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Edible electronics

Electrical circuits developed on cookie dough-based substrate and their sensing applications

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Abstract—Edible electronics present a blossoming path to a greener and eco-friendly future for electronics, whilst being biocompatible with living beings. With this characteristic, edible electronics has been recently proposed for the design and fabrication of edible and digestible sensors. More precisely, it has become a strong and sustainable candidate for continuous and *in vivo* monitoring and diagnosis of patients. Yet, the field is in constant search for new functional materials satisfying the stringent and contrasting requirements of safe edibility and performing electronics. With this in mind, a novel edible substrate, based entirely on cookie dough is presented in this letter. An extensive mechanical and electrical characterization of the edible substrate is provided, aside from a clear step-by-step guide for its fabrication. Additionally, to prove the use of the cookie-dough substrate for food-based electronics, we demonstrate a voltage divider and a resonant circuit fabricated on it. Tests have been conducted in dry and wet conditions, simulating intraoral environment. Sensing capabilities have been also investigated, with variations of temperature and pH. These findings push the boundaries of edible electronics, enabling a growing community of researchers to utilize the proposed substrate and circuits in a broad range of sensor technologies and applications.

Index Terms—edible electronics, cookie-dough, resonant circuit, mechanical characterization, electrical characterization

I. INTRODUCTION

Edible electronics is at the forefront of innovation and research, within an already blossoming field of green electronics [1]. Green electronics, which utilizes biocompatible, biodegradable and non-toxic materials to achieve what classic electronics already does well, has been one of the main focal points of research since the ever-growing threat of global warming [2] and exponential increase in electronic waste [3] have become apparent. What edible electronics adds to the equation is the element of complete safety, aside from biodegradability, biocompatibility and non-toxicity, as it can be entirely digested in the body of a living being [1, 4].

With the evolution of material science, many previously untouched resources have been utilized in the field of electronics. Edible electronic and ionic conductors, as well as edible dielectrics can be easily found in food, however semiconducting materials are not as widespread as their previously mentioned counterparts [1]. Indigo [5], copper phthalocyanine [6] and β-Carotene are some of the ingestible and edible semiconducting materials that have potential use in development of edible active electronic components. Furthermore, these materials have already been integrated and tested in fields such as health monitoring, ingestible sensorics, as well as personalized nutrition. For example, in the domain of health monitoring, edible sensors have been developed. Moreover, edible pills have been developed for wireless in-body monitoring [4]. On the other hand, edible electronics has been used in ripeness, spoilage and food-freshness sensors, as a sustainable alternative to conventional packaging employing hard electronic components. These edible sensors can in principle be eaten alongside the foods, the freshness of which they monitor [7, 8]. The utilization of food-based substrates presents a fascinating avenue in the realm of electronic components.

Cookie dough possesses several attributes that make it an appealing choice for a potential substrate. Its pliable nature allows for easy shaping and molding, easily becoming a surface for printed electronics [9-11]. This flexibility facilitates the integration of components into unconventional shapes and forms, enabling the creation of customized electronics with enhanced functionality [10]. Moreover, cookie dough provides dielectric properties, effectively safeguarding electronic circuitry from external elements and electrical interference [12]. The abundance and affordability of ingredients used in cookie dough production contributes to its attractiveness as a substrate material [13]. Unlike conventional substrates that may require specialized manufacturing processes, such as clean rooms, as well as expensive materials, cookie dough can be readily prepared using day-to-day ingredients found in kitchen pantries [13, 14]. Aside from that, cookies and biscuits have long shelf life [14]. This accessibility not only reduces production costs but also promotes sustainability by repurposing food materials that might otherwise go

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to waste [15].

In this paper, we present a novel use for cookie dough, in the field of edible electronics. The substrate is fully mechanically and electrically characterized, in laboratory conditions, as well as in conditions with varying temperature. Moreover, the substrate is used for the creation of proof-of-principle edible electronic circuits and sensors. More precisely, we have created a resonant inductive-capacitive (LC) circuit and a voltage divider, which are fully edible, and we validated sensor applications of the proposed circuits.

II. MATERIALS AND METHODS

A. Substrate fabrication

For the preparation of the cookie dough-based substrate, three substances were used: finely crumbled cookies, fine corn starch and deionized water (DI). Ground Plazma, soft biscuits (manufacturer Bambi A.D) were used as the crumbled cookie base, as they are the most popular and commonly used biscuit in Serbia, for all generations. Moreover, fine corn starch Gustin (manufacturer Dr. Oetker) was used, alongside DI water as the binding material. For the fabrication of one substrate, with average size of 10 cm × 10 cm, with a thickness of 4 mm, 20 g of corn starch and 40 g of DI water were mixed, and left for 5 minutes to settle. Following that, 20 g of finely crumbled cookies were added to the mass, and everything was steam cooked until the mass bound together, which takes approximately 2 minutes. Furthermore, the sample under preparation was put in a sintering oven, on top of baking paper, with the goal of evenly distributing the cooking mass. The sintering oven was turned on at 150 °C for 1h and 30 minutes. Finally, it was left in laboratory settings to cool down. After fabrication the substrate was mechanically and electrically characterized.

B. Mechanical and electrical characterization

For mechanical characterization, wetting was tested on the drop shape analyzer (DSA25, manufacturer KRÜSS Optronic GmbH). This is important to analyze the wetting performances of the proposed edible substrate in electronics. Furthermore, a tensile testing machine (model 34SC-2, manufacturer Instron) was used for compression and three-point measurements. This was done to see how well this substrate compares to conventional, commercially available ones. Electrical characterization consisted in impedance spectroscopy (Hioki IM3590) tests, when the substrate was dry and after it was submerged in DI water.

C. Implementation and testing of an edible voltage divider circuit

Firstly, a voltage divider circuit was made as a proof-of-concept for the use of the presented cookie dough-based substrate in electronics. For conductive lines, edible gold flakes were deposited on this edible substrate. Aside from that, edible resistors were made out of activated carbon-based paste. The paste was fabricated using a ratio of 1:4 of beeswax and sunflower oil [16]. Activated carbon was then mixed with the substance to achieve 30 % concentration. The sunflower oil was mixed with beeswax on a magnetic stirrer for 15 minutes, then gradually activated carbon is added. Finally, the mass is molded to a certain length, which corresponds to the desired electrical resistance. The circuit was tested by adding a light emitting diode in parallel to one resistor, and the other side was connected to a voltage source. For the voltage source SPD3303C (manufacturer SIGLENT) was applied.

D. Implementation and testing sensing capabilities of an edible LC circuit

A resonant circuit was made out of the edible substrate. An inductor was made out of spaghetti, slightly cooked, then coated with gold and left to dry. Before the drying process, the gold coated spaghetti was bent into a solenoid shape to form an inductor (L). For the realization of a capacitor, the substrate itself, with its dielectric properties, was utilized. On both sides of the substrate, edible gold flakes were added in a 4 cm × 4 cm area to form the electrodes of the parallel plate capacitor (C). With the use of activated carbon paste, the capacitor was connected to the inductor. Furthermore, an impedance analyzer IM7585 (manufacturer HIOK) was used to measure the resonant frequency of the LC circuit (in the frequency range between 1 MHz and 200 MHz). Moreover, all measurements were repeated at different temperatures of 25 °C, 30 °C, 35 °C, 40 °C and 45 °C. This was achieved using a hotplate (manufacturer COLO LabExperts) and a thermovision camera (manufacturer HIKMICRO). Additionally, the edible LC circuit was wet with DI water, artificial saliva, pH 6 and pH buffer solution of pH 6, with the goal of studying the effect of different liquids on the performance of the edible LC circuit. Finally, pH sensing capabilities were investigated, with pH buffer solutions ranging between 6 to 8, as those are typical for intraoral environments.

III. RESULTS AND DISCUSSION

A. Substrate fabrication and characterization

Fig. 1 shows the fabricated cookie-dough edible substrate. All of the substrates, designated for mechanical and electrical characterization have been cut to fit a 5 cm × 5 cm square shape.

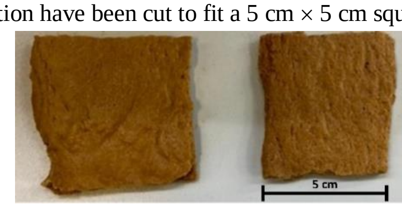

Fig. 1. Examples of fabricated and cut substrates.

Drop shape analysis was done with water to analyze the wetting characteristics of the substrates. Five samples were used to calculate the average wetting angle of the substrate, which was 56.27°, as well as the standard deviation of the samples, corresponding to 2.77°.

With the use of the tension testing system, compression and three-point measurements, were executed for each of the 3 different samples. The compression test yielded relatively uniform results, with the average force of compression being 25.92 N, with a sample standard deviation of 3.56 N. Similarly, the three-point measurement test gave results which averaged to 15.91 N, with a sample standard deviation of 2.29 N. These values are lower than those of substrates conventionally used in printed circuit board (PCB) technology, therefore cannot be exposed to big forces, but that is not an issue in the edible electronics concept.

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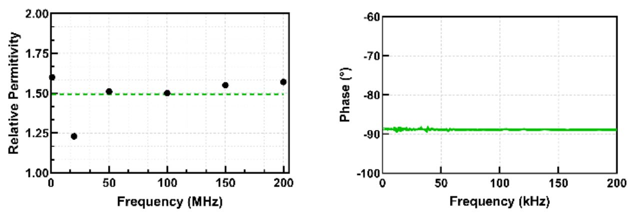
Fig. 2(a) shows the change in relative permittivity over the specified frequency range, showing relative stability around the value of 1.7, which is less than fiberglass epoxy, but comparable with PTFE (Polytetrafluoroethylene) substrate. Furthermore, Fig. 2(b) depicts the change in phase angle over the specified frequency range. The phase angle is close to -90°, showing that the material can be used as a dielectric in capacitive structures.



a) b)

Fig. 2. Electrical characterization of the substrate: a) Relative permittivity from 1 MHz to 200 MHz; b) Change in phase angle over the same frequency range.

Furthermore, Sait et al. [14], has reported that biscuit and cookie type product can be stored at 4 °C for 6 weeks, and at –18 °C for 6 months. This indicates that this edible material can be used for long periods of time as an indirect spoilage detection mechanism, when put next to regular cookies and biscuits. Through contactless antenna measurements, with an LC circuit incorporated with the edible cookie dough-based substrate, changes in the edible material could be detected via changes in relative permittivity of the capacitive part of the LC sensor.

B. Testing an edible voltage divider circuit

The first proof-of-principle circuit we realized on the edible substrate was a voltage divider, the functionality of which was tested by connecting it to a generator on the one side, and a light emitting diode (LED) on the other, as it can be seen in Fig. 3. The resistors, which were made out of paste based on activated carbon, had average resistances of 59.6 kΩ and 72.2 kΩ. We proved the correct operation of the circuit by series connection of two edible resistors as well as connecting the LED, which can be turned on after connection of power supply.

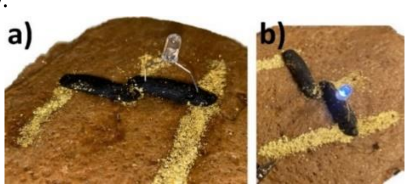


Fig. 3. Voltage divider: a) After fabrication; b) In operation regime.

C. Implementation and testing of an edible LC circuit

The fabricated edible LC resonator, the second proof-of-principle circuit on cookie dough substrate, is shown in Fig. 4. The circuit was successfully characterized at variable temperature and DI water addition, with results presented in Fig. 5(a) and Fig. 5(b). Temperature testing was performed in the range from 25 °C to 45 °C with the goal of simulating the temperature within the intraoral environment.

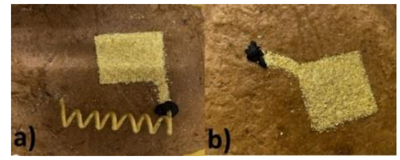


Fig. 4. Edible LC circuit: a) One side comprising one plate of the edible capacitor, made with edible gold leaf, interconnected through a black edible paste (the same used in Fig. 3) to the edible solenoid, made with gold-coated spaghetti; b) The opposite side, presenting the second plate of the edible capacitor.

The resonant frequency of the circuit can be expressed as:

$$f_{res} = {}^{1}/_{2\pi\sqrt{LC}} \qquad (1)$$

Where L is the inductance and C the capacitance of the resonator. f_{res} was recorded as a function of temperature within the tested range (Fig. 5(a)). f_{res} shifts towards higher frequencies with increasing temperature. The initial value at 25 °C was 70.9 MHz, then the resonant frequency increased until 75.9 MHz at 45 °C. This is due to the decrease in the value of the relative permittivity with increasing temperature, which leads to the decrease in capacitance. More precisely, the water content in the substrate decreases as it is heated, and because of that the relative permittivity decreases. As the capacitance is inversely proportional to the square of f_{res} (1), f_{res} increases with increasing temperature. Furthermore, f_{res} drops with the addition of DI water. This can be attributed to the high dielectric constant of DI water, approximately 80, which increases the capacitance of the circuit. The behavior of the LC circuit was then tested with the addition of artificial saliva and pH buffer solution separately, both with a pH of 6, which produced a slight decrease in resonant frequency, as reported in Fig. 5(b). The motivation behind pH testing, is to see how much the resonant frequency changes in different pH environments, as the end goal is to create a substrate that can be edible and functional. That is why the chosen pH solutions were in the range between 6.0 and 8.0, to simulate intraoral behavior. The results of this experiment can be seen in Fig. 5(c) and (d), where a slight decrease in f_{res} with increasing pH is reported: f_{res} drops from 61 MHz at pH 6 to 56 MHz at pH 8 (Fig. 5(c)). Such measurement indicates the possibility to sense the pH of an aqueous solution by tracking f_{res} of the proposed edible circuit. The dependence on pH level can be explained by the interaction of the carbon-based cookie dough surface with the ionic solution. Moreover, as presented in Fig 5(d), the capacitance increases with the increase in pH, as a consequence of an increase in relative permittivity. This is associated with changes on the surface of the dielectric carbon-based material [17]. The variation in capacitance will result in resonant frequency changes as it is presented in Fig. 5(c). Large variations in temperature, can affect the behavior of the LC structure, as an increase in temperature leads to an increase in resonant frequency. We have mainly tested the LC structure in ranges of body temperature, as we intended it to be used in an intraoral environment, however higher changes can occur if the sensor is tested on low and high temperatures, as variations in temperatures can affect the properties of the material. Kim et al. [18] reported that with increase in moisture, the value of the dielectric constant increases, which will lead to a decrease in resonant frequency. With that in mind, increase in temperature leads to a decrease in moisture content in cookie dough, which leads to an increase in resonant frequency, through the decrease of the dielectric constant.

When observing temperature changes, the calculated sensitivity was 0.25 MHz/°C. On the other hand, the influence of pH in the range of 4 to 8, has given a sensitivity of 2.5 MHz/pH. This is much more significant than the sensitivity obtained with change in temperature. Furthermore, the differences in liquids showed in Fig. 5(b), have been subjugated to a t-test between one another. Statistical significance ($p = 0.01$) was seen between the resonant frequency of the dry LC structures and those with liquids on them. An interesting result was obtained when comparing DI water with artificial saliva of pH 6. A statistical significance was obtained when comparing their resonant frequency, with a p value of 0.024. This gives room for further improvements. Moreover, no statistical significance was found when comparing pH buffer of pH 6 and artificial saliva with the same pH ($p=0.5$), which is expected. When comparing the resonant frequency of the LC structures covered with a pH buffer and DI water, no statistical significance was seen ($p= 0.07$).

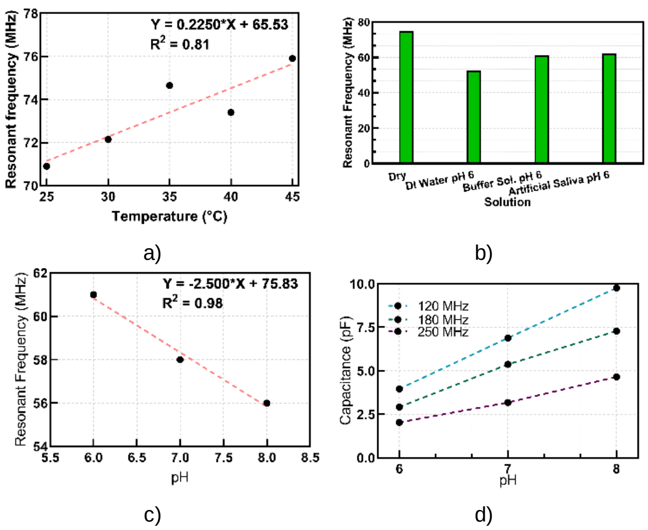


Fig. 5. a) Resonant frequency change of the edible LC resonator fabricated on the cookie dough substrate with a) change in temperature (fitting method: Linear Regression); b) change in dielectric environment: from dry conditions to wet conditions with different fluids at pH = 6. c), d) Parameter testing in different pH buffer solutions - Resonant frequency of edible LC circuit as a function of pH (fitting method: Linear Regression).

IV. CONCLUSION

We have developed an edible substrate based on cookie dough, which shows promising results in the field of printed edible electronics. The substrate, even though is affected by water exposure in terms of dielectric constant, does not degrade. It is rigid enough to provide a stable printing environment, even paving the way to potential uses in pinhole electronic circuits. By developing two functional edible circuits on it, we provide a proof of concept for successful realization of cookie dough-based electronics. Aside from these realizations, cookie dough-based substrates have been used in sensing in edible electronics.

REFERENCES

[1] A. S. Sharova, F. Melloni, G. Lanzani, C. J. Bettinger, and M. Caironi, “Edible Electronics: The Vision and the Challenge,” Advanced Materials Technologies, vol. 6, no. 2, p. 2000757, Nov. 2020, doi: https://doi.org/10.1002/admt.202000757.

[2] B. H. Robinson, “E-waste: An assessment of global production and environmental impacts,” Science of The Total Environment, vol. 408, no. 2, pp. 183–191, Dec. 2009, doi: https://doi.org/10.1016/j.scitotenv.2009.09.044.

[3] N. Singh and O. A. Ogunseitan, “Disentangling the worldwide web of e-waste and climate change co-benefits,” Circular Economy, vol. 1, no. 2, p. 100011, Sep. 2022, doi: https://doi.org/10.1016/j.cec.2022.100011.

[4] L. Lamanna, P. Cataldi, M. Friuli, C. Demitri, and M. Caironi, “Monitoring of Drug Release via Intra Body Communication with an Edible Pill,” Advanced Materials Technologies, vol. 8, no. 1, Oct. 2022, doi: https://doi.org/10.1002/admt.202200731.

[5] A. Bouzidi et al., “Electronic conduction mechanism and optical spectroscopy of Indigo carmine as novel organic semiconductors,” Optical and Quantum Electronics, vol. 50, no. 4, Mar. 2018, doi: https://doi.org/10.1007/s11082-018-1439-7.

[6] B. Petrović et al., “Toothpaste ingestion—evaluating the problem and ensuring safety: systematic review and meta-analysis,” Frontiers in Public Health, vol. 11, Oct. 2023, doi: https://doi.org/10.3389/fpubh.2023.1279915.

[7] R. Priyadarshi and J.-W. Rhim, “Chitosan-based biodegradable functional films for food packaging applications,” Innovative Food Science & Emerging Technologies, vol. 62, p. 102346, Apr. 2020, doi: https://doi.org/10.1016/j.ifset.2020.102346.

[8] I. K. Ilic, L. Lamanna, D. Cortecchia, P. Cataldi, A. Luzio, and M. Caironi, “Self-Powered Edible Defrosting Sensor,” ACS Sensors, vol. 7, no. 10, pp. 2995–3005, Oct. 2022, doi: https://doi.org/10.1021/acssensors.2c01280.

[9] T. Sogabe, R. Kobayashi, P. Thanatuksorn, T. Suzuki, and K. Kawai, “Physical and structural characteristics of starch‐based and conventional cookies: Water sorption, mechanical glass transition, and texture properties of their crust and crumb,” Journal of Texture Studies, vol. 52, no. 3, pp. 347–357, Feb. 2021, doi: https://doi.org/10.1111/jtxs.12585.

[10] D. Sert, M. K. Demir, and N. Ertaş, “Rheological, physical and sensorial evaluation of cookies supplemented with dairy powders,” Food Science and Technology International, vol. 22, no. 3, pp. 196–202, Apr. 2015, doi: https://doi.org/10.1177/1082013215583149.

[11] J. Willmann, D. Stocker, and E. Dörsam, “Characteristics and evaluation criteria of substrate-based manufacturing. Is roll-to-roll the best solution for printed electronics?,” Organic Electronics, vol. 15, no. 7, pp. 1631–1640, Jul. 2014, doi: https://doi.org/10.1016/j.orgel.2014.04.022.

[12] M. G. Gutiérrez‐Cárdenas et al., “Dielectric properties analysis of dough and further microwave baking for cookies,” Journal of Food Processing and Preservation, vol. 46, no. 9, Aug. 2022, doi: https://doi.org/10.1111/jfpp.16940.

[13] B. Pareyt and J. A. Delcour, “The Role of Wheat Flour Constituents, Sugar, and Fat in Low Moisture Cereal Based Products: A Review on Sugar-Snap Cookies,” Critical Reviews in Food Science and Nutrition, vol. 48, no. 9, pp. 824–839, Sep. 2008, doi: https://doi.org/10.1080/10408390701719223.

[14] D. I. Sait. (2006). Effect of Oven Types on the Characteristics of Biscuits Made from Refrigerated and Frozen Doughs. Food Technol. Biotechnol. 44.

[15] S. Grasso and D. Asioli, “Consumer preferences for upcycled ingredients: a case study with biscuits,” Food Quality and Preference, vol. 84, p. 103951, Apr. 2020, doi: https://doi.org/10.1016/j.foodqual.2020.103951.

[16] 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.

[17] Rao, M.A., Rizvi, S.S.H., Datta, A.K., & Ahmed, J. (Eds.). (2014). Engineering Properties of Foods (4th ed.). CRC Press. https://doi.org/10.1201/b16897, pages 533 – 565.

[18] Y. R. Kim, M. T. Morgan, M. R. Okos, and R. L. Stroshine, "Measurement and prediction of dielectric properties of biscuit dough at 27 MHz," *Journal of Microwave Power and Electromagnetic Energy*, vol. 33, no. 3, pp. 184–194, 1998. https://doi.org/10.1080/08327823.1998.11688375