Edible Electronics and Robofood: A Move Towards Sensors for Edible Robots and Robotic Food

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Abstract — Sustainability remains an underdeveloped aspect when designing an electronic device, even though technology is more pervasive in our society. As such, a paradigm change is needed toward the use of more environmentally friendly materials and processes. Edible electronics proposes integrating food-grade materials into more complex system such as robots, thus contributing to reducing e-waste accumulation. Besides sustainability, biocompatibility, and biodegradability, the use of food-grade materials in electronics has unprecedented advantages including minimal toxicity levels, especially in case of ingestion. Thus, edible electronics and robotics opens unprecedented scenarios: in the future rescue drones could integrate edible components, effectively increasing the food payload of the mission; robotic food could be employed as drug delivery vectors for wild animals; ultimately, miniaturized edible robots could enable novel diagnostic tools that can be digested by the body after performing a specific test. The EUfunded "Robofood" project works towards this vision.

In this frame, we present a versatile fully edible electrically conductive ink for edible electronics and robotics. The ink is based on activated carbon - an organic edible electronic conductor with a daily intake up to three orders of magnitude higher than metals - and is formulated to be deposited by spray coating. Successful deposition on different edible substrates was obtained. As a proof-of-principle for the use of this material in edible robotics, a first application for bending sensing is herein reported. The coating was interfaced with a standard microcontroller and data was recorded during finger bending. The materials and methods developed in this work have a high degree of versatility and could be applied to other scenarios. We believe that the vision supported by this project has the potential to open the way for novel edible technologies for applications such as medicine and food quality monitoring among others.

Keywords — Edible electronics, green electronics, green sensors, edible sensors, conductive coatings.

I. INTRODUCTION

Edible electronics is a recent research area which aims at delivering electronic devices using food-derived materials. Depending on how they will be used in the final scenario, edible electronic systems will likely fall into one of two categories: those that operate inside the gastrointestinal (GI) tract and those that execute their tasks outside the body [1]. The former aims at delivering novel technologies for GI monitoring by developing digestible devices where each constituent material is inherently safe to eat. Although many ingestible devices have been documented [2-4], novel digestible materials can open interesting scenarios for next-generation ingestible applications. The latter instead expands its impact to the food business, industries, and robotics where devices can be used directly in contact with food as there is no

contamination risk. Food monitoring, together with models for shelf-life prediction [5, 6], can be useful in reducing food waste [7]. Regardless of the application scenario, edible devices have the potential to represent a more sustainable alternative than traditional components. These devices can benefit from infrastructures already implemented in the food industry for manufacturing, storage, transport, and – above all – disposal, therefore they could be introduced into society with minimal investments. As such, it is possible to dispose of edible electronics in several environmentally friendly ways, for instance by using them to produce compost, biogas, or animal feed.

Preliminary edible components have already been documented in the literature [8–17]. Food-derived materials like silk, pea protein, pullulan and cellulose have been studied as promising flexible substrates for electronics and sensing by printing on their surface resistors, capacitors, inductors with gold/silver ink [14-16]. Partly edible circuits have been developed using honey-gated transistors, in which the active semiconductor is biocompatible [18]. There is ongoing research on edible semiconductors [19, 20] but no fully edible circuits with high degree of complexity have been fabricated so far. RC and RLC filters have been fabricated using either activated carbon and egg white [21] or carbonized cotton, and gold [17]. Other remarkable starch, edible implementations include memristors (using casein or garlic) [22, 23], battery [24], and supercapacitor [25]. Edible sensors are also gradually emerging. Edible sensors have first been explored for use with direct contact with food to detect freshness or spoilage [8-12]. They either use food-based biocatalysts to detect enzyme activity in saliva [8], or exploit the reaction to pH, amines and O_2 of anthocyanin (red cabbage) or genipin [9-12]. A glutinous rice gel has also been used both as strain and temperature sensor, and to detect enzymes in the GI tract [13]. A piezoelectric sensor using a film of broccoli powder and gelatin capable of detecting low frequency sound has also been shown [17]. Pressure sensing using - for instance - a gelatin/glycerol hydrogel has also been demonstrated [25].

Several materials with promising applications in edible electronics have been identified [1, 17]. Remarkably, a recurrent interesting material is activated carbon [8, 24 - 26]. Activated carbon is a form of vegetable carbon approved as edible in the European Union (EU) as E 153. It is made from carbon-rich materials including bamboo, coconut husk, and wood, and it is used in many applications as colourant agent, food additive, dietary supplement, and water filtration. The European Food Safety Authority (EFSA) does not express concern on the maximum daily intake of hundreds of mg/kg reported in food industry [27].

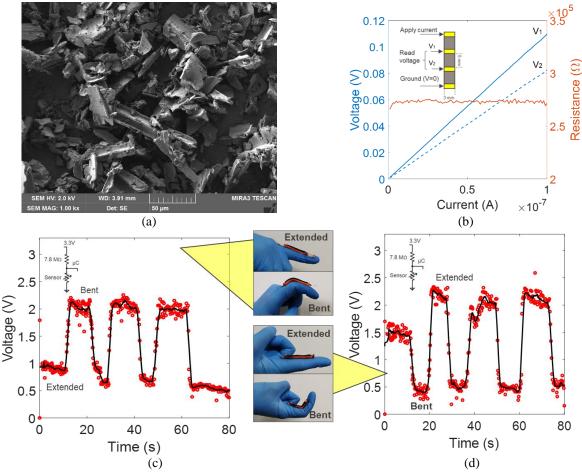


Fig. 1. (a) Morphology of the activated carbon-based edible conductive coating. (b) Typical response for the measurement of the resistance of the material using a four-probe configuration. Inset: measurement setup. (c) Data obtained by connecting the conductive coating deposited onto a viscoelastic edible substrate to a standard microcontroller during finger bending (stretching) (d) Similar experiment as before but during finger bending in compression.

This compares favourably to other metals approved as edible from EFSA - such as gold (E 175) or silver (E 174) - which have a maximum daily intake in the order of μ g/kg of body weight. Activated carbon is an electrical conductor with large surface area and its conductivity depends on many factors including source material and activation process [28] ranging from 0.001 to 1.940 S·cm⁻¹ [1]. As such, activated carbon has been widely used in electronics for supercapacitors [25, 29], functionalised electrodes [30] and flexible [31] applications among others.

Here we present preliminary data on an edible electrically conductive ink based on activated carbon. The ink is produced using a straightforward process using an edible binder to provide adhesion with the substrate and is specifically formulated for spray coating. To demonstrate its use for sensing, we provide a proof-of-concept application. After the successful deposition of the coating onto an edible stretchable material, we interface the material with a standard microcontroller and we show that the coating is successful in sensing the bending movement of a finger. The material and processes herein developed can potentially be applied to edible robotic systems for several application including structural health monitoring, tactile, strain and pressure sensing.

II. PRELIMINARY RESULTS AND DISCUSSION

To prepare the edible electrically conductive ink, commercial gelatin-based candy was first dissolved in water in a concentration of 80 mg/ml. Ethanol was then added to the solution in a 7:1 volume ratio. Activated carbon (Supelco 05105 from Sigma Aldrich) was then introduced in the solution with a 1:1 weight ratio with the binder. The solution was then stirred using magnetic stirring at room temperature for 30 min. Afterwards, the solution was tip sonicated for 15 minutes. The ink was deposited using spray coating. The ink volume sprayed was 6 ml over an area of 75 mm x 25 mm. The pressure used to spray the ink was kept constant to 2 bar. The target substrate was left to dry for at least two hours at room temperature after spray coating. The deposition was successfully achieved over a variety of substrates, including traditional (glass, Polydimethylsiloxane – PDMS, cellulose) and edible (ethyl cellulose, edible wafer paper, gelatin-based film) substrates. An SEM micrograph of the coating deposited onto PDMS is reported in Fig. 1a.

The sheet resistance of the coating was quantified using PDMS as the target substrate. 10 g of PDMS (SYLGARD 184 Silicone Elastomer) was prepared by mixing the polymer in 10:1 weight ratio with the curing agent, poured into a standard 100 mm glass petri dish, degassed into a vacuum chamber, and cured for 2 h at 70 °C. Cured PDMS was then released from the petri dish and cut in smaller target substrates. Before the coating deposition, the PDMS target substrates were exposed

to oxygen plasma for 1 min at 20 W to improve the wettability [32]. The coating was then deposited and dried as described in the above-mentioned procedure. To measure the sample resistance, four equally spaced electrical contacts were created by applying silver paint. The electrical measurements were performed using a semiconductor parameter analyser (SPA Keysight B1500A) connected to a probe station in air. Data shows an ohmic behaviour suggesting that the ratio between activated carbon and binder is enough to overcome the percolation threshold and provide a conductive film (Fig. 1b). The sheet resistance was obtained as average and standard deviation over three different samples and was quantified to be 247.0 ± 61.8 kΩ/sq.

With the aim of demonstrating the potential of the conductive coating in edible robotics, the coating was deposited onto an edible viscoelastic gelatin-based substrate and was used to detect finger bending. The sample was attached to a nitrile glove. Electrical contacts were obtained by using silver paint and copper tape and were positioned far from the area of the sample undergoing bending. The sample was then interfaced to a commercial pull-up 7.8 M Ω resistor in a voltage divider configuration and a voltage of 3.3 V was supplied to the circuit. The output of the voltage divider was connected to the STM32 Nucleo-64 microcontroller board (STMicroelectronics) equipped with a CPU ARM Cortex M4F. Data was digitised using the embedded 12-bit ADC with a sampling time of 196.3 ± 72.0 ms. The microcontroller was interfaced with a laptop (HP EliteBook x360 830 G6) via USB. Data was collected and stored in real-time using a custom Matlab script. Data shows that the sample could detect multiple events of finger bending in both directions (stretching and compression). When finger bending created stretching (Fig. 1c), the resistance of the sample increased therefore the output of the voltage divider was also increased. Differently, when finger bending created a compression of the sample (Fig. 1d), the resistance of the sample decreased, which in turns created a reduction of the output voltage. In both cases, during the hold time, the sample maintained its state apart from a slight output drift probably due to the viscoelastic behaviour of the substrate. This first proof-of-principle shows that these materials hold a great potential for robotics application. However, additional characterisation of the coating and substrate will be needed to enable the use of these materials and methods for strain and bending sensing. Future works include the optimisation of the ink to further reduce the sheet resistance, the characterisation of the rheological properties of the ink and the integration of the component into a more complex system toward the demonstration of in-vivo applications.

III. CONCLUSIONS

Edible electronics and its application in edible robotics have the potential to revolutionise the way we think about technology. Besides being sustainable, devices manufactured using food-derived materials open novel scenarios in many application fields including food quality monitoring, healthcare and well-being. Nonetheless, edible electronics remains an emerging research field where novel materials and processes are needed for the implementation of complex systems.

Here we present a first demonstration of a sprayable edible conductive ink based on activated carbon. The ink can find application in printed electronics to produce electrodes, contacts, or resistors. The technique herein describe is also scalable to larger areas and can be used on a variety of substrates including edible ones. As a proof of concept, we use the conductive ink deposited onto an edible viscoelastic substrate to record real-time data from finger bending using a standard microcontroller board. As such, we demonstrate a successful connection between edible components and traditional electronics.

The data illustrated in this work is preliminary and more characterisation is needed to unlock these materials and methods in more complex systems such as robots. However, the vision of the EU-funded "Robofood" project and shared in this work might have dramatic consequences in our society. We foresee that in the future edible robots and robotic food will be available in real-life scenarios completing tasks specifically designed to benefit from their advantages.

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REFERENCES

- Sharova, Alina S., Filippo Melloni, Guglielmo Lanzani, Christopher J. Bettinger, and Mario Caironi. "Edible electronics: The vision and the challenge." Advanced Materials Technologies 6, no. 2 (2021): 2000757.
- [2] Melino, G., Accarino, C., Riehle, M., Potter, M., Fineron, P., Annese, V.F., Grant, J.P., Al-Rawhani, M.A., Beeley, J., Carranza, I.E. and Cumming, D.R., 2020. Capsule endoscopy compatible fluorescence imager demonstrated using bowel cancer tumours. IEEE Sensors Journal, 20(17), pp.9763-9771.
- [3] V. F. Annese, D. De Venuto, C. Martin, and D. R. S. Cumming, "Biodegradable pressure sensor healthcarecare," 2014 21st IEEE International Conference on Electronics, Circuits and Systems (ICECS), Dec. 2014, doi: https://doi.org/10.1109/icecs.2014.7050056.
- [4] V. F. Annese, C. Martin, D. R. S. Cumming, and D. De Venuto, "Wireless capsule technology: Remotely powered improved highsensitive barometric endoradiosonde," 2016 IEEE International Symposium on Circuits and Systems (ISCAS), May 2016, doi: https://doi.org/10.1109/iscas.2016.7527504.
- [5] G. E. Biccario, V. F. Annese, S. Cipriani, and D. De Venuto, "WSNbased near Real-time Environmental Monitoring for Shelf Life Prediction through Data Processing to Improve Food Safety and Certification," Proceedings of the 11th International Conference on Informatics in Control, Automation and Robotics, 2014, doi: https://doi.org/10.5220/0005102407770782.
- [6] V. F. Annese and D. De Venuto, "On-line shelf-life prediction in perishable goods chain through the integration of WSN technology with a 1st order kinetic model," 2015 IEEE 15th International Conference on Environment and Electrical Engineering (EEEIC), Jun. 2015, doi: https://doi.org/10.1109/eeeic.2015.7165232.
- [7] "Food Waste," Food Safety, 2022. https://food.ec.europa.eu/safety/food-waste_en (accessed Mar. 23, 2023).
- [8] J. Kim, I. Jeerapan, B. Ciui, M. C. Hartel, A. Martin, and J. Wang, "Edible Electrochemistry: Food Materials Based Electrochemical Sensors," Adv Healthe Mater, vol. 6, no. 22, Nov. 2017, doi: 10.1002/adhm.201700770.
- [9] I. K. Ilic, L. Lamanna, D. Cortecchia, P. Cataldi, A. Luzio, and M. Caironi, "Self-Powered Edible Defrosting Sensor," ACS Sens, vol. 7, no. 10, pp. 2995–3005, Oct. 2022, doi: 10.1021/acssensors.2c01280.
- [10] I. Dudnyk, E. R. Janeček, J. Vaucher-Joset, and F. Stellacci, "Edible sensors for meat and seafood freshness," Sens Actuators B Chem, vol. 259, pp. 1108–1112, Apr. 2018, doi: 10.1016/j.snb.2017.12.057.

- [11] B. Kuswandi, N. P. N. Asih, D. K. Pratoko, N. Kristiningrum, and M. Moradi, "Edible pH sensor based on immobilized red cabbage anthocyanins into bacterial cellulose membrane for intelligent food packaging," Packaging Technology and Science, vol. 33, no. 8, pp. 321–332, Aug. 2020, doi: 10.1002/pts.2507.
- [12] I. Mallov, F. Jeeva, and C. B. Caputo, "An edible genipin-based sensor for biogenic amine detection," Journal of Chemical Technology and Biotechnology, vol. 97, no. 4, pp. 830–836, Apr. 2022, doi: 10.1002/jctb.6556.
- [13] S. Tian et al., "A Degradable-Renewable Ionic Skin Based on Edible Glutinous Rice Gel," ACS Appl Mater Interfaces, vol. 14, no. 4, pp. 5122–5133, Feb. 2022, doi: 10.1021/acsami.1c24352.
- [14] H. Tao et al., "Silk-based conformal, adhesive, edible food sensors," Advanced Materials, vol. 24, no. 8, pp. 1067–1072, Feb. 2012, doi: 10.1002/adma.201103814.
- [15] G. Stojanović, M. Pojić, S. Kojić, A. Mišan, and D. Vasiljević, "Mechanical properties of edible biofilm as a substrate for printed electronics," Appl Phys A Mater Sci Process, vol. 125, no. 8, Aug. 2019, doi: 10.1007/s00339-019-2881-5.
- [16] M. A. Daniele, A. J. Knight, S. A. Roberts, K. Radom, and J. S. Erickson, "Sweet substrate: A polysaccharide nanocomposite for conformal electronic decals," Advanced Materials, vol. 27, no. 9, pp. 1600–1606, Mar. 2015, doi: 10.1002/adma.201404445.
- [17] Wu, Yingzhu, Dongdong Ye, Yingfa Shan, Shuohai He, Ziyue Su, Jiahao Liang, Jinren Zheng et al. "Edible and nutritive electronics: materials, fabrications, components, and applications." Advanced Materials Technologies 5, no. 10 (2020): 2000100.
- [18] A. S. Sharova and M. Caironi, "Sweet Electronics: Honey-Gated Complementary Organic Transistors and Circuits Operating in Air," Advanced Materials, vol. 33, no. 40, Oct. 2021, doi: 10.1002/adma.202103183.
- [19] E. D. Głowacki, G. Voss, L. Leonat, M. Irimia-Vladu, S. Bauer, and N. S. Sariciftci, "Indigo and Tyrian purple From ancient natural dyes to modern organic semiconductors," Israel Journal of Chemistry, vol. 52, no. 6. pp. 540–551, Jun. 2012. doi: 10.1002/ijch.201100130.
- [20] M. Irimia-Vladu et al., "Biocompatible and biodegradable materials for organic field-effect transistors," Adv Funct Mater, vol. 20, no. 23, pp. 4069–4076, Dec. 2010, doi: 10.1002/adfm.201001031.
- [21] B. le Borgne, B. Y. Chung, M. O. Tas, S. G. King, M. Harnois, and R. A. Sporea, "Eco-friendly materials for daily-life inexpensive printed passive devices: Towards 'do-it-yourself' electronics," Electronics (Switzerland), vol. 8, no. 6, Jun. 2019, doi: 10.3390/electronics8060699.

- [22] M. Saha, S. M. Nawaz, B. K. Keshari, and A. Mallik, "Natural-Casein-Based Biomemristor with Pinched Current-Voltage Characteristics," ACS Appl Bio Mater, vol. 5, no. 2, pp. 833–840, Feb. 2022, doi: 10.1021/acsabm.1c01188.
- [23] S. Mao et al., "PH-Modulated memristive behavior based on an edible garlic-constructed bio-electronic device," New Journal of Chemistry, vol. 43, no. 24, pp. 9634–9640, 2019, doi: 10.1039/c9nj02433f.
- [24] Ilic, Ivan K., Valerio Galli, Leonardo Lamanna, Pietro Cataldi, Lea Pasquale, Valerio F. Annese, Athanassia Athanassiou, and Mario Caironi. "An Edible Rechargeable Battery." Advanced Materials (2023): 2211400. doi: https://doi.org/10.1002/adma.202211400.
- [25] V.F. Annese, V. Galli, G. Coco, M. Caironi. "Eat, Test, Digest: Towards Diagnostic Food for Next Generation Gastrointestinal Tract Monitoring". 2023 IEEE 9th International Workshop on Advances in Sensors and Interfaces (IWASI), Monopoli, Italy, 2023. In-print.
- [26] P. Cataldi et al., "An Electrically Conductive Oleogel Paste for Edible Electronics," Advanced Functional Materials, vol. 32, no. 23, p. 2113417, Feb. 2022, doi: https://doi.org/10.1002/adfm.202113417.
- [27] "European Food Safety Authority," European Food Safety Authority, Jul. 13, 2021. https://www.efsa.europa.eu/en (accessed Mar. 23, 2023).
- [28] T. Adinaveen, J. J. Vijaya, and L. J. Kennedy, "Comparative Study of Electrical Conductivity on Activated Carbons Prepared from Various Cellulose Materials," Arabian Journal for Science and Engineering, vol. 41, no. 1, pp. 55–65, Nov. 2014, doi: https://doi.org/10.1007/s13369-014-1516-6.
- [29] S. Faraji and F. N. Ani, "The development supercapacitor from activated carbon by electroless plating—A review," Renewable and Sustainable Energy Reviews, vol. 42, pp. 823–834, Feb. 2015, doi: https://doi.org/10.1016/j.rser.2014.10.068.
- [30] Şavk, Aysun, Hediye Aydın, Kemal Cellat, and Fatih Şen. "A novel high performance non-enzymatic electrochemical glucose biosensor based on activated carbon-supported Pt-Ni nanocomposite." Journal of Molecular Liquids 300 (2020): 112355.
- [31] C. Wang, K. Xia, H. Wang, X. Liang, Z. Yin, and Y. Zhang, "Advanced Carbon for Flexible and Wearable Electronics," *Advanced Materials*, vol. 31, no. 9, p. 1801072, Oct. 2018, doi: https://doi.org/10.1002/adma.201801072.
- [32] Annese, Valerio F., Samadhan B. Patil, Chunxiao Hu, Christos Giagkoulovits, Mohammed A. Al-Rawhani, James Grant, Martin Macleod et al. "A monolithic single-chip point-of-care platform for metabolomic prostate cancer detection." Microsystems & Nanoengineering 7, no. 1 (2021): 21.