Organic Permeable Base Transistors – Insights and Perspectives

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Dedicated to Professor Karl Leo on the occasion of his 60th birthday

Vertical organic transistors have emerged as a new device concept holding great promise to overcome the limitations of lateral organic transistors. In this regard, the organic permeable base transistor (OPBT), a special kind of vertical transistor, stands out due to excellent performance figures such as low-voltage operation and high transition frequency measured in a pulse-biasing mode. On the occasion of Prof. Karl Leo’s 60th birthday, his contributions to the development of high-performance OPBTs are honored and a perspective for the future of OPBT development is provided. The current state of the art of OPBTs is reviewed and the principles of the device operation are summarized. New insights into the formation of the permeable base layer, which is vital to the function of OPBTs, are discussed. Additionally, a full yield analysis on a batch of 144 equivalent OPBTs is reported and the device yield and temporal evolution of device parameters are statistically analyzed. This analysis proves a device yield of >90% for these short-channel transistors, rendering the possibility for circuit integration. Finally, the scaling laws of OPBTs are derived and strategies for new materials and device layouts to push the performance to the gigahertz regime are concluded.

1. Introduction

The vision of flexible electronics is to enable lightweight, cost-effective, power-efficient, and versatile electronic devices that can support people in any aspect of life. In this context, organic semiconductor materials are an interesting material class due to their low processing temperature and inherent mechanical flexibility. Unfortunately, besides a couple of prototypes and first landmark products, truly flexible electronics remains a vision for the future. The biggest bottleneck which impedes the rise of organic-semiconductor-based flexible electronics is the challenge to fabricate organic thin-film transistors (OTFTs) that can operate at low voltage (<5.0 V), which possess high on/off ratios (>10^6) and work at high frequencies. In particular, the highest possible operation frequency, given by the unity-gain transition frequency, is an important figure of merit, and ideally, it should be above 100 MHz in order to enable wireless communication and other demanding applications such as active-matrix display driving.

In the last decade, the OTFT community focused on improving the charge carrier mobility in organic semiconductor materials, which is nowadays in excess of 10 cm^2 V^-1 s^-1. Recently, various groups have demonstrated the fabrication of OTFTs with switching operations at a few tens of a megahertz, e.g., Sawada et al. reported a solution-processed organic single-crystal transistor with f_T = 45 MHz at 7 V. Recently, Perinot et al. also reported a solution-processed organic transistor with f_T = 160 MHz at 40 V. The recent achievements open a route for high-frequency OTFTs, but they still need further optimizations in order to go toward industrial production (e.g., fabrication on flexible substrates). The origin of that problem lies in the contact resistance that increasingly contributes to the overall device resistance when the channel length L is scaled down. As a consequence, the effective charge carrier mobility steeply drops for decreasing channel length, ultimately causing the transition frequency to level off with the channel length. Furthermore, when the channel length is reduced, parasitic capacitances originating from the unavoidable gate–source and gate–drain overlap L_{OV} add to the total device capacitance, further impeding a transition frequency increase. Still, reducing the effective channel length and device capacitance seems to be the most effective way to increase the transition frequency (f_T = 1/(L^2 + L_{OV}). The effectiveness of this approach has been successfully demonstrated by Perinot et al., who reported a transition frequency of 160 MHz for a channel length of 1.2 μm and an overlap length of ~250 nm. However, these devices still require a high operation voltage (40 V), high thermal conductivity, rigid substrates, and very high resolution of patterning, which might be challenging to achieve on a flexible substrate.

Vertical organic transistors are an alternative device concept to realize short channel length thin-film transistor (TFT).
these structures, the channel length is not defined by a patterning technique such as lithography or printing but rather by the thickness of the vertically stacked layers. Thus, since layer thicknesses in organic semiconductor devices can be controlled with nanometer precision, the short channel length can be manufactured conveniently. Among all the different vertical transistor designs (e.g., Schottky barrier vertical organic field-effect transistors,[12,13] vertical organic field-effect transistors,[14,15] electrolyte-gated vertical organic transistors,[16–18] organic static induction transistors[19–21]), organic permeable base transistors (OPBTs) stand out due to their excellent performance figures such as a transition frequency of 40 MHz measured in a pulse-biasing mode at $V_{CE} = 8.6$ V,[22] on/off ratio $> 10^8$, current densities as high as 1 kA cm$^{-2}$,[23] and favorable stability.[24] Furthermore, the research on OPBTs is rapidly progressing in the last couple of years, which led to innovative devices,[25] new fabrication methods,[26] and even complementary circuits.[27] However, there are still several problems to solve, such as integration into circuitry and improving device yield and reliability.

In the last decade, Leo and co-workers coined this field,[22,25,33] and on the occasion of Karl Leo’s 60th birthday, we honor his work on OPBTs and highlight current developments in this field of research. Hence, in this publication, we review the current state of the art of OPBTs and summarize the principles of the device operation. We discuss the importance of the base electrode for device operation by providing novel insights into the base layer formation, structure, and composition. Moreover, we report a full yield analysis on a batch of 144 equivalent OPBTs and statistically analyze the device yield and temporal evolution of device parameters. The in-depth understanding of the device operation in combination with the learning from the yield analysis is used to draw a vision for the future of OPBTs. Finally, we predict the performance limits and derive strategies for new materials and device layouts in order to further push the performance possibly to the gigahertz regime.

2. Understanding the Operation of OPBTs

2.1. From Vacuum Tube to Organic Solid-State Triodes

The OPBT concept was inspired by vacuum tube triodes and solid-state triodes based on inorganic semiconductor materials.[28,29] The vacuum tube triode consists of three electrodes, namely cathode, anode, and a grid-like middle electrode, placed in a high vacuum. All charge carriers in the vacuum tube triode originate from the hot cathode, and the current flow from cathode to anode is modified by the grid-like middle electrode. Hence, when transferring the triode concept from vacuum to solid state, it is necessary to embed a strip or mesh electrode, which corresponds to the grid-like electrode in a vacuum tube. It is important that the fine mesh structure of the middle electrode can effectively modulate the potential barrier height against the carrier flow. The basic structure of an OPBT is shown in Figure 1a. Most commonly, OPBTs consist of three electrodes and at least two organic layers. The electrodes are denoted as emitter, collector, and base corresponding to the cathode, anode, and grid electrode in vacuum triodes. The base electrode

![Figure 1. a) Scheme of an n-type OPBT. The transistor consists of metallic emitter, base, and collector electrodes and at least two organic layers. b) Energy diagram of an OPBT in the on-state. With applied base–emitter voltages ($V_{BE}$), electrons can pass through the pinholes in the base and hence reach the collector. Except for a minor leakage current into the base $I_B$, the emitter current $I_E$ can finally arrive at the collector, defining the collector current $I_C$. c,d) Simulation of an n-type OPBT. (c) A transfer curve: fixed emitter potential at 0 V and fixed collector potential at 1.0 V. The base potential is varied. Three different regions can be identified. (d) Electron density and electric potential for full simulation cell (top) and the detailed view around the base (bottom), for base potentials corresponding to regions I ($V_{BE} = 0.5$ V), II ($V_{BE} = 1.0$ V), and III ($V_{BE} = 1.5$ V), as marked in (c). Reproduced with permission.[30] Copyright 2016, AIP Publishing.](image-url)
is permeable for charge carriers, i.e., charges injected at the emitter can pass the base electrode and arrive at the collector. The transmission of charges through the base can be modulated by the potential of the base electrode, influencing the current injected at the emitter. Thus, similar to a vacuum tube triode, the base modulates the current flowing from emitter to collector.

The thickness of the organic semiconductor layers in state-of-the-art OPBTs is in the range of 100 nm, resulting in a total device thickness of typically <300 nm, and as we will discuss, the dimension of the channel region of OPBTs are even much smaller than 300 nm, related to the oxide surface around one pinhole. Compared to lateral-channel OTFTs where the channel length is typically in the range of >1 μm, OPBTs are true short-channel devices that can carry a very large current density. The structure and morphology of the base electrode are essential for the device operation, which will be discussed later. For most OPBTs discussed in the literature, the base electrode is a thin metal film that contains nanometer-size pinhole openings, allowing the current to flow. Furthermore, the pinholes are separated from the metallic base electrode by a thin oxide layer (AlOₓ, thickness 1–2 nm), which is vital to suppress undesired leakage current into the base. The permeable base transistors (PBTs) with polymer grid network was first made by Yang and Heeger in 1994. In 1998, Kudo et al. proposed the concept of metal base transistor, and later, the focus shifted to p-type OPBTs. In a series of publications, Leo and co-workers optimized the n-type OPBTs as proposed by Fujimoto and co-workers and today, OPBTs are the promising alternative organic transistors.

2.2. Operation Mechanism of OPBTs

Kaschura et al. developed a comprehensive understanding of the operation mechanism and charge transport in OPBTs (cf. Figure 1b–d). The basic working mechanism of an OPBT can be understood as follows: For n-type devices, a low base potential represents a high energy barrier for electrons and almost no electrons can pass the base, leading to the off-state. For high base potentials (i.e., the base potential is positive with respect to the emitter), a current is injected at the emitter. At the base, most of this current is transmitted from emitter to collector, the device is in its on-state (cf. Figure 1b). However, the emitter current is not completely transmitted across the base, a fraction of the emitter current is drained at the base, resulting in a current Iₑ flowing from the emitter electrode to the base electrode. The transmitted current is finally collected by the electric field generated by the base—collector diode operated in reverse, giving a current IₑC. The opposite is the case for p-type devices.

By using 3D drift-diffusion simulations, the operation of OPBTs can be divided into three regions according to the base potential (cf. Figure 1c,d). For low base potentials (region I), the openings (pinholes) in the base layer limit the current, yielding an exponential law similar to the subthreshold regime in lateral TFTs. Region II can be understood as a transition region, where the charge channel region forms around the base oxide and within the pinholes. For high base potentials (region III), small changes in the applied base potential have no influence anymore, since the base is shielded by a charge accumulation around the base insulator, and the current is defined by the charge transport through the intrinsic semiconductor layers. Therefore, at low current densities, the applied base potential controls the number of charges that can pass through the pinhole openings and the openings are the current limiting factor. However, at higher current densities, charges accumulate within the openings and in front of the base insulation, allowing for the efficient lateral transport of charges toward neighboring openings. The on-state in the current–voltage characteristics reaches the maximum possible current given by space charge limited current (SCLC) transport through the intrinsic semiconductor layers. Thus, even a small effective area of the openings can drive huge current densities. The channel region of the OPBT is a complex combination of the pinhole and a 2D area along the oxide on the other side of the base. Hence, the function of the OPBTs actually relies on the field effect, which is similar to OTFTs, and thus the electrode emitter, base, and collector could be also called source, gate, and drain, respectively.

Since a large current flow from emitter to collector (up to 1 kA cm⁻²) is controlled by a small base current (ranges from 10⁻⁷ to 10⁻¹ A), the current gain β and transmission factor α can be defined as β = IₑC/Iₑ, and α = IₑC/IₑB, respectively. Thus, α and β are related by β = α(1 − α), and the OPBTs have to show a large current transmission in order to reach a large current gain. In the following subsection, different approaches employed to optimize the performance of OPBTs are summarized.

2.2.1. Device Optimization

The emitter and collector electrodes as well as the charge carrier injection are optimized at first to reduce the contact resistance as much as possible. For C₆₀-OPBTs, a combination of aluminum and chromium has been shown to give the lowest contact resistance to C₆₀ layers. Furthermore, the contact resistance between metal and semiconductor can be further reduced by inserting an additional injection layer of C₆₀-n-doped by Wₓ(pp)₄ (n-C₆₀), underneath the emitter electrode (cf. inset of Figure 2a). Using this injection layer, Klinger et al. demonstrated that C₆₀-OPBTs are purely limited by the SCLC in the intrinsic organic semiconductor layer, which can be understood as an absolute lower limit of the contact resistance. In addition, the currents are demonstrated to scale with a reduced effective length, which is defined as [LₑC/(LₑC)²]²/3 (Lₑ and LₑC are the intrinsic layer thicknesses on top and bottom of the base, respectively). As the major limitation of the current is revealed to be a SCLC, devices have to be as thin as possible, always chosen as a trade-off between performance, device stability (electrical shorts), and reasonable breakdown voltages.

Although the vertical electrode configuration enables a short channel length, there is also a downside of this architecture – obviously an undesired overlap capacitance and restrictions in device scaling. To circumvent these problems, an improved structuring technique for OPBTs was proposed to scale down the active area, so that the influence of the series resistances and overlap capacitances of the electrodes diminishes. To realize a high-performance but simple transistor, Klinger et al.
introduced an indirect structuring method, illustrated in the inset of Figure 2a. An additional insulating layer (either SiO or the poorly conductive organic material 2,2′,7,7′-Tetra(N,N-di-p-tolyl)amino-9,9-spirobifluorene (Spiro-TT)) with a stripe-like open window is patterned on top of the base to define a quadratic active area in conjunction with the narrowed emitter electrode. With such an indirect structuring method, the off-current of OPBTs, leakage current, parasitic electrode resistance, and the other parasitic effects such as charging of nonoverlapping areas by lateral transport are effectively reduced.\[9,10\]

Based on the above device geometry optimization, the performance of OPBTs is significantly improved. As seen in Figure 2a, at a collector–emitter voltage of 7.0 V, current densities above 1 kA cm$^{-2}$ can be achieved, despite processing without any high-resolution structuring. Here, similar to transistors made from many other materials, e.g., amorphous silicon,\[40\] low temperature polysilicon,\[41\] oxide semiconductors,\[42\] as well as 2D semiconductors, electrothermal feedback also occurs in OPBTs. At a current density of 40 A cm$^{-2}$ and a $V_{CE}$ of about 3.6 V, the transistors amplify signals up to 11.8 MHz (cf. Figure 2b). The frequency–current dependence predicts a transition frequency $f_T$ in the range of 100 MHz if a current density of 1 kA cm$^{-2}$ was applied. However, at such high current densities, self-heating of the device sets in (cf. Figure 2c), and $f_T$ measurements at steady state become difficult. Thus, a pulse-biasing characterization circuit was built by Kheradmand-Boroujeni et al., which can turn on the organic transistor and apply an accurate bias $I–V$ to it in less than 10 μs, then apply a small sinusoidal signal to the device and measure several small-signal parameters such as transition frequency, transconductance, and then quickly turn off the device again. By using a pulse-biasing small-signal measurement technique, a transition frequency of 40 MHz is achieved at $V_{CE} = 8.6$ V (cf. Figure 2d),\[22\] which might be further improved employing high-mobility semiconductor materials. Based on Klinger et al.’s work,\[39\] even small temperature changes can significantly change the charge carrier mobility as well as the conductivity of these materials, leading to an increasing performance during Joule self-heating. Thus, operation at a controlled level of self-heating can also be an option to realize electronic devices with much higher switching frequencies. This is especially interesting for high frequency applications where the processed signals are much faster than the thermal system. Thus, self-heating can become a natural way to boost the highest currents and frequencies and extend the operation and thus the application range.

2.2.2. Function and Morphology of the Base Electrode

The function of OPBTs is mainly influenced by the nanometer-thin base electrode and its even thinner oxide layer (typical thickness 1–2 nm). It is observed that devices based on C$_{60}$, which have
the highest performance so far, only work after they have been thermally annealed. Specifically, devices show a strong leakage current into the base electrode and very little charge carrier transmission to the collector electrode before they are annealed. After a heat treatment at 150 °C for 2 h, the base current decreases, while the collector current increases, consequently vastly improving the gain and on/off ratio of the transistors.[34]

In principle, this effect might originate from two independent mechanisms. If there is a strong improvement in the insulating properties of the base oxide during annealing, the base current will decrease, going along with an increase in charge carrier transmission. Likewise, it is possible that the pinhole channels through the base electrode become conductive during annealing, hence rendering the base permeable for electrons. This effect will cause an increase in transmission and enable electrons to reach the collector electrode before falling into the base.

In order to find out which of these two mechanisms is responsible for the performance boost during annealing, and to obtain a better understanding of the nanoscopic processes, thin aluminum films of varying thicknesses have been prepared and the electrical sheet resistance, as well as the optical transmittance have been measured before and after the annealing. In order to provide similar morphology as in an actual OPBT, an underlayer of 100 nm C<sub>60</sub> is used. The results can be seen in Figure 3, with sheet resistance and transmittance represented by the unfilled and filled squares, respectively. Since aluminum oxide is transparent and nonconductive, any change in the oxidation of the base electrode during annealing would be visible in this experiment. Since no such process can be observed in either measurement, thus, the annealing treatment may not affect the insulation of the base layer but make the it permeable to charge carriers, thereby opening up the transistor channels through the pinholes (cf. Section S2 in the Supporting Information). This finding is also supported by the X-ray photoelectron spectroscopy (XPS) measurements shown in Figure 4. This surface-sensitive technique is applied to a thin aluminum film deposited on top of a C<sub>60</sub> layer. A significantly increased signal from carbon bonds can be seen after annealing, thus we conclude that pinholes in the base electrode thin film are filled with C<sub>60</sub> semiconductor material during annealing, making the nanoscopic pinhole channels conductive.

Since we are dealing with very thin self-assembled layers, there are statistical processes involved and not every device is the same. We have previously been able to produce hero devices, always stating that there is a wide spreading in performance of nominally identical devices. In order to shed light on the statistical outcome of the thin-film assembly processes and to quantify the lab-scale production yield of OPBTs, we present a time-resolved observation of 144 OPBT devices produced on the same day.
3. Device Statistical Analysis

In previous publications on OPBTs, the focus has been mostly on the performance of the best devices fabricated, sometimes referred to as hero devices. To complement that, here, an analysis on a series of measurements is conducted with a batch of 144 OPBTs. This analysis gives an insight into the performance development during the first day after production and into the yield of the OPBT fabrication.

3.1. Methodology, Reliability, and Yield

For this analysis, a batch of 36 samples with four pixels each (144 transistors in total) was first processed in the same way as previously done using ambient air to oxidize the base layer. For each device, the transfer curves were measured spanning from a base–emitter voltage of −1.0–1.5 V. On the first day after processing (day 1), all of the transistors were measured before any annealing was done. Starting from the next weekday (day 4), the devices were separated into six equal groups depending on the first digit of the sample and thermally annealed on a hot plate at different times. The “0” devices were never annealed, the “1” devices were annealed for 5 days in a row, the “2” devices started their annealing on day 5 and were annealed for 4 days in a row, and so on, with the last set of devices “5” only annealed once on day 8. A schematic of the annealing days for each sample can be seen in Section S3 (Supporting Information). The annealing was done on a hot plate at 150 °C for 2 h. Once a sample was annealed, we continued the annealing on the following days. A measurement of all transistors was done again on day 26 to observe long-term effects. On that day, no annealing was done. Figure 5 shows a summary of the transistor working status in all measurements.

From the results, the following data can be extracted. 6 devices never worked and 11 devices in total never worked after annealing (4.2% and 76%, respectively). Some of the unsuccessful measurements can be attributed to contact problems in the measurement setup, which were not taken into account for the statistical analysis in Section 3.2 but are included in the calculation of the yield.

3.2. Evolution of Key Parameters

In Figure 5, it is only considered whether a device works or not, but not how well it works. For this reason, further analysis of the data is done by analyzing the distribution and trend of the on/off ratios and the maximum gain values as a function of time. The results are plotted as box plots and are shown in Figure 6. For the on/off ratios (cf. Figure 6a), the median remains approximately constant from day 4 to 8 with an expected variability of the interquartile range (IQR) as samples with different annealing days are measured on the same day. A considerable increase is seen on day 26, which comes with a further increase in the width of the IQR. The existence of the so-called hero devices is evident on this plot, as there is a considerable number of outliers with a higher ratio.

Figure 6b shows that the gain values exhibit a similar trend to the on/off ratios. In this case, a slight increase in the median is also observed from day 4 to 8. On day 26, this increase is considerably larger and the presence of hero devices is more prevalent. The presence of hero devices depends on the pinhole formation during the annealing process and is mostly associated with the annealing of the first day. The repeated annealing steps did not change the performance in a statistically significant way. These results indicate that the presence of hero devices is not a special case but that a group of overperforming devices is generally present. In OTFTs, the gain values can be very high in the range of 10^8 and often are only limited by the measurement instrument (e.g., measurement accuracy for the gate leakage current). These values are high enough for typical applications. The OPBT devices with an oxide made by air exposure can reach a gain in the range of 10^3–10^4. However, investigations about the OPBTs where the base electrode is anodized using citric acid achieved higher values in the range of 10^5–10^6, which is already acceptable for typical applications like driving display pixels. Based on the simulation, we know that the base current is not needed to allow for the operation of the transistor. The base leakage is a pure parasitic effect and once the gate is even better insulated, the performance of the OPBTs can be on par with OTFT in terms of gain. From this analysis, the overall device yield is higher than 90% and the devices show good performance figures (on/off and gain) which steadily increase with time. It is still an open question why there is such a large number of hero devices. We hypothesize that this effect might originate from a statistical variation of the level of perfection of the thin AlO_x layer surrounding the base electrode. However, the existence of hero devices proves that it is possible to obtain very high performance and it is a question of defining a correct treatment procedure to make all devices become hero devices in future.

4. Perspective and Outlook

In the last 15 years, research on OPBTs was mainly focused on single n-type devices that have been optimized almost to their inherent performance limits. However, in order to turn OPBTs from a lab curiosity into a mature technology, further development steps need to be taken. These developments concern: a) the evaluation of robust techniques to form the porous base electrode, b) the optimization of the performance of p-type OPBTs to a level comparable to n-type devices (cf. Section S4 in the Supporting Information), c) the integration of OPBTs in (ideally complementary) circuits, and d) the exploration of new semiconductor materials for even higher transition frequencies. In the following subsections, we discuss strategies to address the above-described challenges. As we will show, if these problems could be resolved, OPBTs would be able to operate in complementary circuits with frequencies beyond 1 GHz.

4.1. Integration of Devices

As discussed in Section 3, the yield of n-type devices allows for the integration of OPBTs in advanced circuits. However,
the integration into logic gates (in particular, unipolar architectures), it is desirable to have control over the threshold voltage in order to adjust the tripping point of inverter gates. Since precise channel doping in organic transistors for threshold voltage control is difficult to achieve,\textsuperscript{[45,46]} alternative approaches such as dual gate transistors need to be developed for OPBTs. The main advantage of organic transistors with dual gates/bases is that the threshold voltages can be set as a function of the applied second gate/base bias, which is crucial for the application in logic gates and integrated circuits. However, incorporating a dual gate/base structure into an ultrashort channel vertical architecture represents a substantial challenge. Based on the OPBT device structure, Guo et al. have now succeeded in realizing vertical organic permeable dual-base transistors (OPDBTs), where the dual base electrodes can be used to tune the threshold voltages and change the on-currents.\textsuperscript{[25]}

A schematic of an OPDBT is shown in Figure 7. Similar to single-base OPBTs, an OPDBT also consists of a simple sandwich-like architecture, four parallel electrodes (gray and green) are separated by an organic semiconductor (orange). The quality of the permeable base electrodes in OPDBTs is first investigated by floating one base. The base leakage current is negligible compared to the collector current, which underlines the high quality of the permeable base electrodes. Figure 7 shows the $I_{C-V_{B2}}$ curves of OPDBTs measured with various $V_{B1}$ from 0 to 2.0 V and $V_{C} = 2.0$ V. As the voltage of base1 increases, the on-state current also increases, the peak of the on-current reaches 1.54 A cm$^{-2}$. Importantly, for the base2
sweeps, the transfer curves shift with the applied base1 bias from 0.5 to 2.0 V, so that the \( V_{th} \) also gradually shifts from 0.04 to 0.52 V. As shown in the inset of Figure 7b, the \( V_{th} \) of OPDBTs depends on the base1 bias during the base2 sweep. They show slight device-to-device variations measured over 61 devices. Thus, by introducing a second base layer, the threshold voltages of vertical organic transistors can be reliably controlled.

By integrating a dual-base transistor with a resistive load, a logic inverter, NAND gate, and AND gate circuits are realized experimentally, as illustrated in Figure 7c–e. Unlike conventional unipolar NAND/AND gates consisting of at least two transistors, the NAND/AND gates are implemented by using only one vertically stacked dual-base transistor. The independent base control enables the transistor to work at different states by tuning the voltages of two bases (two inputs). In addition, by integrating n-type OPDBTs and p-type OPBTs, complementary inverters can be realized. The fabricated organic complementary inverters can operate with a fall and rise time of <10 ns, a gain of >20, and a voltage of <4.0 V. Furthermore, complementary inverters with OPDBTs enable precise control of the tripping point and switching point of the inverters, which represents a significant step toward the application of vertical-gate/base transistors in power-efficient organic complementary inverters. The OPDBT concept facilitates the development of high-performance and complex organic integrated circuits. OPDBTs with two inputs simplify the fabrication of integrated circuits without compromising performance, and the reduction of the total number of transistors offers great advantages for integrated circuits. Hence, since OPDBTs represent a compact and technologically simple hardware platform, they offer admirable application perspectives of vertical-channel organic transistors in complex logic circuits. Furthermore, the transition frequency of single-base OPBTs can reach 40 MHz at \( V_{CE} = 8.6 \) V, hence OPDBTs may also have great potential to be used in fast circuits. However, an overall device yield of >90% still somehow limits the utilization of OPBTs in circuits. Hence, additional strategies, e.g., new processes, new semiconductor materials, new geometries, etc., need to be considered to further improve the yield of device fabrication.

4.2. New Processes for OPBTs

The favorable performance of OPBTs is due to their vertical structure and the utilization of the full device area for current injection and transport. Thanks to these advantages, record devices have been achieved even with low quality material films. Thus, there is a large potential for further development and optimization of OPBTs. Apart from the semiconducting material itself, which is the topic of the next section, the passivation of the base electrode is a crucial part of the OPBT device stack, which can be tuned. While conveniently a simple oxidation in ambient air is used to create an insulating layer around the base electrode, more controlled oxidation by wet electrochemical anodization has been introduced recently. The anodization technique allows for a more controlled growth of the base oxide layer at an arbitrary thickness. Such an oxide layer not only displays better insulating properties, leading to negligible base leakage current, but also decreases the device capacitance without compromising the device performance. At the same time, it has been shown that thicker base electrode layers can be rendered permeable for electrons using anodization, thus decreasing the series resistance of the electrode, which opens the door for even larger device operation frequencies. The research on OPBTs with anodized base electrodes is ongoing in several groups and expected to yield more results in the upcoming future.

4.3. New Semiconductor Materials for PBTs

As reported above, for the n-type OPBTs, which are based on C_{60}, the current density is neither limited by the density of pinholes nor the charge carrier injection at the emitter but rather by the
charge carrier transport in the bulk of the semiconductor, which manifests itself in a SCLC (mobility of only 0.06 cm$^2$/V·s) in C$_{60}$. Hence, to boost the performance of OPBTs, alternative semiconductor materials that offer higher mobility in vertical direction need to be investigated. However, mobility is only one criterion for the semiconductor material. Additionally, the material should be dopable to minimize the injection resistance at the semiconductor–metal interface, and its surface properties should allow for the formation of pinholes in the base metal to which this material is required to form a Schottky-type contact.

To the best of our knowledge, organic semiconductors with electron mobility in the vertical direction better than C$_{60}$ do not exist at present. However, an alternative class of materials that are also partially compatible with low-temperature processing on flexible substrates are electron-transporting transparent conductive oxides such as ZnO, indium gallium zinc oxide (IGZO), In$_2$O$_3$, etc. In particular, IGZO has been used to demonstrate flexible Schottky-type diodes with a record transition frequency of 2.45 GHz at a voltage of <2.0 V.

**Figure 7.** a) Schematic structure of an OPDBT measured in common-emitter configuration. With applied base1–emitter voltages ($V_{B1}$) and base2–emitter voltages ($V_{B2}$), electrons can pass through the bases and hence reach the collector (a). Except for a minor leakage current into the bases $I_{B1}$ and $I_{B2}$ (blue dotted arrows), the emitter current $I_E$ (solid blue arrow) can finally arrive at the collector, forming the collector current $I_C$. b) Transfer curves as a function of $V_{B2}$ at different $V_{B1}$ of 0, 0.5, 1.0, 1.5, and 2.0 V, respectively. Inset: threshold voltage shifts. The threshold voltages are dependent on the base 1 bias during base 2 sweep, error bars indicate the slight device-to-device variations measured over 61 devices. c–e) Circuit diagrams and static voltage transfer characteristics of an inverter (c), a NAND gate logic circuit (d), and an AND gate logic circuit (e). Reproduced under the terms of the CC-BY-4.0 license. Copyright 2020, The Authors, published by Springer Nature.
different metals as well as treatment processes. For example, oxides such as ZnO or others have been shown to form excellent Schottky-type contacts with noble metals such as Au, Pd, or Pt. However, even the use of less noble metals like Ni, Cr, or Ti for the base electrode might be possible depending on treatment conditions and specific interface interactions. Overall, oxide-based semiconductors can be an excellent choice to further improve the performance of n-type OPBTs. However, the processes allowing for the formation of semiconductor-filled pinholes in the base layer still need to be explored.

Due to their comparably low mobility, hole-conducting oxide-based semiconductors are not an alternative for p-type OPBTs. Currently, the p-type OPBTs are based on pentacene. However, since pentacene has a high roughness and an undesirable molecular orientation, pentacene-based OPBT fell short when comparing the performance to C60-based devices. Furthermore, although the charge carrier injection can be improved using self-assembled monolayers or molecular doping, pentacene does not offer great potential for further device improvements due to the limited mobility in the vertical direction. Other materials such as 2,7-dioctyl[1]benzothieno[3,2-b][1]benzothiophene might offer higher charge carrier mobility. However, due to the preferred molecular orientation, the mobility is highly anisotropic with much lower values in the vertical direction. Thus, since the material development for p-type organic transistors is focused on lateral TFTs, there is a lack of appropriate materials for p-type OPBTs. An alternative that just recently emerged is rubrene grown as triclinic thin-film crystals. In this crystal phase, record mobility values in the vertical direction of up to 10 cm² V⁻¹ s⁻¹ have been reported as well as the possibility to dope the crystals, thereby improving charge carrier injection.

4.4. Perspective for High-Frequency Operation

Thus, looking at the recent improvements in the OPBT processing, e.g., the anodization technique allowing for thick base electrode layers and an adjustable thickness of the oxide-layer surrounding the base, it is interesting to investigate where the inherent performance limits of OPBTs will be. In the following text, we derive important scaling laws of OPBTs to predict the transition frequency for various device parameters.

The assumptions on which our calculations are summarized in the Section S4.4 in the Supporting Information. In brief, we adapt the design of the OPBTs operating at 40 MHz, and describe the collector current by a space-charge limited current. Effects of self-heating are not taken into account in our model. The results of our calculation are summarized in Figure 8. In the simulation, the semiconductor thickness refers to both top and bottom semiconductor layers, which are considered equal.

We discuss three important dependencies: a) base layer accumulation capacitance (given by the oxide thickness) versus semiconductor thickness, b) base electrode thickness versus semiconductor thickness, and c) charge carrier mobility versus semiconductor thickness (parameters of the simulation are given as insets). These parameters have the largest impact on $f_T$. Furthermore, they can be modified by the fabrication method, i.e., using anodization, the base layer and the thickness of the AIO$_x$ surrounding the base can be as thick as 50 and 10 nm, respectively, without reducing the permeability of the base layer for electrons passing from emitter to collector. We draw these three dependencies for different transition frequencies of 40 MHz, 100 MHz, and 1 GHz. As a reference, we also show the device operating at 40 MHz. In all three plots in Figure 8, this device performs slightly better than predicted by our simulations, which probably originates from the effect of self-heating. In Figure 8a, the base layer accumulation capacitance (given by the oxide thickness) is plotted versus semiconductor thickness. Most interestingly, if the base layer capacitance is reduced (e.g., by anodization at higher voltage), then the semiconductor layer thickness can be chosen larger for a fixed transition frequency. Having a thicker semiconductor layer has two substantial advantages: 1) it might help to improve the device yield in OPBTs due to fewer shorts, and 2)
the overall power dissipation is lower due to less collector current and less displacement current at the base. Following this graph, $f_T$ of 1 GHz is possible for a layer thickness <40 nm, which in practice though results in short-circuits. In Figure 8b, base electrode thickness is plotted versus semiconductor thickness. Here, a similar trend can be seen as in Figure 8a. An increased thickness of the base electrode (i.e., using the anodization) reduces Ohmic losses and hence, the given $f_T$ can be reached for a larger thickness of the semiconductor film. However, in this case, a transition frequency of 1 GHz is also practically impossible since it would require semiconductor layers thinner than 40 nm. In Figure 8c, the mobility is plotted versus the semiconductor layer thickness. For higher mobility, the given $f_T$ is reached even for thicker semiconductor layers. Most interestingly, already for a mobility of $\approx 0.4 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$, the layer thickness for the 1 GHz line would be above 50 nm, rendering such devices physically possible (ideal would be a mobility of 2 cm$^2$V$^{-1}$s$^{-1}$, allowing for a thickness of 100 nm). Furthermore, this is even a conservative estimate since a base thickness of >15 nm and an oxide capacitance of <1 $\mu$F cm$^{-2}$ will further lower the required mobility values. Thus, 1 GHz operation is in reach for OPBTs if semiconductor materials with sufficiently high charge carrier mobility can be successfully implemented in OPBTs. This strong dependency of $f_T$ versus the semiconductor thickness purely originates from the SCLC which allows OPBTs to outperform lateral OTFTs.

Besides this promising perspective, it should be noted that such high-frequency operation cannot be achieved without increasing power dissipation. In particular, for $f_T$ of 1 GHz at a mobility of 0.4 cm$^2$V$^{-1}$s$^{-1}$, we calculate a power density of $\approx 6 \text{ kW cm}^{-2}$ which is too high for low thermal conductivity substrates such as flexible foils. Thus, a direct current continuous-mode operation at high alternating current signal frequencies at the base electrode will be difficult for OPBTs on flexible substrates. For applications where the transistors are driven in a pulsed regime though, e.g., the switching transistor in an active-matrix display, it might be possible. However, the excellent features of OPBTs, such as 40 MHz, 1 kA cm$^{-2}$, and excellent stability, are not obtained simultaneously because the transition frequency of 40 MHz is measured in a pulse-biasing mode. OPBTs also show some additional nonideal features, e.g., relative leakage current (nanoampere regime) than lateral-channel OTFTs (below picampere regime), which leave plenty of room for future improvements of these vertical transistors.

5. Conclusion

Due to steady improvements, OPBTs are today the most promising alternative organic transistors with the potential to operate even at 1 GHz. However, to further advance with the OPBT technology toward complete electronic circuits, it is vital to control the mechanisms behind the pinhole formation within the base layer in order to develop robust and scalable fabrication techniques. The yield analysis reported in this paper proves that even with the simple oxidation process in air and subsequent thermal annealing, a device yield of >90% can be achieved. However, such device yield of OPBTs would not be high enough for future production, which still needs to be further optimized. At the same time, it is still unclear, why some devices (hero devices) perform much above the average. Further investigations are required to push the average closer to the performance we see in hero devices.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

Research data are not shared.

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