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Recent progress on biodegradable materials and transient electronics

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ABSTRACT

Transient electronics (or biodegradable electronics) is an emerging technology whose key characteristic is an ability to dissolve, resorb, or physically disappear in physiological environments in a controlled manner. Potential applications include eco-friendly sensors, temporary biomedical implants, and data-secure hardware. Biodegradable electronics built with water-soluble, biocompatible active and passive materials can provide multifunctional operations for diagnostic and therapeutic purposes, such as monitoring intracranial pressure, identifying neural networks, assisting wound healing process, etc. This review summarizes the up-to-date materials strategies, manufacturing schemes, and device layouts for biodegradable electronics, and the outlook is discussed at the end. It is expected that the translation of these materials and technologies into clinical settings could potentially provide vital tools that are beneficial for human healthcare.

1. Introductions

Electronics has made tremendous impacts on human society and has been widely used in almost every field, including telecommunication, entertainment, and healthcare, to name a few. While the long-lasting stable operation is a hallmark of conventional electronics, an emerging type of device that possesses "transient" function is gaining increasing attention recently [1–7]. Such devices are made of biodegradable materials, and can completely or partially dissolve, resorb or physically disappear after functioning in environmental or physiological conditions at controlled rates, and are termed as "transient electronics", or "biodegradable electronics" for biomedical or eco-friendly applications. Biodegradable electronics as temporary implants can be safely absorbed by the body after fulfilling its therapeutic and diagnostic functions, similar as biodegradable sutures or cardiovascular stents, and as a result eliminates second surgeries for device retrieval and decreases associated risks of infection. For green electronics, introducing biodegradability to consumer electronics or environmental monitors is expected to greatly alleviate landfill and environmental issues caused by electronic waste (E-waste, more than 50 million tons each year) [8,9] and eliminate associated costs and risks resulting from recycling operations. Furthermore, transient devices capable of self-destruction that protect information from unauthorized access can be used as data-secure hardware.

Demonstrated transient devices so far are mostly associated with degradation in aqueous solutions targeting biomedical or environmental applications [1–4,10]. Researchers have performed studies on biodegradable materials for transient electronics, including materials dissolution chemistry, degradation modeling, fabrication techniques, device integration, etc. Early attempts have been focusing on organic materials including natural or synthesized biodegradable polymers, and partially degradable devices have been achieved with contributions mainly from substrate components [11–13]. Recent studies have showed that monocrystalline silicon nanomembranes (mono-Si NMs) dissolve in physiological environments with rates ranging from a few nanometers to more than 100 nm per day [1,2,14,15], depending on the types of aqueous solutions. Together with degradable inorganic dielectrics, metals, and polymer substrates, dissolvable Si NMs enable fully biodegradable electronics with superior operation characteristics that can also be compatible with semiconductor foundry process [16,17]. Novel fabrication techniques have been developed to adapt the sensitive nature of biodegradable materials to device integration, preventing materials destruction by solvent, temperature, or water. A variety of fully biodegradable devices in physiological solutions have been demonstrated, including thermal

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therapy device [1], intracranial pressure sensor (ICP) [18], electrocorticography (ECoG) recording systems [19], radio frequency (RF) electronics [20], batteries [21], drug delivery systems [22], etc. In order to achieve both stable operations for a certain period of time and then transience at a later stage, encapsulation materials are critically important. The functional lifetime of achieved transient devices is mostly defined by the degradation time and water permeability of encapsulation materials, and the thickness of active electronic components. External trigger stimulus (moisture, temperature, light, mechanical force, etc) represents an alternative factor to determine the starting point of transience, and demonstrated triggered degradation in non-aqueous environments are mostly associated with non-biological applications. In these scenarios, devices are either capable of full transience, or partial degradation.

As an emerging technology, transient electronics has made fast development since it has been first proposed on 2012 [1], and more possibilities are to be explored to further expand its opportunities to be used for healthcare and green electronics. This review summarizes recent progress on biodegradable materials and electronics, focusing on biomedical and eco-friendly applications. It is noted that most biodegradable electronics designed for biomedical applications can be well adapted for eco-friendly usage. A wide range of biodegradable materials will be first reviewed, followed by an introduction of various novel fabrication techniques. Representative biodegradable functional electronic systems and eco-friendly devices will be described, and perspectives to further advance high-performance multifunctional transient electronics will be discussed.

2. Materials

Biodegradable electronic materials capable of complete degradation and physical transience in physiological or environmental solutions have been studied in order to establish a comprehensive material database for the construction of biodegradable electronics. Biodegradable organic materials (natural or synthesized polymers) that have been extensively studied as biomedical implants (e.g. sutures, stents, scaffolds for regenerative medicine) often serve as passive components where the function is defined by their mechanics and structure. While inorganic semiconductors, metals, dielectrics are shown to have excellent degradation behavior as well as superior electronic properties. Combination of these two categories of materials enables high-performance active devices and sensors that can significantly expand the possible applications of biodegradable electronics. It is noted that although the degradation of inorganic materials has been studied for years in the context of corrosion science, most studies focus on materials in the bulk format and in relatively more corrosive environments (e.g. strong acid or basic solutions). Exploration of materials degradation in biological solutions in the thin film format relevant to electronics standards is therefore necessary, as well as establishing the correlations between materials degradation and electronic properties. Available studies have investigated the degradation rates of dissolvable thin film electronic materials, including semiconductors, dielectrics, metals, etc, mostly in de-ionized (DI) water and simulated bio-fluids such as phosphate buffered saline and phosphate buffered solutions. It is noted that the *in vitro* degradation behavior could be very different from the *in vivo* degradation due to the presence of complex components (e.g. proteins, cells). The influence of various *in vivo* components on the degradation behavior is under investigation, e.g. recent studies have shown that proteins can possibly slow down the dissolution rates of Si [23]. Biocompatibility of these materials has also been evaluated both *in vitro* and *in vivo* through cell toxicity tests and animal models. Overall, the research on biodegradable materials for electronics is still in progress. Although materials candidates capable of full degradation have been proposed and various functional devices have been demonstrated, further materials studies are necessary, including extensive investigations of material interactions with components in real biological solutions (e.g. proteins, cells, etc), local enrichment of dissolution by-products and the associated biocompatibility, exploration of effective encapsulation materials, etc. In the following sections, materials proposed for biodegradable electronics are discussed in terms of semiconductors, dielectrics, metals, and substrate and encapsulation materials. Available modeling tools capturing materials degradation rates will also be given.

2.1. Semiconductors

Semiconducting materials are the most important component in electronics that determines the performance. Literature studies have showned that mono-Si NMs (30–300 nm), polycrystalline silicon (poly-Si), amorphous silicon (a-Si), germanium (Ge), silicon germanium alloy (SiGe), indium-gallium-zinc oxide (a-IGZO), and zinc oxide (ZnO) are dissolvable in physiological aqueous solutions [1,2,14,15,24,25]. Great efforts have been devoted to the investigation of dissolution mechanism of single crystal Si NMs in various bio-fluids and aqueous solutions, as Si offers excellent operational characteristics as well as alignment with the deep base of scientific and engineering knowledge associated with the well-established electronics industry. In order to investigate the dissolution kinetics of mono-Si NMs, arrays of Si NMs are fabricated on silicon oxide/silicon (SiO_2/Si) substrates using a commercially available silicon on insulator (SOI) wafer. The changes of the thickness of Si NMs are monitored as a function of time by profilometer or atomic force microscopy (AFM). Representative dissolution process of Si NMs in bovine serum is captured by the AFM as shown in Fig. 1(a) [14]. It is found that doping levels, pH, temperatures, concentrations and types of ions, and proteins in the solution can all have significant effects on the dissolution rates of mono-Si NMs, as shown in Fig. 1(b) and 1(d) [2,14,15,23]. Higher temperatures and pH levels speed up dissolution rates of Si NMs, while high doping levels ($>10^{20}$ cm^{-3}) significantly decrease the rates [2,14]. Moreover, chlorides and phosphates concentrations above a certain level greatly accelerate silicon dissolution even in a near-neutral aqueous solution (pH~7.5) [15]. A recent report suggests that the calcium concentration can significantly increases the dissolution rates, while the presence of silicic acid and proteins (albumin) slows down the reactions [23]. Density functional theory (DFT) and molecular dynamics (MD) simulation tools can be applied to reveal the underlying physics of silicon dissolution behavior [15]. It is found that silicon dissolution proceeds through the nucleophilic attack of silicon surface bonds, which weakens the interior bonds of surface silicon atoms (backbonds) and therefore increases their susceptibility to further ion attack, as shown in Fig. 1(c). Similarly, dissolution rates of poly-Si, a-Si, Ge, and SiGe are greatly affected by pH, temperatures, proteins and types of ions. For example, the rates of these materials at the physiological temperature (37 °C) are higher than those at room temperature. At similar pH, bovine serum leads to dissolution rates that are 30–40 times higher than those of a phosphate buffered solution at 37 °C for poly-Si, a-Si, and single crystal Si [24]. *In vitro* cytotoxicity studies on mono-Si, poly-Si, a-Si, Ge and SiGe materials, and *in vivo* studies of mono-Si membranes implanted in subdermal regions of BALB/c mice suggest good biocompatibility [2,14,24]. Besides the aforementioned inorganic semiconductors, PDPP-PD synthesized from diketopyrrolopyrrole (DPP) has recently been proposed as an eco-friendly, biocompatible

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