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Fully Degradable Food-Based Solenoids and Radio Frequency Circuits for Green Electronics

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Herein, edible solenoids are introduced, which are realized by coating spaghetti with edible gold leaves, creating fully edible and functional radio frequency (RF) electronic components. As a proof-of-principle of their use in RF circuits, a completely edible passive inductor-capacitor (LC) resonator at ≈200 MHz is demonstrated. The results significantly expand the applications of edible electronics to RF regime, supporting future developments in edible sensing and edible robotic systems, emerging fields with a high grade of sustainability.

1. Introduction

The recent decades have witnessed a rapid increase in the production and distribution of electronics for both consumer and industrial applications, leading to a dramatic increase in e-waste that now represents a significant contributor to the large-scale release of toxic pollutants into the environment.[1] As the demand for electronics products keeps surging, increasing attention is hence being devoted to technological solutions that are compatible with the circular economy paradigm through either reuse and/or recycle mechanisms, or that are inherently less reliant on conventional electrical and electronic materials.[2] The latter approach hence exploits bioderived materials for the realization of electronic components and circuits, possibly by employing low-energy and low-cost fabrication techniques. For these reasons, this approach is particularly suited for applications where devices operate for relatively short time windows prior to disposal. In recent years, there has been a growing inclination toward the development of edible electronic devices, i.e., devices exclusively derived from food sources.[3] These ecologically sustainable devices present the intriguing opportunity for 100% environmentally friendly disposal, through the generation of compost, biogas, animal feed, or even human consumption.[4] In addition, the potential of edible electronics in novel application scenarios is evident, particularly in the context of smart pills designed for remote monitoring of human health,[3] for nontoxic tags in the food industry contributing to the creation of tools aimed at reducing food waste,[5] as well as components of edible actuators, supporting the development of edible robotics.[4] Edible electronic components have been already explored in the literature, including edible substrates,[6] resistor–capacitor (RC) and resistor–inductor–capacitor (RLC) filters,[7] memristors,[7b,8] batteries,[9] supercapacitors,[10] and coils.[11,12] Importantly, many edible sensors have also been explored, such as food spoilage sensors,[13] strain and temperature sensors,[14] piezoelectric sensors,[7a] pressure sensors,[10] irradiation sensors,[15] pH sensors,[16] and humidity sensors.[17] The primary obstacle within the domain of edible electronics lies in the discernment and effective processing of food-derived substances and food additives, i.e., substances regulated for use as food ingredients or in the food processing, which may exhibit functional suitability in the electronics sector. This challenge becomes particularly pronounced when considering the incorporation of essential components like conductors and semiconductors. Regarding the formers, despite the availability of organic alternatives, such as activated carbon composites[18] and melanin,[19] their electronic conductivity typically proves inadequate for ensuring the functionality of critical components like wires, interconnectors, antennas, and inductors (generally necessitating a minimum conductivity of $\approx 10^3$ S cm^{-1}).[3] Nevertheless, since 1994 in Europe, gold has received approval as a food colorant (identified in Europe with the E-number E-175), opening the potential for leveraging the conductive attributes of gold in edible devices. To date, gold is only regulated and commercially available for food consumption in an extremely limited range

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of formats within the food market, such as micrometric and fragile gold leaves and powders with specific thicknesses and granulometries. Demonstrations exist in which commercial edible gold leaves have been successfully integrated into supercapacitors and batteries as current collectors, without the necessity of any further processing and modification,[9,10] and, similarly, edible gold leaf powder has been added into carbon-based conductive pastes to improve their conductivity.[20] Nevertheless, such shape limitation critically hinders the exploitation of gold-favorable mechanical properties and ease of metallurgical processing to create convenient form factors, suitable for fundamental electronic applications, such as solenoids and antennas. In this study, we demonstrate that it is possible to manufacture radio frequency (RF) electronic components combining the electrical conductivity properties of nonprocessed, fragile edible gold leaves with the processability and robustness inherent in some food products. In particular, numerous centuries of culinary experimentation have unequivocally illustrated the outstanding rheological properties and processability exhibited by durum wheat derivatives, such as semolina dough. These attributes, specific to pasta production and formation processes, such as drawing, lamination, and drying, originate from the synergistic interaction of water with the starch and the surrounding protein gluten mesh, of which the waste and byproducts of the agri-food sector may represent abundant sources.[21] To demonstrate this concept, we leveraged the enhanced processability of commercial Italian spaghetti to effectively template gold leaf-coated solenoidal components, resulting in inductance values ranging from 70 to 130 nH and quality factors superior to 10 for spaghetti with label no. 5 (Figure S1, Supporting Information). By interconnecting the edible inductor with an edible capacitor, composed of the same edible leaves and seaweed as dielectric, proof-of-principle edible resonant circuits operating from 180 to 280 MHz were successfully demonstrated. Such results open a path for exploitation of sustainable circuits for RF applications in sensing and control with components made of waste-derived food.

2. Results and Discussion

Figure 1 displays the procedure applied to fabricate the edible solenoids. A set of commercially available Italian spaghetti is placed in boiling water for ≈2 min to induce a second hydration process, after the first that occurs within the pasta production process. During this second hydration step, the starch granules present in the pasta, absorbing part of the water, swell, increase in volume, and gelatinize, i.e., they change structure from crystalline to gel-like, softening the spaghetti structure. Correspondently, the gluten proteins mesh that surrounds the starch grains undergoes as well a process of water intake and swelling, which expands the individual threads constituting the mesh. Meanwhile, the protein structure of gluten undergoes a denaturation process, leading to a further coagulation that increases the degree of crosslinking and strengthens the overall mesh, strongly improving its elasticity.[22] This process is crucial for providing cohesion and elasticity to pasta, enough to avoid mechanical damages or breakage during its handling in the following forming steps. Spaghetti was then coated with a commercial 24 carat gold leaf, with thickness of ≈3 μm (**Figure 2**b) and measured conductivity of ≈100 S cm^{-1}, by mechanically rolling the spaghetti onto the leaf. The gold leaf adheres to the spaghetti thanks to the gluing effect of the moist starch collected in the spaghetti's outer layer during the boiling water soaking.[23] After this coating step, while still wet and viscoelastic, the

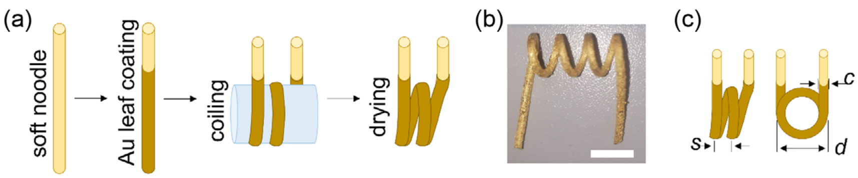

Figure 1. a) Graphic representation of the solenoid fabrication process. b) Photograph of the resulting solenoid (scale bar is 1 cm). c) Geometrical details of the solenoid, where $s = 5$ mm is the step between coils, $d = 10.35$ mm is the outer diameter, and c is around 1.85 mm the diameter of the coated noodle (for spaghetti labeled with no. 5).

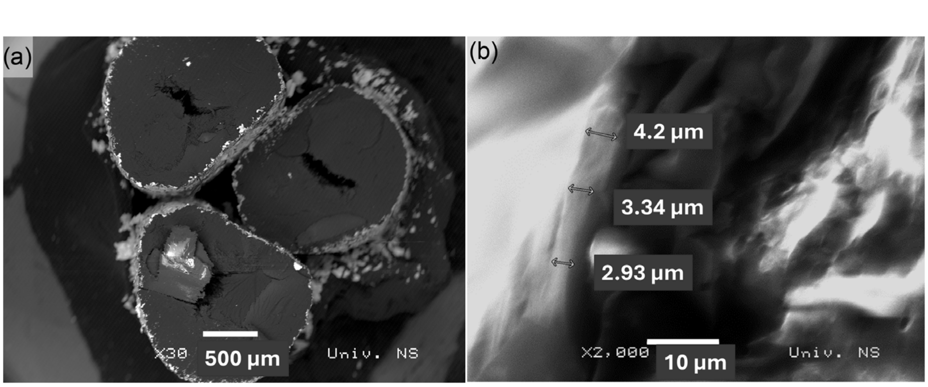

Figure 2. SEM micrographs of cooked spaghetti covered by the gold leaf, a) cross-section of spaghetti with gold layer and b) thickness of the gold layer.

spaghetti was coiled around a cylindrical object to shape the inductors, and then left to dry. Upon drying, the starch in the gold-coated spaghetti recrystallizes, making the solenoidal spaghetti stiff again. Using this procedure, solenoids with a number of coils ranging from 4 to 8, with step lengths $s = 5$ mm and $d = 10.35$ mm, respectively, were fabricated (see Figure 1c). It is worth noting that the form factors of this work are mostly limited by the size of commercial spaghetti and by the dexterity of the manual operator; we can safely assume that an automated process based on ad hoc preformed spaghetti-like structures can result in a considerable miniaturization of the coil. Importantly, no damage to the gold leaf was procured during the coil manufacturing, as exemplified by the scanning electron microscopy (SEM) images reported in Figure 2a, where the gold leaf adhering to the spaghetti surface can be observed.

The benefits of applied processing techniques compared to other methods (chemical vapor deposition, evaporators, etc.) are cost-effectiveness, wider accessibility, and enabling completely "green" process which is very important for food-based or edible electronics. The impedance of the thus formed solenoids was measured to extract the Bode plots. The Impedance Analyzer HIOKI IM7585 with sample holder IM9202 was used to measure the electrical characteristics, in the frequency range from 1 to 650 MHz. The characterization of the electrical properties of edible solenoid was performed, measuring impedance (Z), inductance (L), and quality factor (Q) as a function of frequency. The measurements were conducted on five identical samples of solenoids and the average measurement values of impedance, inductance, and quality factor were calculated and presented. We also analyzed the influence of diameter of spaghetti on quality factor, creating solenoids from three of the most often commercial pasta, labeled with no. 3 (diameter around 1.55 mm), no. 5 (diameter around 1.85 mm), and no. 7 (diameter around 2.1 mm). Increasing the pasta's diameter increases the cross-sectional area, which reduces the resistance of the coil. Since the quality factor is inversely proportional to resistance, lowering the resistance can help increase the Q-factor. **Figure 3**a shows the typical impedance Z (magnitude and phase) of the fabricated edible solenoids, which shows a characteristic 90° phase angle above 100 MHz, up to its self-resonance at ≈550 MHz, which emerges from the parasitic capacitance of the inductor. At lower frequencies, the device exhibits a resistive behavior, as evident from the phase angle reaching ≈5° at 1 MHz. This characteristic is explained by the relatively low conductivity of the gold conductor, yielding an overall DC resistance in excess of 5 Ω. How the DC electrical resistance was changed when the structure was exposed to different pH values can be seen in Table S1, Supporting Information. By adopting a conventional RLC equivalent model (see inset of Figure 3b, the fitting characteristics are reported in Figure S2, Supporting Information), we have extracted inductance values L varying with the number of coils, spanning from 70 to 130 nH; importantly, a linear trend with increasing the number of coils is obtained, as expected by ideal inductors. The parasitic capacitance C (in inset of Figure 3b) is a consequence of two effects, capacitance between each turn in solenoid structure and the geometry of the conductor, which is essentially a coiled hollow cylinder (which can be also seen in Figure 2a).

To showcase the operation of the component in a circuit, we realized proof-of-concept LC tanks by coupling the inductors to edible parallel plate capacitors (**Figure 4**a). The capacitors were fabricated in a form of edible parallel plate capacitors, whereas a dielectric layer, edible seaweed, was used with thickness of ≈0.2 mm. Square-shaped electrodes were created using edible gold in four different dimensions (5 × 5 mm, 15 × 15 mm, 25 × 25 mm, and 35 × 35 mm). The impedance analysis of such formed capacitors (reported in Supporting Information) has highlighted a purely capacitive behavior (extracted ε_r between 5 and 10) in the frequency range from 1 to 40 MHz. By applying a formula for parallel plate capacitor $C = \varepsilon_r \cdot \varepsilon_o \cdot A/d$, capacitance values increase with the surface area (A). The measured values were from 2.5 to 5.5 pF (for various dimensions of capacitors) at frequency of 20 MHz, as can be seen in Figure 4b and an increasing trend of capacitance with capacitors area was confirmed. As expected, the LC circuit demonstrated a characteristic resonant behavior with resonant frequency in the range from 180 to 280 MHz. We found that the resonance frequency is finely tuned by modifying either the inductive (Figure 3d) or the capacitive components (Figure S3, Supporting Information), as expected in ideal LC circuits.

The theoretical resonance frequency values f_0 can be extracted using the following equation

$$f_0 = \frac{1}{2\pi\sqrt{\mathrm{LC}}} \quad (1)$$

and compared to the experimental values, extracted from the impedance analysis. It can be observed that a good

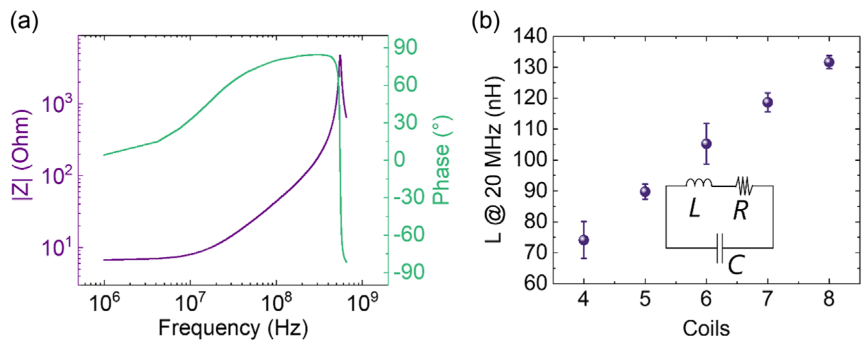

Figure 3. a) Bode plot of the impedance magnitude and phase of a 4-coil inductor. b) Inductance values for solenoids with different number of coils (turns), adopting a conventional equivalent model for data analysis (see inset).

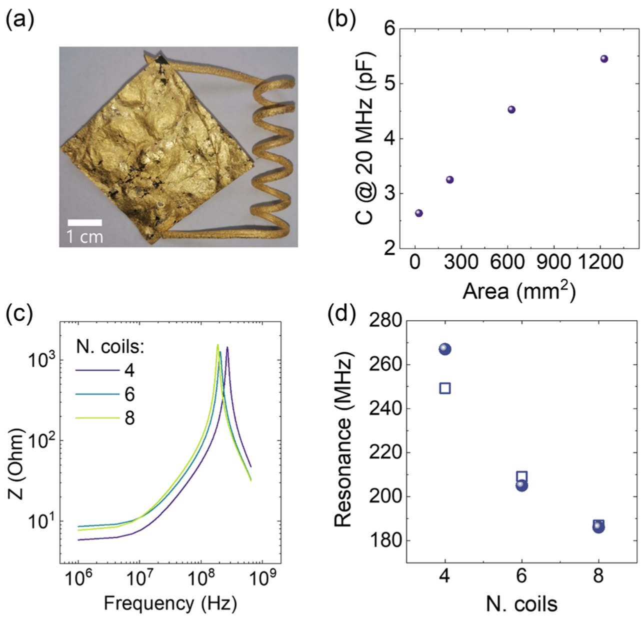

Figure 4. a) Photograph of a biodegradable LC tank circuit. b) Capacitance versus seaweed area for a parallel plate capacitor based on gold-coated seaweed. c) Impedance magnitude plots for a set of LC with variable number of coils within the inductor. d) Experimental (spheres) and theoretical (squares) resonance frequency values of the LC circuits versus the number of coils of the inductors.

correspondence exists between the experimental and the theoretical data (Figure 4d), highlighting the close-to-ideal behavior of the edible RF resonators.

The intended usage of this circuit is for applications in oral cavity to detect different biomarkers or pH or bleeding, measuring shift in the resonant frequency, which is the consequence of exposing the LC circuit to different media in our mouth. The edible gold represents recyclable material since it has a purpose only during measurement. After measurements, the coil can be removed from the oral cavity, and any standard procedure can recycle it.

3. Conclusion

In this study, we have demonstrated the possibility of realizing edible passive components such as solenoids, as required in future RF circuits. Our approach involves harnessing the chemical and physical properties inherent in food, employing processes inspired by the culinary domain, with the vision of taking advantage of food waste. To validate this concept, a series of solenoids were shaped by leveraging the successive softening and stiffening of Italian spaghetti, along with their inherent elasticity and resistance to mechanical stress. Spaghetti serves as mechanical scaffold for delicate, micrometric thick edible gold leaves, perfectly glued at the spaghetti surface by the action of moist starch, finally imparting metallic conductivity up to hundreds of $S\,cm^{-1}$ to the structure. The resultant edible solenoids exhibit inductance values that linearly correlate with the number of coils. These values range from 70 nH for 4× rounded inductors to 130 nH for 8× rounded inductors. Proof-of-principle edible resonant circuits which operate above 200 MHz and are entirely fabricated through the coupling of edible gold foils with food derivatives, were ultimately successfully demonstrated. This study lays the groundwork for a systematic exploration and utilization of strategies and materials directly derived from the culinary domain for application in the field of edible electronics. Our future study will be oriented toward analyzing properties of other edible conducting materials such as zinc and zinc-beta-carotene. Furthermore, the focus of this manuscript was to demonstrate, for the first time, the proof of feasibility in successfully manufacturing spaghetti-based solenoids as one of the inductor types. In the future, fabrication can be automatized, as well as manufacturing of other types of inductors such as square, circular, etc., using 3D bioprinters could be explored.

4. Experimental Section

Materials: The materials used in this work included: edible thin gold leaf, 24 carat, originally in dimensions of 80 × 80 mm (bought from company Muzejsko snabdevanje, Belgrade, Serbia), Barilla classic spaghetti no. 5, (no. 3 or no. 7), (bought in local store, Novi Sad, Serbia), Seaweed leaves, Sushi Nori, Miyako, Japan (bought in local store, Novi Sad, Serbia), and plastic straws (bought in local store, Novi Sad, Serbia) for rolling spaghetti around them into solenoid form.

Manufacturing of Edible Solenoids: In the first step, Barilla spaghetti was cooked in boiling water for around 2 min. After that the boiled spaghetti was rolled in the edible thin gold leaf. One edible thin gold leaf with standard dimensions of 80 × 80 mm is enough to cover completely one spaghetti, which means for creation of one inductor. The spaghetti coated with edible gold was rolled on plastic straws to achieve solenoid shape with desired number of turns. After the gold leaf application, the coated coils were blow-dried at 60 °C to eliminate excess water content between the spaghetti and the gold leaf held by capillary forces for ≈5 min. As the final step, the coils were dried at room temperature for ≈20 h, or until the coil mass was close to the combined mass of dry spaghetti and gold leaf.

Manufacturing of Edible Capacitor: Seaweed (thickness around 200 μm) was cut in the desired shape of electrodes, in our case, square shape. To provide tunability of the resonant frequency, we fabricated parallel plate capacitors with the following dimensions: 5 × 5 mm, 15 × 15 mm, 25 × 25 mm, and 35 × 35 mm. The seaweed was slightly created to be wet (with four drops of tap water) and consequently sticky. After that, prepared gold leaf was deposited on the seaweed from one side and after that from another side. It is important to take care that there is no short circuit between these parallel plate gold electrodes, which was achieved by selecting dimensions of seaweed to be slightly larger that dimensions of edible gold electrodes.

Electrical Characterization: The Impedance Analyzer HIOKI IM7585 with sample holder IM9202 was used to measure the electrical characteristics, in the frequency range from 1 to 650 MHz. The following signal levels were used for measurements: power: −40.0 to +1.0 dBm, voltage: 4 mV to 502 mV rms, and current: 0.09 mA to 10.04 mA rms. Three measurements were repeated for each sample and average values are presented.

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.

Author Contributions

Milan R. Radovanović: data curation (equal); formal analysis (equal); methodology (lead). **Željko Popović**: formal analysis (lead); validation (equal). **Sanja Kojić**: investigation (equal); supervision (supporting). **Danica Piper**: investigation (equal). **Alessandro Luzio**: conceptualization (equal); visualization (equal); writing—original draft (equal). **Giorgio Ernesto Bonacchini**: methodology (equal); validation (lead); writing—original draft (equal). **Mario Caironi**: conceptualization (equal); resources (equal); writing—original draft (equal). **Goran M. Stojanović**: conceptualization (equal); resources (equal); writing—original draft (lead).

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

edible electronics, edible solenoids, green electronics, inductance, quality factors

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[1] D. Perkins, M.-N. Drisse, T. Nxele, P. Sly, *Ann. Glob. Health* **2014**, *80*, 286.
[2] O. A. Ogunseitan, J. M. Schoenung, J. Lincoln, B. H. Nguyen, K. Strauss, K. Frost, E. Schwartz, H. He, M. Ibrahim, https://www.theworldcounts.com/stories/household-hazardouswaste-statistics (accessed: November 2024).
[3] A. S. Sharova, F. Melloni, G. Lanzani, C. J. Bettinger, M. Caironi, *Adv. Mater. Technol.* **2021**, *6*, 2000757.
[4] V. F. Annese, G. Coco, V. Galli, P. Cataldi, M. Caironi, in *Proc. 2023 IEEE Int. Conf. on Flexible and Printable Sensors and Systems (FLEPS)*, IEEE, Piscataway, NJ **2023**.
[5] F. Safety, https://food.ec.europa.eu/safety/food-waste_en (accessed: November 2024).
[6] a) H. Tao, M. A. Brenckle, M. Yang, J. Zhang, M. Liu, S. M. Siebert, R. D. Averitt, M. S. Mannoor, M. C. McAlpine, J. A. Rogers, D. L. Kaplan, F. G. Omenetto, *Adv. Mater.* **2012**, *24*, 1067; b) G. Stojanovic, M. Pojić, S. Kojic, A. Mišan, D. Vasiljevic, *Appl. Phys. A* **2019**, *125*, 2881; c) M. A. Daniele, A. J. Knight, S. A. Roberts, K. Radom, J. S. Erickson, *Adv. Mater.* **2015**, *27*, 1600.
[7] a) Y. Wu, D. Ye, Y. Shan, S. He, Z. Su, J. Liang, J. Zheng, Z. Yang, H. Yang, W. Xu, H. Jiang, *Adv. Mater. Technol.* **2020**, *5*, 2000100; b) B. Le Borgne, B.-Y. Chung, M. O. Tas, S. G. King, M. Harnois, R. A. Sporea, *Electronics* **2019**, *8*, 699.
[8] S. Mao, B. Sun, Y. Tian, W. Mao, S. Zhu, Y. Ni, H. Wang, Y. Zhao, Y. Chen, *New J. Chem.* **2019**, *43*, 2433.
[9] I. Ilic, V. Galli, L. Lamanna, P. Cataldi, L. Pasquale, V. Annese, A. Athanassiou, M. Caironi, *Adv. Mater.* **2023**, *35*, 2211400.
[10] V. F. Annese, V. Galli, G. Coco, M. Caironi, presented at *2023 9th Int. Workshop on Advances in Sensors and Interfaces (IWASI)*, Monopoli, Bari, Italy, June **2023**.
[11] W. Xu, H. Yang, W. Zeng, T. Houghton, X. Wang, R. Murthy, H. Kim, Y. Lin, M. Mignolet, H. Duan, H. Yu, M. Slepian, H. Jiang, *Adv. Mater. Technol.* **2017**, *2*, 1700181.
[12] K. Fukada, T. Tajima, M. Seyama, *ACS Appl. Mater. Interfaces* **2022**, *14*, 44697.
[13] a) I. Ilic, L. Lamanna, D. Cortecchia, P. Cataldi, A. Luzio, M. Caironi, *ACS Sensors* **2022**, *7*, 2995; b) I. Mallov, F. Jeeva, T. Caputo, *Biotechnology* **2022**, *97*, 830; c) I. Dudnyk, E.-R. Janeček, J. Vaucher-Joset, F. J. S. Stellacci, *Sens. Actuators, B* **2018**, *259*, 1108.
[14] S. Tian, Y. Xing, Y. Long, H. Guo, S. Xu, Y. Ma, C. Wen, Q. Li, X. Liu, M. Zhang, *Interfaces* **2022**, *14*, 5122.
[15] B. Kuswandi, N. P. Asih, D. K. Pratoko, N. Kristiningrum, T. Moradi, *Science* **2020**, *33*, 321.
[16] G. Matzeu, G. R. S. Naveh, S. Agarwal, J. A. Roshko, N. A. Ostrovsky-Snider, B. S. Napier, F. G. Omenetto, *Adv. Sci.* **2021**, *8*, 2003416.
[17] H. M. Ur Rehman, P. A. Sathya, M. Rehman, M. Khan, S.-J. Kimb, W. Y. Kim, *Sustainable Mater. Technol.* **2023**, *36*, e00596.
[18] P. Cataldi, L. Lamanna, C. Bertei, F. Arena, P. Rossi, M. Liu, F. Fonzo, D. Papageorgiou, A. Luzio, M. Caironi, *Adv. Funct. Mater.* **2022**, *32*, 2113417.
[19] Y. Zhang, Q. Tang, J. Zhou, C. Zhao, S. Wang, *Engineering* **2023**, *10*, 191.
[20] K. Fukada, T. Tajima, K. Hayashi, *ACS Appl. Mater.* **2022**, *4*, 6087.
[21] a) D. Kringel, Á. Dias, E. Zavareze, E. Gandra, *Starch/Starke* **2019**, *72*, 1900200; b) D. Lesmana, Y. M. Vianney, Y. A. Goenawan, K. Natalie, J. Sukweenadhi, G. Buschle-Diller, Y. P. Mukti, C. M. Erawati, S. Purwanto, *Technology* **2021**, *2*, 3; c) L. Principato, L. Ruini, M. Guidi, L. Secondi, *Resour., Conserv. Recycl.* **2019**, *144*, 82.
[22] a) M. Wagner, M.-H. Morel, J. Bonicel, A. Cuq, *J. Agric. Food Chem.* **2011**, *59*, 3146; b) H. Liu, J. Qi, K. Hayakawa, *J. Food Sci.* **1997**, *62*, 813; c) N. Edwards, M. Izydorczyk, J. Dexter, C. Biliaderis, *Cereal Chem.* **1993**, *70*, 122; d) S. Cafieri, M. Mastromatteo, S. Chillo, M. A. Nobile, *J. Food Eng.* **2010**, *100*, 336; e) S. Cafieri, S. Chillo, M. Mastromatteo, N. Suriano, M. A. Del Nobile, *J. Cereal Sci.* **2008**, *48*, 857.
[23] S. Perez, P. Baldwin, D. J. Gallant, *Starch*, Elsevier Inc., Amsterdam, Netherlands **2009**, p. 149.

