Lignin and Keratin-Based Materials in Transient Devices and Disposables: Recent Advances Toward Materials and Environmental Sustainability

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Cite This: ACS Omega 2022, 7, 10854−10863

ABSTRACT: Rising concerns and the associated negative implications of pollution from e-waste and delayed decomposition and mineralization of component materials (e.g., plastics) are significant environmental challenges. Hence, concerted pursuit of accurate and efficient control of the life cycle of materials and subsequent dematerialization in target environments has become essential in recent times. The emerging field of transient technology will play a significant role in this regard to help overcome current environmental challenges by enabling the use of novel approaches and new materials with unique functionalities to produce devices and materials such as disposable diagnostic devices, flexible solar panels, and foldable displays that are more ecologically benign, low-cost, and sustainable. The prerequisites for materials employed in transient devices and disposables include biodegradability, biocompatibility, and the inherent ability to mineralize or dissipate in target environments (e.g., body fluids) in a short lifetime with net-zero impact. Biomaterials such as lignin and keratin are well-known to be among the most promising environmentally benign, functional, sustainable, and industrially applicable resources for transient devices and disposables. Consequently, considering the current environmental concerns, this work focuses on the advances in applying lignin and keratin-based materials in short-life electronics and single-use consumables, current limitations, future research outlook toward materials, and environmental sustainability.

1. INTRODUCTION

The environment and human society are systems that coexist and interact. In some cases, these interactions are weak and, in others, strong. The extent of human interactions with the environment, and the consequential impacts on each other, partially or wholly, are unpredictable and can have far-reaching ramifications. For example, the concerns expressed over the detrimental effects of pollution from resource use, such as plastics and electrical/electronic wastes (i.e., e-wastes), have grown over the last two decades. However, more disturbing facts behind many of the warnings on the negative impacts of these pollutants are the threat to environmental sustainability, the quality of human life, and by extension, the future of humanity. Hence, humankind’s persistent calls for a concerted attitudinal change toward limiting the undesirable impacts of environmental stressors cannot be overemphasized since we cannot continue in the same mindset that brought us to the current situation and expect a better environment and future. Broadly speaking, e-wastes are superfluous and discarded electrical and electronic devices or equipment in the environment, which may be stand-alone or part of a more extensive system or machine functioning together, such as end-of-life vehicles (e.g., fuel-based or electric vehicles), secure memory devices, batteries and accumulators, mobile devices, solar panels, air conditioners, toys, light bulbs, robotic systems, television sets, and so on, to mention but a few. In 2016, the global e-waste generated exceeded 44 million metric tons (Mt). With exponents such as economic growth, advancement in technologies, and the continual hardware overhaul (i.e., modernization and upgrade), e-waste generation is expected to grow in the future (an estimated 74.7 Mt by 2030). Table 1 shows the global e-waste generation (volume and per capita values) and collection rate in 2016, demonstrating the disparity in waste generated, collected, and recycled, a clear indication that the challenges presented by e-wastes and component materials are an urgent environmental issue.

Received: December 30, 2021
Accepted: March 7, 2022
Published: March 25, 2022
Table 1. Regional e-Waste Generation and Collection Rate in 2016

| region   | by volume (Mt) | per capita (kg/capita) | the collection rate for recycling (%) |
|----------|----------------|------------------------|---------------------------------------|
| Asia     | 18.2           | 4.2                    | 15                                    |
| Europe   | 12.3           | 16.6                   | 35                                    |
| United States | 11.3     | 11.6                   | 17                                    |
| Africa   | 2.2            | 1.9                    | unavailable                           |
| Oceania  | 0.7            | 17.3                   | 6                                     |

*This table summarizes data available in ref 2c.*

E-waste streams are comprised of heterogeneous mixtures of metals (e.g., gold, aluminum, copper, zinc, silver), metalloids (e.g., boron, silicon, tellurium, antimony, etc.), rare earth metals, glass, plastics, etc. Besides, the exact compositions of e-wastes vary at a given point in time, depending on the current technological advancement, which makes the e-waste stream a complex and ever-evolving threat to the living and nonliving components of the environment. Moreover, plastic materials are usually an integral component of electrical and electronics systems/equipment, sometimes employed as encasements (e.g., mobile phones), insulators or insulation (e.g., electrical wirings), as well as functional materials (e.g., electronic displays); coupled with the fact that most of the conventional plastics, currently employed in industry, are not only fossil-based, they have been associated with various adverse health and environmental burdens and also exhibit delayed mineralization in natural environments, further exacerbating the complex challenge of e-waste. Various detrimental health and environmental impacts are associated with e-waste and plastic pollution. For example, e-wastes are significant sources of high levels of toxic metals and compounds in the environment such as lead, cadmium, mercury, copper, persistent organic pollutants, etc., polluting the air, water, soil, and even sediments near the dump and operational sites for electronic system manufacturing; consequently, biological exposures of humans to these hazardous systems present detrimental health challenges ranging from respiratory disorders, chronic gastritis, gastroduodenal ulcers, skin infections, leukemia, and so on. Moreover, plastic pollution is associated with myriads of undesirable health and environmental consequences such as carcinogenesis of living cells, initiation of mutagenic processes of microbial communities, respiratory disorders, obesity, reproductive impairment, diabetes, thyroid dysfunction, loss of ocean productivity, and global warming.

Hence, the urgency in ameliorating and potentially reversing the damaging impacts of materials used on the living and nonliving components of the environment have led to a concerted pursuit for the accurate and efficient control of the life cycle of materials (e.g., e-waste and plastics) and their subsequent dematerialization (with net-zero impact) in target environments, which is an integral component for attaining the mandate of the sustainable development goals (SDG). Consequently, necessity is laid upon the scientific community and stakeholders along the materials value chain, toward ecodesign (i.e., the idea of removing environmentally associated challenges and problems during a product development life cycle, without compromising on cost, efficiency, or quality), as demonstrated in Figure 1, toward lessening the growing detrimental impacts and environmental burdens resulting from material usage, thereby lending credence to the emerging concept of transient technology (i.e., “green technology”), which broadly encompasses the idea of purposefully designing or fabricating materials, devices, or systems and sundry components thereof, from materials that possess the inherent ability to partially or wholly mineralize or lapse into the natural environment (usually initiated by a stimulus), within a fleeting period after the desired functional lifetime of such material or component is reached. Thus, by extension, the dematerialization and subsequent environmental resorbing should have a net-zero impact or offer a positive ecological impact and must be associated with biocompatibility (i.e., nontoxicity in living systems), a fundamental benchmark for “green” or ecofriendliness.

As part of the literature review, essential reports on the emerging field of transient technology within the past decade were collated, which is imperative for assessing and reevaluating biomaterials in transient technology to inform current insights and inferences toward establishing facts in forming new research directions. In this short report, we examine important advancements in using biomaterials such as lignin and keratin as efficient and industrially applicable resources for fabricating transient devices and single-use bioreabsorbable consumables. Underlining critical factors hindering the applications of biorenewable materials as alternatives to conventional materials, we highlight significant initiatives aimed at sustainable solutions, with an outlook on future research directions toward more efficient and benign biodegradable materials for sustainability.

2. PROMISE OF TRANSIENCY IN MATERIALS APPLICATIONS (“GREEN” TECHNOLOGY)

There is no gainsaying that materials consumption will continue to rise in the coming decades as a result of advances in materials development and subsequent replenishment of in-use stocks, thus setting the stage for the need to develop consolidated strategies in mitigating associated detrimental
impacts of materials exploration and applications through programmable life cycle systems.5c Transient technology is a promising opportunity to meet modern materials needs and assuage a number of persistent environmental challenges presented by conventional material use as enumerated by Olivetti et al. 5c Whether it entails understanding material degradation pathways, modeling, and fabrication techniques, the general concept of transiency is rooted in closing the material loop toward a friendlier and sustainable materials system. On this basis, the core requirements for transient devices and disposable materials are anchored in efficient functionality, low-cost implications, ease of disposability, ease of fabrication, and design flexibility, biodegradability, biocompatibility, and environmental benignity. 6a,8a,b Humanity must demonstrate commitment in the direction of greener material systems and embrace the transition toward biorenewable materials (e.g., cellulose, starch, lignin, chitosan, silk, etc.) that offer the benefits of materials transiency, benignity, and ecology as substrates and encapsulating materials for the fabrication of end-use applications in electronic devices, associated components and disposables in a range of industries such as healthcare, single-use disposable devices, brain–computer interfaces, advanced displays, next-generation storage systems, to mention but a few.8c The growing demands for the prevention of sensitive data leakage, the need for self-destructing devices, reducing the impact of e-waste, assuaging the damaging effect of plastic pollution, and eliminating supplementary surgeries after surgical implantations, further demonstrates the possibilities and promises of transient technology as an emerging and ecofriendly technological concept in materials science and engineering, a revolutionary paradigm shift in the quest for greener materials and sustainability.1c,8f,9a Moreover, as pointed out by Green-gard,9b the application possibilities of transiency in devices and materials are almost endless. For example, it will allow for the deployment of sophisticated drones and robots into enemy lines without the worries of adversaries gaining control of the technical know-how of such technologies. Furthermore, single-use and disposables such as diapers, latex gloves, condoms, straws, syringes, plastic carrier bags, sanitary pads and wipe napkins, medical plasters, facemasks, and so on will become less environmental burdens,b a s e do nt h e p r i n c i p l e o f engineering the materials used for these products to depolymerize and disappear on exposure to specific environmental stimuli. In addition, green materials technology is central to the emerging field of neurotechnology and next-generation tissue engineering and regeneration (e.g., scaffolding materials).8c,9c In summary, because Earth is currently the only habitable place humanity calls home, it is incumbent on us to sustain this planet and ensure we do not squander the enormous opportunities offered by the concept of transient materials technology.

3. BIOMATERIALS IN TRANSIENT DEVICES AND DISPOSABLES

The future and sustainability of the chemicals and allied industries will be driven by critical technological trends, which will require innovations, adaptability, and technologies geared toward environmentally friendly materials that demonstrate commensurate or better functionality and efficiency to those traditionally employed.9d,10a Biomaterials (i.e., biorenewables) as alternative resources (examples in Table 2) have shown, among others, the benefits of energy efficiency (e.g., low-cost), efficient substitutes for conventional applications, materials for sustainable and next-gen manufacturing, ecofriendliness, advantages of water conservation and carbon sinks, impressive functionalities in materials applications, etc.9d,10a,11a,12 With regard to transient devices or green technology, biorenewables and their derivable materials such as lignin and keratin have shown promise as functional, efficient, and applicable greener substrates and encapsulating materials for transient devices and single-use disposable materials.

3.1. Lignin. This biopolymer comprises about 15–30% of the dry mass of lignocellulosic biomass, and ranking top in the emerging technologies of the future, lignin and derivable materials will play critical roles toward the circular economy,
next-generation manufacturing, and sustainable material- s. However, recent developments have seen an increasing interest in utilizing lignin and derived materials in various application ranges (Figure 2), for example, in the fabrication of transient devices and single-use disposable materials.

3.1.1. Transient Electronics. Chupka et al. established that lignins demonstrate a work function as current carriers in the conductivity region, between 2.0 and 2.2 eV, thereby making lignins and their derivable materials promising candidates in dielectric and semiconducting applications. The electrical properties of lignin are inherent and diverse, with the utmost influence arising from the electronic and ionic components in the molecular structure (i.e., functional groups such as carboxyl, carbonyl, quinoid, phenolics, etc.). The electrical conductivity of lignin results from carbocations at phase separations favoring the activation mechanism toward conductivity; in addition, the lignin polymer as an amorphous heterogeneous system demonstrates a considerable difference in functionality depending on the chemical composition, method of isolation, and experimental conditions (e.g., temperature) during preparation, thus conferring on them tunable electrophysical properties. Lignins, as bioresources, offer the possibilities for the production of a wide range of new class of biodegradable, biocompatible, and ecological materials from carbon-based materials for anodic electrodes to photo-cross-linked polymer systems (referred to as BAMTU-SH). The glass transition temperature ranged between 20 and 49 °C and exhibited a tensile strength and elongation at break of 2.9–18.2 MPa and 103.5–305.6%, respectively. The authors determined that the mechanical properties of BAMTU-SH networks could be adjusted to suit application demand by controlling the functionality of thiols. It was further shown that, below 310 nm, the BAMTU-SH networks’ transmittances were near zero, a demonstration of their ability to hinder short-wave ultraviolet light. It was asserted that the materials have potential applications as flexible substrates for transient devices and can be easily degraded in mild acetic acid (acid/water at 1:1) within 2.5–6 h, degradation time. Other studies evidenced that fossil-based poly(ε-caprolactone, PCL), a common material employed as a flexible substrate and encapsulant in transient electronics, could be effectively and efficiently replaced with biobased poly(methyl-ε-caprolactone, PMCL), polymerized from methyl-ε-caprolactone monomer, a derivative of lignin-based p-cresol. The benefits of PMCL over PCL are noteworthy. They include improved amorphousness (i.e., ease of processability, better dimensional stability and impact resistance), lower glass transition temperature (~60 °C), and lower overall economic and environmental cost implications. Nevertheless, despite lignin-based materials having enormous potentials for the production of transient devices, the irregular chemical structure and polyfunctional composition of lignins is a fundamental challenge in the optimization of their electrophysical properties, thereby, demanding extensive studies in both theoretical and basic principles of understanding the parameters required for the optimal application in electronic devices toward materials sustainability.

3.1.2. Disposable Materials. Lignins are well-known for their functional role as structural and support systems in plant cell walls. Hence, recent years have seen increasing research studies in applying lignin-based materials (e.g., composites) as high-performance and greener alternatives to fossil-based polymers employed in single-use disposables, e.g., packaging films and membranes, toward potential solutions of plastic pollution. For example, a biodegradable disposable packaging capable of composting over 60% of the material within 90 days has been fabricated. By modifying a blend of polyhydroxalkanoates (PHA) and high crystalline poly(3-hydroxybutyrate, PHB) with grape seed lignin (GS-L), the antioxidative capacity, gas barrier, and degradability of the derived polymer film were enhanced appreciably. The PHB/PHA-derived packaging film exhibited a Young modulus in the range of 240 MPa, optimal tensile strength of 6.6 MPa, and more than 90% elongation at break. At about 1 wt % of the GS-L incorporation into the PHB/PHA blend, the evolution rates of oxygen and carbon were 7.3 and 36.3 cm³ m⁻² 24 h 0.1
MPa, respectively. As demonstrated in Figure 3, it was established that, depending on the lignin concentration, the inhibition percentage for the radical scavenging assay ranged between 29.2 and 100%. In conclusion, it was determined that toxicological studies demonstrated the nontoxicity of the PHB/PHA/GS-L after biodegradation and even had a net positive impact on the germination rate of white mustard seeds (Sinapis alba L.), an indication of the ecofriendliness toward closing the material use loop.

Jiang et al. by drawing inspiration from wood-based structures, fabricated a facile and low-cost lignin-cellulose with UV-blocking, water-stable, and biodegradable paper composite material as a robust and efficient replacement for fossil-based plastic packaging and as a substrate for flexible integrated circuit boards, 3-D printing, wearable devices, and so on. On the basis of the fundamental principle that the mechanical properties of woody materials are directly dependent on the extent of inter- and intramolecular chemistries between lignin and cellulose in woody structures, by partial delignification and controlled successive intercalation of lignin, as reinforcement, into the cellulose-based paper, without destroying the cellulose structure (Figure 4a), a high-performance material with impressive flexibility, Young’s modulus of 10 GPa, and tensile strength of 200 MPa, was produced, exceeding those of petroleum-based commercial plastics (Figure 4b–d).

Wood-based (liquid-wood polymer), biodegradable, and easily compostable drinking straws referred to as BIO LIG have been developed. Entirely fabricated from lignin and cellulose, these straws have demonstrated superior performance compared to conventional plastics, offering a sustainable alternative for reducing environmental impact. The development of such materials exemplifies the potential of lignocellulosic biorefineries in addressing global sustainability challenges.

Figure 3. Inhibition percentages of the radical scavenging assay 2,2′-azino-bis (ethylbenzothiazoline-6-sulfonic acid) (ABTS). Reproduced from ref 16j. This article is licensed under a Creative Commons Attribution-Noncommercial 3.0 Unported License.

Figure 4. Overview of the wood-inspired preparation method and results. (a) The delignification process and intercalation process. (b) Demonstrating the comparative robustness of the lignin-cellulose composite and cellulose papers. (c) Comparing the tensile strength of the developed lignin–cellulose paper against commercial plastics. (d) This lignin–cellulose composite with impressive high strength, thermal and water stability, and UV-shielding capability is a promising candidate for reducing humanity’s dependence on fossil-hydrocarbons, mitigating plastic pollution, and improving environmental and material sustainability. Reproduced from ref 16k. Copyright 2021 John Wiley & Sons, Inc.
3.2. Keratin. Waste poultry feathers are a rich source of renewable and sustainable keratin-derivative material (composed of more than 90% keratin protein); it is estimated that the global poultry industry generates 8–9 million Mt of waste bird feathers per annum.17d And with the sector currently growing at a compound annual growth rate, CAGR, of 3.8% and expected to reach 7% CAGR by 2025, it is deducible that the volume of waste feathers will increase consequentially.17b Moreover, these bonds, while conferring structural integrity on the material, consequently confer polarization characteristics (e.g., interfacial polarization, dipole–dipole moments, atomic/electronic interactions),17i a fundamental feature of all matter containing elementary particles, e.g., atoms, thereby making protein-based materials such as keratin a promising candidate for the fabrication of electronic devices. For example, solid-state electrolytes were obtained through the gradation arrangement of thin films of human-hair keratin and then used to fabricate transient resistive switching memory devices.17k In addition to the nonvolatile nature of Ag/keratin/FTO (i.e., silver/keratin/fluorine-doped tin oxide) memory devices, it is transparent (with good transmittance), and the memory retention performance is reproducible. Capable of retaining operational stability for over seven weeks, these devices exhibited a sufficient resistance level larger than 10^3 OFF/ON and more than 10^4 s retention time. It was determined that these memory devices are dissolvable in deionized water in less than an hour.17l Elsewhere, wool-derived keratin combined with graphene nanoparticles (GnPs) has been used to manufacture green resistor plane capacitors and inductors, subsequently assembled to obtain analogue electrical circuits for high-pass filters and resonators.17k A 40 cm ribbon of the conductive keratin composite was used to fabricate the inductors. The covering of the dielectric core with the conductive strip was expedited by using about 5% wt of polyglyceryl to enable the casting process, which was done after conditioning under high humidity. Moreover, the freestanding capacitors were obtained by assembling keratin-GnPs electrodes, consisting of about 30% wt of GnPs and the keratin as the dielectric material.17l The discrete components were subsequently made into a primary analogue circuit with an assessed capacitance of 86 pF. When linked up in series to a 40 kΩ nanocomposite resistor, the circuit exhibited the typical behavior of a high-pass filter with a cutoff frequency range of 46 kHz, thereby functioning as a differentiation for frequency below the targeted cutoff range.17k

In another report, a green, biocompatible, recyclable, tunable artificial wearable skin was developed from conductive and skin-look-like hydrogels consisting of hydrolyzed keratin, sodium chloride, and poly(vinyl alcohol).17l The artificial skin exhibited remarkable mechanical properties, comparable to that of natural leather, such as a considerable strength of 1.36 MPa, toughness of 3.45 MJ/m^3, and a low modulus determined to be in the range of 110 kPa, while possessing an antifatigue-fracture ability, i.e., capable of over a thousand loading–unloading cycles with no physical damages, thereby mitigating the instances of discomfort when worn and also enhancing durability.17l The fabricated skin was demonstrated to be suitable as a full-body suit as it effectively...
detects motion signals uninterruptedly with outstanding sensitivity toward distinguishing precisely detailed actions, rapid responsiveness, and negligible hysteresis, and the ion-transporting properties were proportional to that of natural skin, with a remarkable strain-sensitive gauge factor (GF) of 4.92 and impressively low detectable strain of 0.25%. \[1\]

3.2.2. Disposable Materials. Studies have evidenced that bioprotein packaging materials hold promise as efficient replacements for fossil-based packaging due to their biodegradability, low-cost, low toxicity, renewability, and sustainability; however, most bioprotein materials need to be modified to meet basic food packaging requirements such as moisture contents, thermal stability, low-microbial activities, etc. \[184\] Scientists have endeavored to graft, \[185\] cross-link, \[186\] plasticize, \[187\] mix, or incorporate \[188\] protein-based materials with other materials to obtain desired results. However, most protein-based films are modified by employing synthetic modifying agents such as glutaraldehyde, glyoxal, and formaldehyde to enhance their gas barrier properties, water and thermal stability, flexibility, and processability; \[189\] nevertheless, associated health and environmental concerns of the potential migration of these chemicals into the food web cannot be trivialized; hence, recently, efforts to find benign and ecofriendly modifying techniques toward ecosustainable and disposable bioprotein materials have intensified. For example, to enhance the water stability of starch as an ecological and sustainable material, Oluba et al. \[18h\] incorporated keratin, extracted from waste chicken feathers, into ginger starch to produce a biocomposite film. An increase in the keratin content resulted in the loss of materials transparency, indicating better UV-barrier properties, an attractive quality for food packaging materials. It was observed that keratin enhanced the toughness of the ginger starch films by improving the tensile strength and elongation at break, possibly resulting from the intermolecular interactions between the functional groups of the starch (i.e., hydroxyl groups) and that of the keratin (i.e., carboxyl and amino groups). \[188\] Moreover, the moisture content of the fabricated biofilms was observed to be significant, demonstrating the possibilities for a better shelf life of packaged products and lower microbial activities. It was concluded that the films when buried in soil degraded by over 50% of the initial mass in 12 days. \[18i\] In another report, edible dialdehyde carboxymethyl cellulose (DCMC) cross-linked with feather keratin (FK) subsequently plasticized with glycerol was cast into a packaging film as a potential replacement for fossil-based plastic packaging. \[18j\] The produced film, which is 100% biodegradable and ecofriendly, exhibited good UV-barrier properties (attributable to the aromatic/amino functional groups) and transmittance. \[18a\] As already mentioned, from the viewpoint of food safety regulation, the higher the UV-barrier, the better for food packaging because oxidation and discoloration of packaged food items are not desirable. It was, however, observed that although the FK/DCMC films offer low-cost, environmentally benignity, transiency, and potential for food packaging applications, the moisture content stability of the FK/DCMC films was irregular owing to the presence of hydrophilic groups inherent in the DCMC. \[18a\]

It is known that synthetic polymers such as polyethylene (PE), polystyrene (PS), polyvinyl chloride (PVC), and urea-formaldehyde (UF) are generally non-biodegradable. For example, Otake et al. \[18i\] investigated the susceptibility of various fragments of synthetic polymers to biodegradation under bioactive soil for 32–37 years; it was evident that low-density PE (LDPE) among other synthetic polymers (PS, PVC, UF) showed a detectable level of biodegradation. Because plastics have and will continue to play critical roles in the advancement of human civilization and a world without plastics is improbable, recent years have seen increasing efforts toward the fabrication of greener and environmentally benign alternatives to fossil-based plastics that are biodegradable and biocompatible, and have transient characteristics. \[18k,16i\] Following this line of thought, Du et al. \[18l\] fabricated a high-performance, biodegradable, biocompatible, and transient flowerpot based on fermented straw fibers urea-formaldehyde adhesive (UF) modified with hydrolyzed keratin (HK) from waste poultry feathers. It was argued that the keratin acting as an acidic catalyst resulted in the self-polymerization of the UF; moreover, because of the multitude of reactive sites in the HK, free formaldehyde content in the composite material was significantly reduced, thereby mitigating the possibilities of gaseous emissions of free formaldehyde into the environment. \[18m\] The authors determined that the HK effect on the UF solid content and pH was nonexistent. The presence of keratin in the flowerpot provided the microbial community with a rich nutrient source, thereby resulting in the rapid degradation of the UF adhesive and, subsequently, the flowerpot, which is beneficial to the environment. \[18n\]

4. SUMMARY AND OUTLOOK

The pursuit of transiency in material applications toward environmental and material sustainability will continue to rise in the following decades with global trends such as innovative materials, artificial intelligence, security and protection of personal data, mobility, and territorial integrity. Bioneroenewable materials have demonstrated capacity as efficient, effective, and formidable alternatives toward replacing fossil-based materials and their derivatives in the quest for a sustainable Earth. Notwithstanding, the sophistication required for transiency in materials and potential applications has given rise to the demand for further research into understanding the interfacial chemistries of various bioneroenewables and their applications or incorporation into a new class of novel materials. There is a need to establish a database of dissolution rates for different bioneroenewable materials relevant to the design and fabrication of substrates and components for transient devices and disposal materials to determine demonstrable vehicles for the application of these materials in electronic devices and disposables with real-world time scales. Furthermore, the need to understand the kinetics of bioneroenewable materials dissolution in various solutions/solvents for improved material-function parameter estimations toward efficient functionality and optimal application cannot be overemphasized.

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ACKNOWLEDGMENTS

The authors would like to acknowledge financial support from the Department of Science and Technology (G6ACH35), Council for Scientific and Industrial Research, Pretoria (086ADMIN), and the University of Johannesburg (086310), South Africa.

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