polymeric, rollable and biodegradable organic transistors, and represent a first step toward the realization of mechanically robust biodegradable electronics onto a Mater-Bi commercial substrate. By demonstrating low carbon foot print additive processes combined with biodegradability at the end-of-life for OFETs based on printed polymers, this works shows a feasible pathway towards sustainable large-area printed electronics applications.

2. Experimental section

2.1. OFETs fabrication

Bottom-contact/top-gate organic field-effect transistors have been realized onto a commercial Mater-Bi film. For the realization of the substrate, biodegradable plastic bags have been bought from the supermarket, and $(2 \times 7) \text{ cm}^2$ rectangles have been cut with the help of a scalpel and attached onto a glass slide for an easier handling during the fabrication of the devices. No treatment of any kind has been performed on the substrate surface, aside from blowing them with a nitrogen gun to remove dust.

Poly(3,4-ethylenedioxi-thiophene):polystyrene sulphate (Clevios PJ700 formulation, purchased from Heareus) source and drain contacts have been inkjet printed by means of a Fujifilm Dimatix DMP2831. The channel of the so obtained transistors has a width of about 1000 μm and a length equal to 65 μm .

Small pads have been printed in correspondence of the points where electrical connections are created during the measurements with a silver. This is needed because the electrode pads are otherwise transparent, and the characterization of the devices would be extremely complex, but the silver pads are not required for the actual operation of the printed transistors.

A PEI-based injection layer has been printed on top of the contacts (Figure 1(c)). This solution is composed of ethylene glycol (20% vol, purchased by Sigma Aldrich), a solution of zinc oxide (ZnO) nanoparticles in isopropanol (nanoparticles concentration 2.5% wt, the solution amounts for 30% of the volume of the final solution) and PEI (branched, purchased from Sigma Aldrich, $M_w \approx 10000$), dissolved in water with a weight concentration of 0.2% (50% vol of the final solution). The presence of PEI, as described by Zhou et al. [84], gives rise to the electron-injecting/hole-blocking features of this solution, thus enhancing the injection of charges in the n -type semiconductor thus improving the electrical performance of OFETs.

P(NDI2OD-T2) (ActivInk N2200, Flexterra corporation) has been used as an organic semiconductor, patterned by inkjet printing Figure 1(d), starting from a mesitylene-based solution, with a concentration of 7 mg ml^{-1} .

The Parylene C dielectric film has been deposited via chemical vapor deposition, using a SCS Labcoater 2 – PDS2010 system.

Lastly, PEDOT:PSS gate contacts have been printed on top of the dielectric layer (Figure 1(e)), again with inkjet printing as the technique of choice.

2.2. Characterization

We performed the electrical characterization (transfer curves and output curves) of the transistors in nitrogen atmosphere using an Agilent B1500A Semiconductor Parameter Analyzer.

Parylene C capacitance was extracted using an Agilent 4294A Impedance Analyzer with a two probes configuration.

2.3. Biochemical oxygen demand

About 6 mg of sample were added to 432 ml of seawater as the single carbon source. The seawater mimics real environmental conditions, as it contains naturally

occurring microbial consortia and the saline nutrients needed for their growth.

The experiment was conducted at room temperature inside dark glass bottles with a volume of 510 ml, hermetically closed with the OxiTop[®] measuring head. NaOH was added to the system, not in contact with the water, in order to sequester carbon dioxide produced during the biodegradation, while the biotic consumption of the oxygen present was measured as a function of the decrease in pressure.

Raw data of oxygen consumption ($\text{mg O}_2/\text{L}$) were corrected, subtracting the mean values of the blanks, obtained by measuring the oxygen consumption of the seawater in absence of any test material. After this subtraction, values were normalized on the mass of the individual samples, referred to 100 mg of material ($\text{mg O}_2/100 \text{ mg material}$) and plotted vs time.

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Ksenija Maksimovic received her BS and MS degree in Materials Engineering and Nanotechnology from Politecnico di Milano, Italy, during which she spent a semester abroad at KULeuven, in Belgium. She did her Master Thesis at the Center for NanoScience and Technology in Milan during which she worked on Printed Organic Electronics based on Edible and Biodegradable materials.



Laura Bertolacci graduated in chemistry in 2009 and earned her PhD in Drug Discovery from the Italian Institute of Technology in 2012. After a two-year experience in a pharmaceutical company, she embarked in a career as a Postdoctoral researcher, working in a wide range of sectors, from biochemistry to material sciences, including nanotechnology, analytical chemistry, microbiology, and biocatalysis. She is currently focused on the field of sustainability and the evaluation of materials biodegradation.



Fabrizio Antonio Viola received his MS degree in electronic engineering in 2014 and earned his PhD in Bioengineering and Robotics, working on multimodal tactile sensors based on organic field-effect transistors – under the supervision of Prof. Annalisa Bonfiglio – from the University of Genova in 2018. He is currently

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Mario Caironi obtained his PhD in Information Technology with honors at Politecnico di Milano (Milan, Italy). In 2007 he joined the group of Prof. Sirringhaus at the Cavendish Lab. (Cambridge, UK) as a post-doc, working for three years on high-resolution printing of downscaled organic transistors and circuits and on charge transport in high mobility polymers. In 2010 he was appointed as Team Leader at the Center for Nano Science and Technology@PoliMi (CNST) of the Istituto Italiano di Tecnologia (IIT, Milan, Italy), obtaining tenure in 2019. He is currently interested in solution-based high-resolution printing techniques for micro-electronic, opto-electronic and thermoelectrics devices, in the device physics of organic semiconductors based field-effect transistors, in biomedical sensors and electronics for the healthcare. He is a 2014 ERC Starting grantee and a 2019 ERC Consolidator grantee.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

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