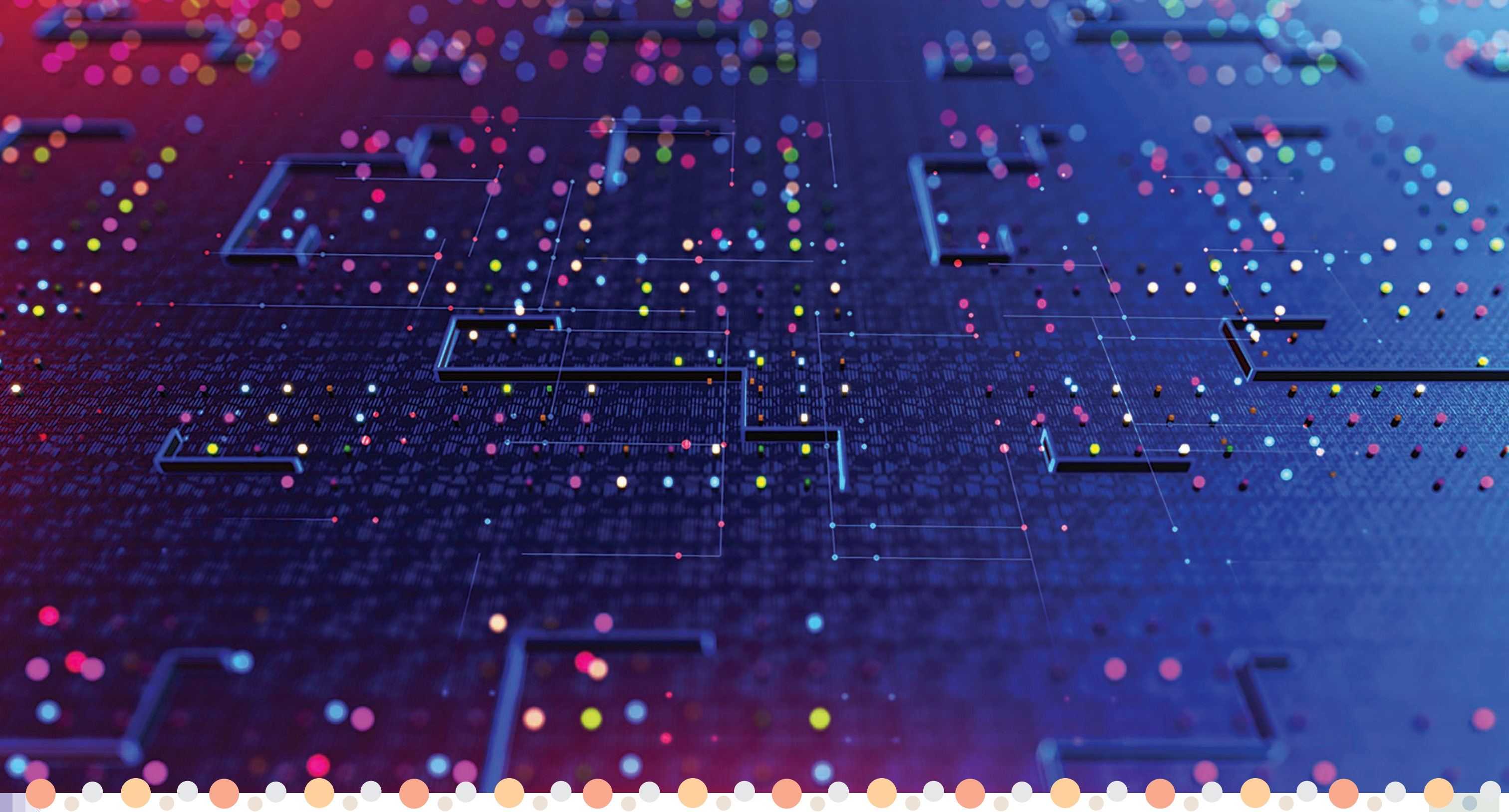
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Route Toward Sustainable Oxide TFTs

Does Thin Film Oxide Technology Offer a Pathway Toward More Sustainable Electronics?

By Niko Münzenrieder, Luisa Petti, Giovanni A. Salvatore, and Giuseppe Cantarella

Wearable, flexible, lightweight, and large-area electronics, able to adapt to arbitrary shapes, is a recent technology that promises to integrate sensing and data processing capabilities into everyday objects. This will allow connecting the digital and the physical worlds we live in and enable new innovative applications such as intuitive interfaces and smart textiles for continuous health and safety monitoring. Different technologies, including devices based on organic semiconductors and conventional silicon, are used to develop such wearable systems, however, devices made from novel oxide-based semiconductors currently have one of the best changes to result in electronic systems with sufficient electrical performance on flexible and large-area substrates. Yet, these exciting possibilities come at a price. If all the objects and surfaces around us are equipped with electronic sensor systems, these objects themselves transform into electronic devices, which in turn would massively increase the amount of electronic waste at the end of their lifetime. The same is true for plastic waste as currently most unobtrusive electronics are fabricated on polymer substrates. Furthermore, electronics that are operated near the human body must be safe and biocompatible. The perfect solution for these problems would be to fabricate electronics using environmentally friendly materials and processes, and to guarantee that the electronic components, after fulfilling their intended task, dissolve into their basic constituents, which are then entirely passive or safely absorbed into the nutrient cycle of our environment. Such transient and bioresorbable behavior has been demonstrated for several types of devices but is an underdeveloped approach in the field of oxide semiconductors. Here, motivations and challenges to combine the excellent electrical and mechanical

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properties of flexible oxide electronics with the environmental friendliness of bioresorbable electronics are explored. This is done by using the most important active building block of such electronics, namely, the TFT, as a model device (Figure 1). Besides assessing the various definitions of biocompatibility and environmental friendliness used in today's research, the available classes of suitable materials and their impact in the electrical and environmental domain are described. Finally, an evaluation of how such materials can be integrated into working oxide transistors and which performance levels are achievable by these first examples of sustainable oxide TFTs is presented.

Introduction

Electronic devices are evolving from only being integrated into dedicated rigid devices used for work, entertainment, or communication, and are on their way to becoming more unobtrusively integrated into our environment. This is possible thanks to the use of flexible electronics, enabling deformable electronic devices to be made from thin-film structures and directly fabricated on large-area substrates such as plastic foils. Rather than considering it a replacement for conventional silicon-based technology, flexible TFTs, circuits, and systems represent an alternative technological platform able to tackle innovative market sectors, such as smart textiles, wearables, and the Internet of Things. This is because flexible systems, while unable to match the electronic performance of traditional electronics, offer distinct advantages when it comes to their mechanical properties, costs, ability to integrate different materials and device technologies into a monolithic system, and the possibility they offer to realize human-sized interfaces and systems. Such capabilities enable the integration of imperceptible sensing, processing, and communication functionalities into everyday objects and even into the human body itself, which in turn allows for the first intimate connection of the digital and physical worlds. The resulting opportunities and potential benefits of items such as sensor patches attached to human skin, electronic textiles for smart clothes, or conformable interfaces for interactive surfaces are nearly limitless. Continuous physiological information from patients, smart assistance for athletes and professionals, data for efficient production processes, or the intuitive manipulation of virtual objects are just some of the possibilities. These benefits, however, come at a cost, namely, the fact that if nearly every object in our environment gets equipped with electronic functionality, the associated use of nonrecyclable materials such as plastic foil substrates and toxic and rare-earth elements could potentially have a dramatic impact on the environment once the end of life of these items is reached. This, in combination with the potentially very short lifetime of certain types of wearables, has the potential to significantly exacerbate the problem of so-called electronic waste (e-waste). E-waste, as visualized in the left part of Figure 2, is hardly recyclable, contains pollutants, and represents a potential risk for human health and the environment [1]. Additionally, it

FIGURE 1. Ultrathin oxide TFT on a natural plant leaf substrate.

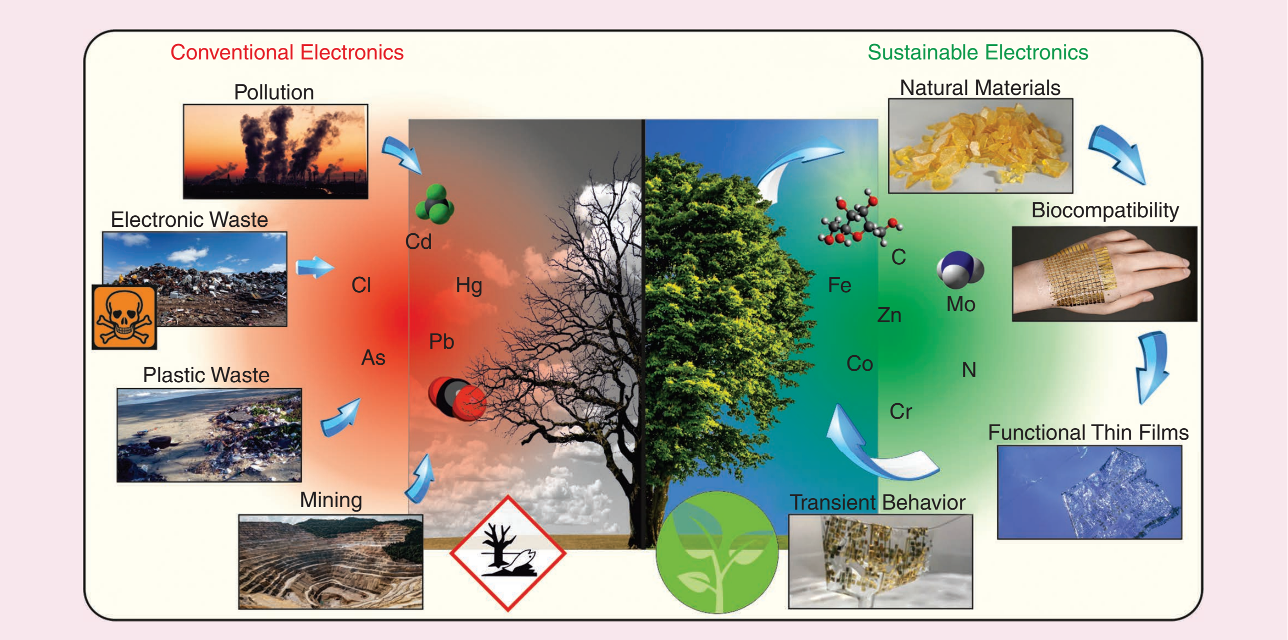

FIGURE 2. The transition from conventional electronics to sustainable devices and systems made from natural materials, and environmentally friendly methods will enable circular processes, waste-free products, and safe electronics operating in proximity to or even inside the human body. Images reproduced from [7] ©2013 IEEE (biocompatibility), and [8] (transient behavior).

consumes valuable resources which, on their own, can have negative environmental and social effects while being mined and processed. The last reports have pictured a dramatic situation, with 53.6 million tons (Mt) of e-waste generated in 2019, an exceptional amount that is also growing by 2 Mt/year [2]. Furthermore, the potentially negative effects of traditional electronic materials become more relevant by the fact that wearable systems are designed to be in close contact with the human body, where harmful substances can have especially severe consequences.

Although traditional recycling and reuse approaches can partially mitigate the effects of e-waste, the sheer amount of envisioned wearable devices and their intimate contact with biological systems and food sources calls for more extreme solutions. This solution comes in the form of transient and bioresorbable electronics, which are made from biocompatible materials, degrade after their use, and are then reintroduced into an industrial recycling process or metabolized by nature. Such transient behavior can come at different levels and is well suited for the world of thin-film electronics as the amount of material needed to create the functional layers is minimal. The most straightforward approach consists of the dissolution of only the electronics devices, while reusing the substrate. Here, the employment of biocompatible and dissolvable materials, such as magnesium, allows rapid dissolution in easily accessible liquids, including water. Alternatively, the use of degradable and nature-inspired substrates complicates the electronics fabrication process but can support the implementation of fully dissolvable systems. If in this case the raw materials are also sourced in a sustainable way, fully circular electronics without negative impacts on humans or the environment can be realized. Such behavior has already been demonstrated by silicon and organic semiconductor-based technologies, which even resulted in edible electronics [3].

In this article, the possibility of realizing sustainable and bioresorbable electronics using oxide semiconductors is explored. Oxide semiconductors are an innovative class of materials that have rapidly gained popularity from their discoveries in the 1960s [4], [5], and especially after room-temperature deposition and large-area processability was demonstrated in the early 2000s [6]. The advantage of oxide semiconductors is their unique combination of electronic performance, mechanical flexibility, and processability on large-area temperature-sensitive substrates. This rapid success has allowed the implementation of these semiconductors into TFTs, circuits, and complex systems, which have already reached the market. At the same time, there are still only a few examples of sustainable oxide devices. Nevertheless, if combined with suitable materials and fabrication processes, these innovative semiconductors have the potential to enrich new research areas and pave the way to a green transition (see the right part of Figure 2).

THE ADVANTAGE OF OXIDE SEMICONDUCTORS IS THEIR UNIQUE COMBINATION OF ELECTRONIC PERFORMANCE, MECHANICAL FLEXIBILITY, AND PROCESSABILITY ON LARGE-AREA TEMPERATURE-SENSITIVE SUBSTRATES.

Transient, Biocompatible, or Bioresorbable?

Realizing sustainable electronics requires a good understanding and definition of what should be achieved. Currently, this is not always clear, as various levels of sustainability have been proposed, and the associated terms like “degradable,” “biodegradable,” and “compostable” can have varying interpretations, highlighting the evolving understanding of how biomaterials interact with living organisms. For instance, a material is deemed “biocompatible” if it does not induce adverse effects when in contact with a living organism. However, due to the intricate nature of immune responses and tissue repair mechanisms in living organisms and interactions between multiple materials, it is insufficient to assess the biocompatibility of a single material solely in relation to a specific type of tissue. Before proceeding, it is therefore important to clarify the meaning of some terminology and, if available, the standard tests usually performed. The most relevant classifications are also illustrated in Figure 3:

- *Degradable/transient*: This behavior refers to the process of breaking down materials through physical actions and chemical agents, without any specification on the transformation process or the toxicity of the resulting products.
- *Compostable*: This usually refers to industrial tests occurring at controlled temperature conditions (i.e., 50–58 °C) and the fragmentation and loss of visibility of an object in the final compost. It is measured using a pilot-scale composting test (EN 14045). A material qualifies as compostable if, after a three-month period, the fraction of residues with dimensions exceeding 2 mm constitutes less than 10% of the initial mass. To be compostable, a material should also comply with additional requirements like having no adverse effects on the composting process, while salt, volatile solids, nitrogen, phosphorus, magnesium, and potassium concentration stay within the established limits.
- *Biocompatible*: This refers to the ability of a material or a medical device to interact with the biological systems of a living organism without causing harm, adverse reactions, or immune responses. Biocompatibility is a complex and multifaceted concept, and the specific requirements can vary depending on the intended use of the material or device. The term is extensively used for medical devices. ISO 10993 provides guidelines and procedures for the biological

evaluation of medical devices. Ecotoxicity testing, instead assesses the potential environmental impact of materials and devices. They involve evaluating how a material or chemical may affect ecosystems, including aquatic and terrestrial organisms, and the environment. Ecotoxicity tests help determine the potential risks associated with the release of substances into the environment. Typical tests aim at assessing water (ISO 6341, ISO 7346, and ISO 10706) and soil (ISO 16734) quality. However, both definitions do not concern the decomposition of a material or device at the end of its lifecycle.

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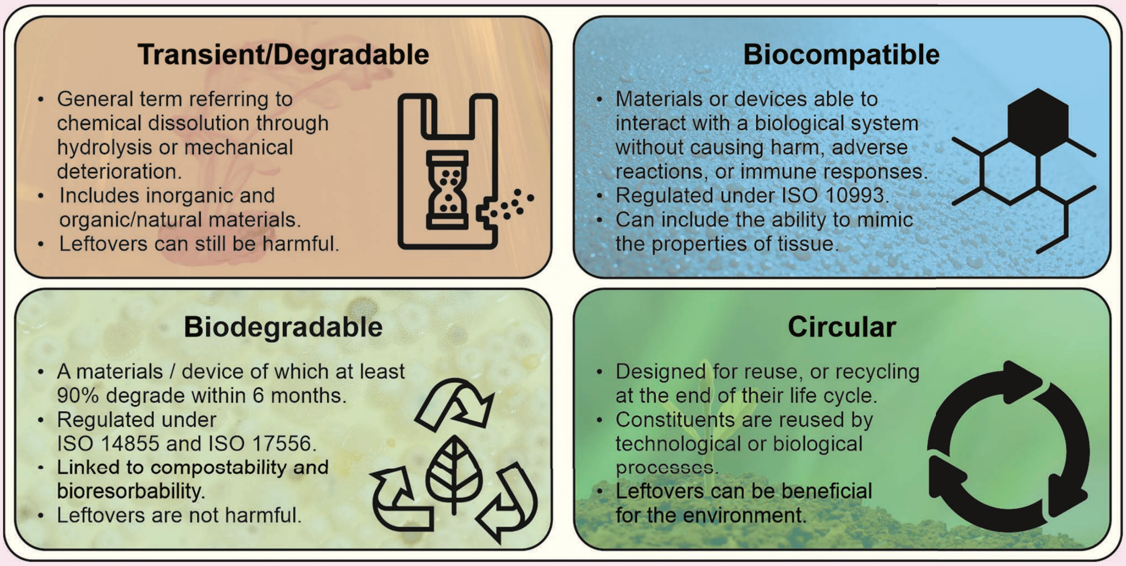
A MATERIAL IS CONSIDERED BIODEGRADABLE IF AT LEAST 90% OF ITS BASIC COMPONENTS DEGRADE WITHIN SIX MONTHS.

- *Biodegradable*: A material is considered biodegradable if at least 90% of its basic components degrade within six months. Biodegradation assesses the actual metabolic conversion of compostable material into carbon dioxide (ISO 14855). Biodegradation is the process of decomposing organic compounds, facilitated by microorganisms under two distinct conditions: either aerobic biodegradation under the presence of oxygen, or anaerobic biodegradation under the absence of oxygen. In both cases, the process allows organic substances to break down into carbon dioxide, water, mineral salts, and new biomass. The biodegradability of materials is a relative property that varies depending on environmental conditions and the periods in which it occurs. Therefore, various laboratory standards are established based on different contexts, including biodegradability in soil (ISO 17556), biodegradability in water (ASTM D6691, ASTM D6692, and ASTM D5209), or biodegradability in waste treatment facilities (ISO 14851-14852).
- *Bioresorbable*: Although the term *biodegradable* usually refers to materials that break down through natural environmental processes, bioresorbable materials are absorbed by living organisms, including humans, animals, or plants. A bioresorbable material is, therefore, specifically designed to be processed by a system's biological processes. There is no dedicated International Organization for Standardization standard for resorbable materials and devices. ISO 10993 is considered the reference as it covers various related aspects of biocompatibility testing.
- *Circular*: This is a term associated with devices that are intended for reuse, refurbishment, or recycling at the end of their lifecycle. The design may prioritize longevity, ease of disassembly, and recyclability. Currently, the term refers to materials that can be recycled through industrial processes with typical examples including metals or glass. At the same time, the circular process must also consider the original source of the used materials and, e.g., the influence of mining operations on the environment. The concept, however, can also be extended to transient devices that decompose into bioresorbable constitution, which, in turn, have a positive effect on the biological system by which they are assimilated. Such biological circularity could, for example, be realized by electronic



Transient/Degradable

- General term referring to chemical dissolution through hydrolysis or mechanical deterioration.
- Includes inorganic and organic/natural materials.
- Leftovers can still be harmful.

Biocompatible

- Materials or devices able to interact with a biological system without causing harm, adverse reactions, or immune responses.
- Regulated under ISO 10993.
- Can include the ability to mimic the properties of tissue.

Biodegradable

- A materials / device of which at least 90% degrade within 6 months.
- Regulated under ISO 14855 and ISO 17556.
- Linked to compostability and bioresorbability.
- Leftovers are not harmful.

Circular

- Designed for reuse, or recycling at the end of their life cycle.
- Constituents are reused by technological or biological processes.
- Leftovers can be beneficial for the environment.

FIGURE 3. Currently used keywords that describe sustainable electronics represent significantly distinct levels of environmental friendliness. In the future, it will even be necessary to go beyond these classical terms and create devices with a positive environmental impact.

devices made from beneficial trace elements so that these can effectively act as a food supplement or fertilizer after being naturally degraded.

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FREQUENTLY OVERLOOKED, THE ELECTRICALLY PASSIVE SUBSTRATE MIGHT BE ONE OF THE MOST IMPORTANT COMPONENTS OF SUSTAINABLE TFTs.

Materials and Processing

The biocompatibility and environmental friendliness of sustainable thin-film devices strongly depends on the properties of the used materials, including their transient and bioresorbable properties, the source of the materials, amount of the materials used, and the resources needed to process them. This goes in parallel with the electrical and mechanical properties, which, to a larger extent, can also be influenced by the device geometry and how exactly the different materials are combined. The creation of bioresorbable oxide thin-film electronics by simultaneously optimizing this plethora of different properties requires expertise from traditional complementary fields, such as electrical and mechanical engineering, physics, materials science, and chemistry, but also biology, process engineering, and environmental sciences. Fortunately, it is possible to benefit from some inherent properties of oxide thin-film technology, as well as from some substantial work available in conventional flexible and large-area electronics, such as the following:

- Thin-film devices are made from only tens of nanometers-thick layers. One gram of titanium, for example, is enough to coat more than 20 m^2 with a 10-nm-thick layer. Hence, the amount of raw material required is very small.
- Only the not necessarily thin layers, namely, the substrate and the encapsulant, can be made from natural or not highly processed, and hence resource-efficient materials. This is possible because in contrast to the very resource-intensive single-crystal semiconductor wafers used for standard electronics, the substrate of thin-film devices is often solely a mechanical support with no active electrical properties.
- Finally, the growing interest in flexible electronics has already led to processes that allow the fabrication of devices on temperature-sensitive, deformable, and comparably rough supports, mostly plastic foils. These developments, in turn, can act as a starting point to realize devices on degradable and natural substrates with similar mechanical properties.
- Finally, in the field of flexible electronics, it is comparably common to integrate new materials, such as sensing layers or soft organic composites, into the fabrication process. This is not easily possible in standard silicon-based processes, as there, the requirements concerning electrical performance, cleanliness, and process integration are much stricter. This flexibility concerning alternative materials also simplifies the evaluation and eventual use of environmentally friendly substitutes.

Nevertheless, simply using established processes and replacing the currently used materials with bioresorbable ones is generally not successful. The reasons for this are manifold: the materials used for a system have to processable on their own but also all together, e.g., once a material with a maximum processing temperature is added to the device stack, this maximum temperature must not be overcome in all subsequent processing steps. Similarly, it is also possible that materials that work fine on their own can cause issues when combined with others. For example, it must be excluded that one material diffuses into another layer and negatively affects its properties, or that interface effects between certain pairs of thin films prevent the proper functionality of TFTs by causing, e.g., an excessive number of interface trap states. Also, individual materials can change their behavior depending on the context. For example, strain-sensitive piezoelectric materials might exhibit desirable properties in the context of rigid conventional electronics but can be less suitable if used in wearable systems fabricated on flexible substrates. Additionally, it must be ensured that the novel bioresorbable devices are able to operate for the intended period and only degrade afterward. This can be done passively by tuning the layer thickness, preferably utilizing the substrate and encapsulation because these do not affect the electrical performance of the devices. Alternatively, active triggering can be realized by using, e.g., heat, light, or electrical stimuli to initiate the dissolution process. This, however, increases design complexity even more [9], [10]. Finally, it is simply challenging to replace materials that are established in the semiconductor industry with more sustainable alternatives without compromising the electrical performance of the resulting devices. In fact, it should not be forgotten that even the most bioresorbable device is only useful if it also provides sufficient electrical performance.

In the context of wearable electronics, in particular unobtrusive sensor systems, the main tasks of TFTs are to act as switches in active-matrix arrays or to be used as building blocks in analog amplifiers and other sensor conditioning circuits. Oxide TFTs are field-effect devices, as shown in Figure 4(a), and their electrical performance must be optimized toward their specific intended application, but generally should exhibit a large current-driving capability combined with a low off current (and hence a large ON–OFF current ratio), low gate-leakage current, low output conductance (i.e., the drain current should saturate and be independent of the drain-source voltage), an as-high-as-possible operation frequency, and be functional at as-low-as-possible voltages. To create such oxide TFTs, at least four distinct types of materials that are classified for their functionality in the devices' design are needed:

1) *Substrate/encapsulation*: Frequently overlooked, the electrically passive substrate might be one of the most important components of sustainable

TFTs. With a thickness normally in the micrometer or millimeter range, the substrate is responsible for most of the mass and volume of the system. It provides mechanical support and determines the mechanical, optical, and haptic properties of the system; often triggers decomposition; and controls the operation timescale of bioresorbable devices. Even in the rare cases in which substrate free electronics are realized, temporary support during fabrication is needed [11], [12].

2) *Conductors*: Conductors transport charge carriers and inject them into the semiconductor layer, but they are also used to create interconnections and contact pads to interface devices to other components. Besides their conductivity, an important parameter is the work function, which must match the requirements of the semiconductor used. Additionally, the

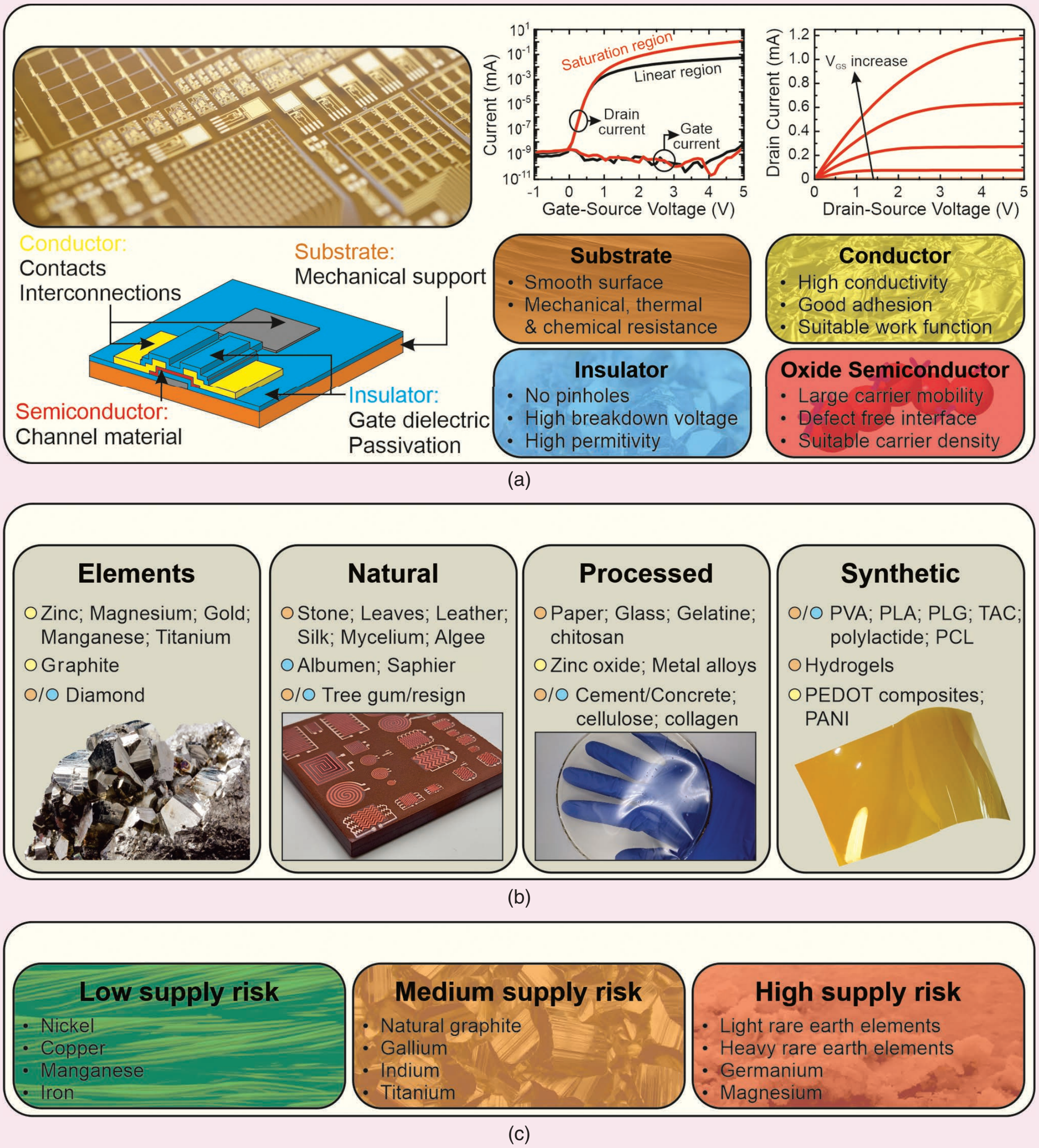

FIGURE 4. Materials for TFTs. (a) A representative image and the electrical characteristics of a TFT with a channel width/length ratio of 280 μm/10 μm, and the simplified device structure of a TFT, illustrating the broad variety of required materials, where each component needs its own set of electronic, mechanical, and chemical properties. (b) The available classes of materials not often associated with electronics, and their potential to enable environmentally friendly oxide transistors with good electrical performance. (c) Availability of the selected relevant raw materials. PEDOT: poly(3,4-ethylenedioxythiophene); PANI: polyaniline; PLG: poly(lactic-co-glycolic acid); TAC: cellulose triacetate; PCL: polycaprolactone.

adhesion on the various substrates and surfaces must be considered. This often requires dedicated adhesion layers or additional treatments.

3) *Insulators*: Insulators are primarily needed to electrically insulate the gate contact of a TFT. These layers are normally demanding to realize as it can be challenging to create pinhole-free, thin, insulating layers on unconventional substrates. Besides an extremely low specific conductivity, the material should exhibit a high dielectric constant, high breakdown voltage, and form a defect-free interface with the semiconductor. At the same time, insulating layers are also used as a passivation layer to improve electrical device performance and environmental stability [13]. Additionally, certain fabrication processes also ask for extra insulating layers acting as, e.g., the etch-stop layer [14].
4) *Semiconductors*: These are normally the core of any TFT, which needs optimized carrier density and as-high-as-possible carrier mobility. Although it is difficult to deposit high-performance semiconductors on sustainable substrate, oxide semiconductors, some of which can even perform well in their amorphous form, represent a good tradeoff between electrical and nonelectrical properties. Some common materials are InGaZnO (IGZO) [6], indium-tin-oxide [15], nikel-oxide [16], and zinc-oxide [17]. Of these, ZnO in particular offers very good biocompatibility and sustainability. An additional advantage of oxide semiconductors is that these are normally transparent for visible light. A disadvantage is that oxide semiconductors are nearly always n-type semiconductors [18].

There are multiple approaches to create or to identify environmentally friendly bioresorbable materials with the required electrical properties. Figure 4(b) illustrates that pure elements, fully natural composites, processed natural materials, and fully synthetic materials can be used. All these options come with their own advantages and disadvantages:

- *Pure elements*: As the basic constituents of matter, pure elements could be considered the most natural materials. In particular, numerous metals, e.g., Zn, magnesium, and tungsten, but also carbon-based materials, can be used as conductive layers in bioresorbable TFTs [19]. This is because certain metals such as gold or titanium do not affect biological processes, while others, such as Zn and magnesium, can even be metabolized. However, pure elements are not necessarily bioresorbable as various elements such as lead or arsenic can oxidize and decompose over time but are also poisonous for the ecosystem. Furthermore, oxide semiconductors cannot be pure elements, and there are no corresponding solid-state insulators.
- *Natural materials*: Fully natural materials directly harvested from the environment are traditionally not associated with any form of semiconductor technology. This is because such materials are normally not considered clean or pure enough. Such materials can be inorganic, such as natural minerals and stones, which can be well suited as substrates; plant-based materials; e.g., leaves, wood, natural resigns, and gums; or char, which can function as bioresorbable substrate, insulators, or conductors [20], [21], [22]. Finally, animal-based products, including silk, gelatine, and egg albumen, are also used to create environmentally friendly and bioresorbable substrates and insulators for transient electronics [23], [24]. In any case, it must be considered that natural materials can also be poisonous and hence harmful.
- *Processed natural materials*: Diverse types of materials are fully based on natural resources, but nevertheless do not, or only rarely, exist in nature. This can be because their natural concentrations are too low, and they are not stable in the environment (such as most pure metals). Additionally, it is often more scalable and sustainable to re-create natural composites with optimized properties and dimensions on an industrial scale, for example, artificial versus natural concrete, or paper versus wasp nests, both of which can be used as substrate or dielectric [20], [25]. Other examples of such materials are leather as substrate, and purified chlorophyl and other organic pigments, which can even exhibit bespoke electronic properties [26]. Furthermore, degradable plastics based on polymers from plants such as starch, cellulose, biomass, or alginate, and animal-derived materials including chitosan, collagen, urea, thioester, or silk, developed to reduce plastic waste, are suitable materials [21], [[27], [28], [29].
- *Synthetic materials*: Artificial materials are often considered not very biocompatible, however, lately, there has been a lot of innovation, which can be utilized for the fabrication of bioresorbable electronics. Synthetic polymers, which can either undergo a full chemical breakdown or be recycled into their basic monomeric and oligomeric building blocks, are suitable degradable material, which can mostly function as substrate or insulators. Examples are poly(L-lactide), polylactide, polyglycolide, polylactide, or polyvinyl alcohol (PVA) [27], [30]. Additionally biocompatible hydrogels, and vitrimers show immense potential to be integrated into thin-film fabrication processes [31], [32].

The large variety of potential bioresorbable materials for the fabrication of TFTs is encouraging. However, the mere fact that suitable or promising materials for the realization of sustainable oxide devices exist is not sufficient. Three other dimensions must also be considered:

1) *Availability and economics*: On the one hand, source materials can simply be hard to obtain or rare, and hence expensive. This is not necessarily a major issue as the minuscule amounts are used in thin-film technology, and their performance can offset the economic cost, e.g., in the case of gold. However, over the last decades, the concern of guaranteeing

a constant supply of raw materials to the electronics industry has raised increased attention. Although organic and renewable materials, or resources abundant in nature (e.g., egg albumen [23]), are normally easily available, key elements for oxide semiconductors, such as indium and gallium, come with larger supply-chain risks. For example, 48% and 80% of the global demand for indium and gallium, respectively, are covered by a single country, and also metals, such as magnesium and titanium, widely employed as transient and biocompatible conductors, have a limited supply base [33]. These risks are partially quantified in Figure 4(c), utilizing the recent list of critical raw materials, which was issued by the European Union to rate the importance and availability of certain materials [34].

2) *Social and environmental impact*: These hard-to-quantify effects concern the destruction of the landscape, creation of toxic waste, energy consumption linked to the mining, refinement, the purification of raw and recycled materials, unsatisfactory working conditions of the involved workforces, child labor or the risk of utilizing resources from conflict regions, and negative influences on the ecosystems, e.g., by the unsustainable use of animal products [35].
3) *Processability*: This concerns the use of unconventional raw materials, their thin-film deposition and structuring, and the creation of functional oxide transistors with sufficient electrical performance on the device level. The processability of materials varies significantly. Metals, on the one hand, are well established and can be processed in many ways. If natural or organic materials, which are often humid, outgassing, sensitive to temperature, or attacked by solvents, are employed, the device fabrication process has to be carefully optimized. The advantage of oxide semiconductors is that they can be processed at room temperature using both vacuum and solution processing [18]. Vacuum-based devices often structured using lithography exhibit excellent performance, but the process is very demanding. It normally involves high vacuum, elevated temperatures, and solvents. This is often not possible if natural materials are involved. Additionally, their often-rough surfaces can complicate processing. Solution processing, such as printing, on the other hand, does not require vacuuming, and the structuring is done together with the deposition itself. However, it can still require elevated temperatures, the feature sizes are limited, and formulation of the printing inks involves advanced chemistry, solvents, and significant optimization time. A less established, and currently hardly scalable, option, which combines the advantages of printing and vacuum processing, is the transfer technique. Here, the devices, or parts of a device, are fabricated on a robust carrier, and then released and placed on a biocompatible substrate [36]. This way, incompatible materials and processes can be separated until the last step of the fabrication process. In all cases, it is worth mentioning that the sustainability of the used process chemicals must also be considered. This issue often receives less attention, but there are some promising examples, such as the replacement of Heptane by coconut oil [37].

Devices

To attain sustainability in electronics, a comprehensive strategy is imperative, encompassing three key avenues: 1) the development of novel sustainable materials, 2) streamlined manufacturing processes, and 3) effective recycling methods. This converges into the overarching concept of “safe and sustainable by design,” wherein safety, environmental, social, and economic sustainability requisites are integrated into the specifications. Nevertheless, the Life Cycle Assessment (LCA) of electronic devices and circuits is intricate. This is even more true when it comes to thin-film technologies that are relatively new. The lack of information on and the large variation of processes and materials hinders the development and implementation of a holistic approach. In the realm of thin film devices, recent LCA studies have focused on batteries [38], photovoltaics [39], organic light-emitting diodes [40], and solar panels [41]. LCA for TFT technology, whether based on organic materials or oxides, has not yet been performed to the same extent.

As highlighted in the previous paragraphs, oxide semiconductors and their associated sustainable materials have nevertheless recently emerged as viable solutions for sustainable electronics. The tremendous advancements oxide TFTs have undergone in the last decades rely mostly on nonsustainable materials and processes. Up to now, considering the impact that the substrate has on overall device sustainability, the main efforts have been devoted to the realization of oxide TFTs on alternative substrates that are able to show (bio)-degradable, compostable, biocompatible, or bioresorbable properties. Examples of such works include paper substrates, such as the recently reported bilayer IGZO/Al_2O_3 TFTs realized without the use of photolithography on transparent cellulose nanopaper and demonstrating the channel mobilities of 22 cm^2/Vs [42]. Further examples are IGZO TFTs, fabricated again without the use of photolithography on seaweed-derived sodium alginate substrates, which yield good electrical performance including a mobility of 26.8 cm^2/Vs [28]. Still, the biocompatibility and biodegradability of the used sodium alginate substrate was only evaluated elsewhere [43].

Besides substrates, further efforts have also been devoted to the use of sustainable conductors and insulators for metal oxide TFTs. As regards conductors, there are palettes of well-known degradable, biodegradable, compostable, biocompatible, or bioresorbable metals, including magnesium, Zn, molybdenum, and tungsten [44]. These metals, especially molybdenum, which is biodegradable, are well reported for metal oxide TFTs and circuits [18].

Concerning dielectric materials, a well-known and first-of-its-kind example is the use of chicken albumen insulators extracted from chicken egg white as gate dielectric for the realization of

IGZO TFTs on paper substrates, yielding channel mobilities of 6.48 cm^2/Vs [23]. Biodegradability of the chicken albumen insulator has been tested elsewhere [21]. Following this work, more examples have been proposed, including chitosan biopolymers as electrolyte dielectrics in combination with indium tin oxide (ITO) TFTs, showing an ON–OFF ratio of 10^5 at 2-V operation voltages [45]. Biodegradability of chitosan has been well tested in soil [46]. Also worth mentioning are eco-friendly solution processing routes of gate dielectrics, such as the exploration of water-induced aluminum oxide dielectrics for hybrid metal oxide/polymer In_2O_3: polyvinylpyrrolidone (PVP) TFTs yielding channel mobilities of 14.1 cm^2/Vs [47]. Similarly, aqueous and green routes to realize Ga_2O_3 dielectrics for In_2O_3 TFTs were successfully demonstrated [48].

At the same time, green approaches were also employed to produce printable metal oxide semiconductors [49], [50]. Concerning metal oxide semiconductors, ZnO is surely one of the most investigated, considering not only its good electrical properties and simple fabrication via solution processing but also its dissolution kinetics in, e.g., liquid electrolytes with an ionic strength similar to those of physiological fluids, pointing out its bioresorbable properties [51]. Using sol–gel solution-processed ZnO, a bioresorbable liquid-electrolyte-gated TFT with 10^2 and 10^3 current ON–OFF ratios and 0.5-V operation, for water and phosphate-buffered saline solution electrolytes, respectively, was shown, [51]. Very recently, degradable TFTs and simple logic gates based on ZnO active layers, Al_2O_3 gate dielectrics, and molybdenum electrodes were realized on a poly(3-hydroxybutyrate-co-3-hydroxyvalerate) degradable substrate produced naturally by bacteria, and planarized through a spin-coated polyvinyl acetate layer [17]. Using room-temperature sputtering and evaporation, a TFT channel mobility of 1.3 cm^2/Vs, an ON–OFF current ratio $>10^6$, and stable device performance was demonstrated. Interestingly, not only the degradability of all materials was demonstrated from dissolution studies, but also a method to control the transience of the devices was implemented by utilizing a printed heater able to accelerate the decomposition [17]. At the same time, IGZO TFTs with dissolvable materials, including SiN_x, SiO_x, molybdenum, and PVA exhibiting field-effect mobilities of ~10 cm^2/Vs and ON–OFF current ratios of $\sim 2 \times 10^6$ were also demonstrated [52].

Simultaneously, studies of dissolution kinetics for IGZO in deionized water, bovine serum, and phosphate buffer saline solution were also provided. IGZO has also been utilized for biocompatible Na^+ ion-sensitive TFTs (ISTFTs) composed of ITO source/drain electrodes, a Ta_2O_3 gate insulator, and an Ag/AgCl reference electrode. A cytotoxicity evaluation of the Na^+-sensitive membrane and the ISTFT were also conducted, demonstrating full environmental biocompatibility of the devices [53]. Also, degradable ITO has been utilized as a channel, source/drain contact, and gate electrode with drop-casted pectin-based polysaccharide gate dielectrics for the realization of TFTs and simple logic circuits on polyethylene terephthalate (PET) [15]. Good electrical performance (channel mobility of 14.5 cm^2/Vs and ON–OFF current ratio $>10^7$) and dissolvability of the TFT stack (excluding the PET substrate) in water within 20 min were demonstrated [15].

It is important to highlight that this list does not serve as a complete overview of the state-of-the-art oxide TFTs incorporating sustainable materials, but rather, provides a picture of what has been done to allow a better understanding of where we currently are. In this regard, besides the still-large confusion that exists among the terms *degradable*, *biodegradable*, *compostable*, *biocompatible*, or *bioresorbable* in the literature, it is surely worth pointing out that the first examples of degradable/biocompatible/bioresorbable TFTs and simple circuits are becoming a reality. Figure 5

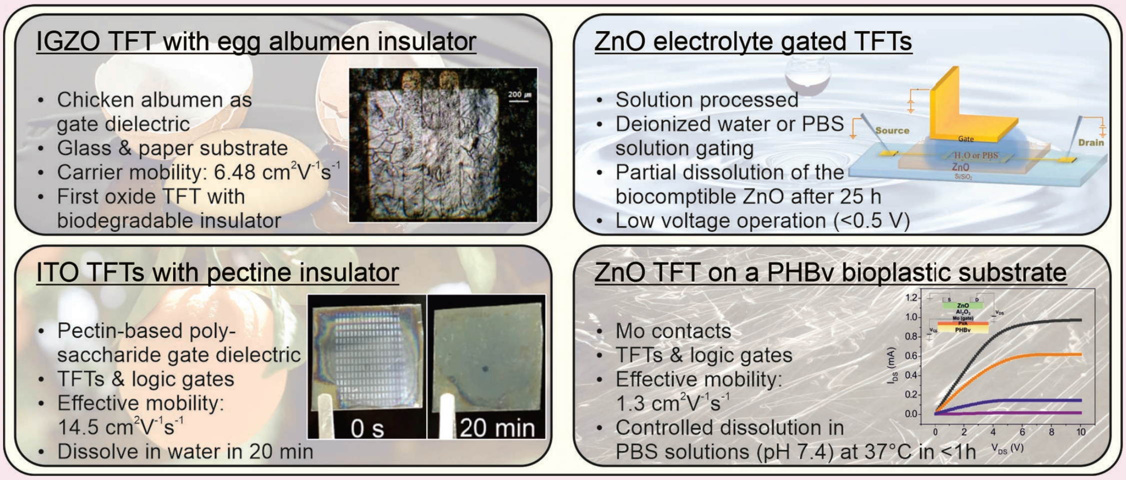

FIGURE 5. Examples of functional oxide TFTs partially or completely made from natural and bioresorbable materials and utilizing different oxide semiconductors [15], [17], [23], and [51]. Reprinted adapted with permission from [15]. Copyright 2022 American Chemical Society. [17] ©2023 IEEE. Used with permission of IOP Publishing on behalf of the Japan Society of Applied Physics (JSAP), from [23]; permission conveyed through Copyright Clearance Center, Inc. Figure reproduced with permission from Royal Society of Chemistry [51].

summarizes the most relevant examples of functional oxide TFTs incorporating sustainable materials.

An open issue is that the so-far realized examples are based on mostly dry processing of rather large structures, which can hardly be integrated into more complex circuits. Furthermore, integration is also limited by device performance and variability. Finally, existing solutions look mainly into one of the aspects of sustainability, e.g., degradability of materials. However, as already anticipated, sustainability in electronic devices needs to account for various levels of environmental, social, and economical impact throughout the entire lifecycle to reduce natural resources and CO_2 footprint. The implementation of this necessarily requires a holistic approach, which manifests itself along the following various pathways:

- eco-design that incorporates environmental requirements into specifications
- research and use of sustainable materials, critically limiting raw elements and pollutants
- sustainable manufacturing; minimizing the use of high-demanding energy, chemicals, and gases; and utilizing, in general, green fabrication routes
- usage and lifetime according to the required needs
- end-of-life strategies to reuse, collect, and recycle.

As shown by these examples and summarized in Figure 5, individual examples of eco-friendly design, sustainable materials, sustainable manufacturing are available, while complete usage and lifetime evaluations and end-of-life strategies, which can differ significantly according to the intended application, are still in their infancy.

Conclusion

The field of sustainable electronics started to attract considerable interest more than 10 years ago when the first transient transistors and systems based on ultrathin silicon were demonstrated. Immediately afterward, the related activities were dominated by silicon and organic semiconductors. These materials were relatively quickly used to demonstrate transient systems with very good electrical performance, and devices with excellent biodegradability. At this point, oxide-based semiconductors made a late entry into the field. Despite the late entry, the outstanding electrical performance of oxide TFTs demonstrate that, even when fabricated on large-area organic substrates using low-temperature processes, as well as their compatibility with a wide range of natural materials, makes them a serious contender for the realization of future eco-friendly electronics. This is highlighted by the already-available demonstrations of oxide TFTs made entirely or partially from biodegradable materials, and the integrated degradable logic circuits that have been realized. These devices are, however, far from any commercial use as their performance still requires improvement. This concerns in particular the stability of the devices, which first requires optimization when oxide semiconductors are combined with new, unconventional but biodegradable materials and fabrication processes, and, second, is affected by oxide semiconductors being generally very sensitive to hydroxy groups and humidity. This is in addition to the more general issue that all kinds of transient electronics must work reliably for the intended lifetime, but then degrade within a reasonable and defined period.

Oxide semiconductors have many opportunities to overcome these current limitations because the performance of such semiconductors is not only determined by the active material itself but is also affected by their exact chemical composition and deposition conditions, which can include sputtering, evaporation, solution processing, and pulsed laser deposition. Simultaneously, and similarly to all other semiconductors, they can be combined with various transient metals or, as in the case of ITO, can act as transient conductors themselves. By contrast to silicon, they can also be easily combined with a huge variety of biocompatible, natural, and large-area substrates. Thin-film insulators with high permittivity and defect-free interfaces with oxide semiconductors are mostly made from oxides too. These in particular, if the insulators should also be made from biodegradable materials, are more challenging to deposit on temperature-sensitive biodegradable substrates. In this context, it is important to acknowledge that the selection of materials, their combination and processing, and the device's design are not isolated problems but must be performed hand in hand to optimize biocompatibility, cost, scalability, and mechanical and electrical performance of corresponding TFTs simultaneously. Here, one major open issue is the fact that the currently most used and proven biodegradable oxide semiconductor, ZnO, normally forms polycrystalline thin films. This in turn promotes the formation of cracks under mechanical strain, and hence complicates the realization of flexible and wearable systems, which are one for the main drivers for more environmentally friendly electronics. This can be seen as a motivator to intensify the efforts on realizing bioresorbable electronics using amorphous oxide semiconductors.

Finally, it should not be underestimated that the biggest advantage of normally n-type oxide semiconductors might be their potential to be combined with predominantly p-type organic semiconductors to realize bioresorbable large-area CMOS electronics. By overcoming the traditional borders between oxide and organic electronics, the best of both worlds can be combined, and truly functional and circular thin-film electronics seem achievable. This in turn could ultimately enable approaches to create electronics that go beyond being not harmful over their entire lifecycle but decompose into constituents that have the capability of even creating a positive impact on the environment.

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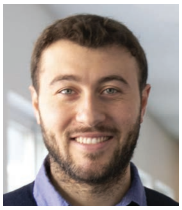

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