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DOI:

[10.1016/j.orgel.2020.105735](https://doi.org/10.1016/j.orgel.2020.105735)

Document Version

Accepted author manuscript

[Link to publication record in Manchester Research Explorer](#)

Citation for published version (APA):

Seck, M., Mohammadian, N., Diallo, A. K., Faraji, S., Erouel, M., Bouguila, N., Ndiaye, D., Khirouni, K., & Majewski, L. A. (2020). Organic FETs using biodegradable almond gum as gate dielectric: A promising way towards green electronics. *Organic Electronics: physics, materials, applications*, 83, 105735. <https://doi.org/10.1016/j.orgel.2020.105735>

Published in:

Organic Electronics: physics, materials, applications

Citing this paper

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ORGANIC FETs USING BIODEGRADABLE ALMOND GUM AS GATE DIELECTRIC: A PROMISING WAY TOWARDS GREEN ELECTRONICS

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Abstract

Green electronics is an emerging field of research which aims to manufacture devices in an environmentally friendly and sustainable way. Usually, the involved electronic materials are naturally occurring and non-toxic. Also, they can be processed using simple, low energy deposition processes and fabrication techniques. In this work, we present low voltage organic field-effect transistors (OFETs) using almond gum (AG) as the gate dielectric. AG is a natural, biodegradable insulator material that can be directly collected from almond trees and used without any further purification. Moreover, AG possesses interesting properties such as water solubility, ease of processing, good insulation, low leakage current, good film quality, and high capacitance making it a promising dielectric for OFET devices. Bottom gate/bottom contact p-channel OFETs have been processed on glass substrates with poly(3,6-di(2-thien-5-yl)-2,5-di(2-octyldodecyl)-pyrrolo[3,4-c] pyrrole-1,4-dione)thieno[3,2-b] thiophene) (DPPTTT) polymethyl methacrylate (PMMA) blend as the active layer and gold as the source and drain electrodes. The transistors operate at low voltage ($V_{GS} \leq 3$ V), with threshold voltages V_{th} as low as -0.8 V, saturated field effect mobilities μ_{sat} above $0.75 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, subthreshold swings SS around 270 mV/dec and ON/OFF current ratio equal to 10^3 . The combined favourable properties of both almond gum and low voltage operated OFET devices have a high potential to pave a way towards using naturally occurring, biodegradable electronic materials in future disposable sensors or throwaway, low-end electronics.

Keywords: Green electronics, almond gum, dielectric, low voltage organic transistors (OFETs)

1. Introduction

We live in a world where electronic devices play an important role in our daily life becoming a necessity in every home, office and almost in every street corner. With the rapid progress of new technologies in mobile phones, tablets and smart wearables, electronic devices become quickly an electronic waste (e-waste) in a very short time. Each new innovation in electronic technologies reduces the lifespan of electronic devices leading to an increase in the rate of obsolescence without increasing its reparability or recyclability [1]. As a result, electronic waste has become one of the worst ecological threats that our planet is currently facing. E-waste is considered as one of the fastest growing pollution problems in the world because of the presence of several substances considered as toxic that can negatively impact the environment and human health. According to the United Nations University, in 2016, the generation of e-waste in the world was estimated to be about 44.7 million metric tons annually, equivalent to 4500 Eiffel Towers and this amount is expected to increase to 52.2 million metric tons by 2021 [2, 3]. One potential way to reduce e-waste is either to use already available materials or synthesize new materials that can be partially or fully decomposed. From that point of view, naturally occurring, biodegradable materials are perfect candidates to be fully compatible with the idea of green electronics.

Since the advent of green electronics that aims to use environmentally friendly materials and processes there is hope to develop more ecological and recyclable electronic devices. It is worth noting that the key element of modern electronics still remains the field-effect transistor (FET). FETs are present in almost all electronic circuits and systems ranging from analog to digital. Amongst FETs, organic field-effect transistors (OFETs) have recorded a rapid development in recent years because they can be easily fabricated from solution on large areas using a wide range of substrates such as glass or paper [4]. To date, several types of biodegradable materials have been used in OFETs [5]. Considering the biodegradable dielectric materials, cellulose [6-8], alginate [9], silk [10, 11], chitosan [12, 13], collagen [14, 15] and gelatin [16] have been previously shown to result in well working organic transistors. However, most of the demonstrated OFETs displayed relatively high threshold voltages and subthreshold swings, and as result, they operated with gate voltages ($V_{GS} \geq 5$ V). This shows that the fabrication of high performance, low voltage organic FETs using biodegradable dielectric materials is not easy. Here, we study the structural and dielectric properties of naturally occurring almond gum (AG) and show that it can be successfully used to fabricate low threshold voltage OFETs that can be operated at $V_{GS} \leq 3$ V without any special treatments or precautions.

Almond gum is a substance produced by almond trees (*Prunus dulcis*) as a result of either a disease called gummosis or a mechanical injury. AG is a natural polysaccharide polymer known to possess anti-cancerous [17, 18], antioxidant [19, 20] and anti-inflammatory properties [21]. Its non-toxicity, solubility in water, as well as its environmentally friendly character allowed this material to attract significant interest in both food and pharmaceutical industries. Recently, it has been studied as an alternative adsorbent for the removal of methylene blue dye from wastewater [22]. What is important, AG is an exceptionally inexpensive material (circa \$5 per kg) and is readily available in the market. As such, we chose to study almond gum as a low cost, biodegradable dielectric material for solution-processed organic transistors.

The electrical characteristics of the fabricated poly(3,6-di(2-thien-5-yl)-2,5-di(2-octyldodecyl)-pyrrolo[3,4-c] pyrrole-1,4-dione)thieno[3,2-b] thiophene) (DPPTTT) polymethyl methacrylate (PMMA) blend OFETs using AG as dielectric show that the devices can be operated at gate voltages (V_{GS}) ≤ 3 V, threshold voltages (V_{th}) as low as -0.8 V, saturated field effect mobilities (μ_{sat}) above $0.75 \text{ cm}^2.\text{V}^{-1}.\text{s}^{-1}$, subthreshold swings (SS) around 270mV/dec, and ON/OFF current ratios up to 10^3 .

2. Experiment

Fresh almond gum was collected from the almond tree (Figure 1a) in Djerba, southern region of Tunisia, and was dried in a stove at 80°C for 4 hours. The dry AG was crushed with mortar and pestle and sieved using a sieve of size 100 μm (Figure 1b). A solution of almond gum was obtained by dissolving 0.25 g of powder of AG in 29.75 ml of distilled water in a small vial. To obtain uniform solution, it was stirred with magnetic stirrer on a hot plate at 40°C and then filtered through a 0.2 μm PTFE filter (Figure 1c). Next, 8ml of the filtered solution was deposited by spray pyrolysis on gold-coated glass slides. The distance between spray nozzle and substrate was 20 cm with nitrogen carrier gas flow maintained at 0.8 bars. During spray pyrolysis, the sample temperature was initially kept at $T=200^\circ\text{C}$ and spray process was stopped after 5min. To conduct capacitance measurements, 100nm of aluminium (Al) was thermally evaporated through a square shaped shadow mask with 0.01 cm^2 of area. The capacitance values were measured at variable frequencies ranging from 100Hz to 1MHz. For the fabrication of OFETs, firstly, 50 nm of Au was deposited to form the gate electrode. Then, (900 $\pm$ 100) nm of almond gum film was formed using the process described above, and the thickness was measured by a Dektak 3 Profilometer. Subsequently, 50 nm of Au was evaporated through a

shadow mask to form the source and drain contacts. The channel length (L) and width (W) were 30 and 1000 μm , respectively. Afterwards, Gold electrodes were treated with a 2 mM 2,3,4,5,6-pentafluorothiophenol (PFBT) solution in isopropanol (IPA) for 2 min and dried at 100 °C for 5 min. Finally, the organic semiconductor blend was prepared by dissolving 0.5 wt. % DPPTTT and PMMA in 1, 2-dichlorobenzene (DCB) at 70:30 ratio. Then, the polymer blend was spin-coated at 2000 rpm for 2 min and the resulting thin films were annealed at 100 °C for 1 h under dry nitrogen. The structure of the fabricated bottom gate/bottom contact OFETs is illustrated in Figure 1d. The electrical characterization was performed in ambient air using an Agilent E5270B measurement mainframe with Karl Suss PH100 micromanipulator probes. The X-ray diffraction (XRD) of almond gum powder was obtained with a Philips PW1840 diffractometer using copper anode which X-ray wavelength is $\lambda=1.54060 \text{ \AA}$ and XR generator parameters 40 kV and 25 mA. Using the automatic diffractometer software, the angular scanning was carried out from 10 degrees up to 80 degrees with steps of 0.02 degrees. Fourier Transform Infrared Spectroscopy (FTIR) analysis of almond gum was recorded in the region 400-4000 cm^{-1} with Perkin-Elmer Spectrum Two Spectrophotometer.

3. Results and discussion

To study the structural properties of the almond gum, X-ray diffraction (XRD) was carried out. A typical XRD of almond gum is depicted in Figure 2a. As can be seen, there are large diffraction peaks at around $\theta = 20^\circ$ and $2\theta = 42^\circ$ in the diffractogram that indicates the presence of very small crystallites. However, the overall structure of the studied almond gum films can be considered to be amorphous [23-25].

Subsequently, FTIR has been used to qualitatively check the functional groups present in the almond gum. These groups are identified by the presence of peaks in the FTIR spectrum. In inset of the Figure 2a, the shape of the obtained curve is similar to those obtained in previous works [21, 23]. The peak observed at 3289.99 cm^{-1} corresponds to the stretching vibration of O-H group and that at 2913.7 cm^{-1} is assigned to the vibrational modes of C-H group. The bound at 1602.24 cm^{-1} is associated with the asymmetrical stretching of carboxylate (COOH) groups or intramolecularly bound water and the peak at 1415.17 cm^{-1} may be attributed to a symmetrical stretching of carboxylate groups of uronic acid [23].

To study the dielectric properties of the prepared almond gum films, metal-insulator-metal (MIM) capacitors (cf. inset in Figure 3b) have been fabricated. Both Au/AG/Al and ITO/AG/Ag structures were studied. The overlap area of the electrodes was 10⁻² cm^2 for capacitance and

0.06 cm² for leakage current density measurements, respectively. Figure 3a shows a typical capacitance vs. frequency of Au/AG/Al, as well as current density vs. applied voltage bias characteristic of ITO/AG/Ag devices. As can be seen, the capacitance decreases as the frequency increases. The capacitance changes are recorded from 8.2 nF/cm² to 6.3 nF/cm² for frequencies ranging from 100 Hz to 1 MHz, respectively. This capacitance decrease with frequency is thought to be due to polarization effects in the dielectric. Indeed, the polarization phenomenon is commonly attributed to the charges present in the material [26]. Without an electric field, the dipoles present in the material are randomly oriented and their resultant net polarization is zero. However, to observe a significant polarization, it is therefore necessary to disturb the distribution of these dipoles by an application of an electric field. Under these conditions, some of the dipoles are oriented in the direction of the applied field. Capacitance increase is related to the ability of dipoles in the material to be polarized, i.e. the higher the dipole polarization the higher the capacitance. For relatively low frequency values, the dipoles have enough time to follow the electric field, thus causing an increase in the capacitance, whereas for high frequency values, these dipoles can no longer follow the electric field leading to a decreased capacitance. Such behaviour has been previously observed in naturally occurring materials because natural-resource-based gate dielectrics often display high dependence of capacitance on frequency [27]. Inset in Figure 3a shows the leakage current density versus applied voltage bias of the almond gum dielectric. Leakage current is an important parameter for assessing the dielectric behaviour of insulators. It is worth noting that in low voltage thin film transistors (TFTs), leakage current has to be minimized not only to reduce the power consumption but also to improve the switching efficiency of the transistors [28]. In our MIM structures, the leakage current density reaches 10⁻⁶ A/cm² for an applied voltage bias of 1 V. Since gum exudates like almond gum usually possess several types of ions such as calcium, sodium, potassium, iron, etc. and OH groups, the cause of the leakage current observed in AG is probably related to the presence of mobile charges and hydroxyl (OH) groups [27]. Previously reported FTIR chemical analysis composition of almond gum [21, 29] and our results (inset Figure 2a) suggest that this is indeed the case. Figure 3b shows the dielectric vs. applied voltage of an ITO/AG/Ag MIM structure at 1 kHz. As can be seen, no changes have been observed and the dielectric constant remains stable whatever the applied voltage. The calculated dielectric constant has been found to be between 7 and 9. It is important to mention that the dielectric constant of AG films strongly depends on the composition of the material but also on how the films were prepared, i.e. the material deposition technique, humidity, solvent, thermal treatments, etc. all affect the dielectric constant of AG films. As a result, more detailed

studies of the dielectric properties of almond gum thin films are needed to establish how their dielectric constant depends on all the aforementioned factors.

Figures 4a and 4b show typical output and transfer characteristics of the fabricated DPPTTT-PMMA OFETs measured in ambient atmosphere, respectively. After polarizing negatively the gate and drain electrodes with respect to the grounded source electrode, well-defined linear and saturation regimes with a good pinch-off region were obtained. As can be seen, the device operates in the accumulation mode and shows p-channel behaviour. A small hysteresis is observed during forward and back gate voltage scan in the transfer characteristic. This instability is probably due to the charge trapping at the semiconductor-insulator interface and is highly undesirable in electronic circuits [30]. All the electrical OFET parameters were calculated in the saturation regime using the standard metal-oxide-semiconductor field-effect transistor (MOSFET) equation [31]:

$$I_{DS,sat} = \frac{W}{2L} \mu C_i (V_{GS} - V_{th})^2. \quad (1)$$

where $I_{DS,sat}$ is the drain current in the saturation regime, μ_{sat} represents the field effect mobility, W is the channel width and L the channel length, C_i is the capacitance per unit area, V_{GS} the gate voltage and V_{th} the threshold voltage.

The electrical parameters extracted from the transfer curves are summarized in table 1. To calculate the field-effect mobility in the saturation regime the areal capacitance measured at 1 kHz was taken (i.e. $C = 7.2 \text{ nF/cm}^2$, cf. Fig. 3a). It has been found that $\mu_{sat} = 3 \text{ cm}^2/\text{Vs}$. However, we speculate that this μ_{sat} value is largely overestimated because the current-voltage characterizations of our transistors were carried out in semi DC conditions. It is reasonable to assume that at low frequencies ($f < 100 \text{ Hz}$) the areal capacitance of our AG films is about 3-4 times higher. As a result, we estimate that the factual field-effect mobility in the saturation regime is approximately $0.75 \text{ cm}^2/\text{Vs}$. This value is of the same order of magnitude than that obtained with cyanoethyl cellulose dielectric [32] using DPPTTT-PMMA as the active layer. The relatively high μ is attributed to a good charge transport that is related to the high interface quality between the organic semiconductor and the AG dielectric. As shown in the transfer and output characteristics, our transistors operate at $V_{GS} \leq 3 \text{ V}$ and display relatively low threshold voltage $V_{th} = -0.8 \text{ V}$. Low V_{th} is very important especially for reduction of power consumption in portable electronic devices which are increasingly miniaturized. Another important parameter playing a critical role in power consumption is the subthreshold swing (SS). SS determines how much gate voltage is necessary to increase the drain current by one order of

magnitude [33]. It is extracted from the logarithmic scale of transfer characteristic by using the following equation [31]:

$$SS = \frac{dV_{GS}}{d \log I_{DS}} \quad (2)$$

In most studies that have been carried out on OFETs using polymers as the active layer the subthreshold swings have usually been much higher than 500 mV/dec. This severely limits the suitability of these devices for integrated circuits [34]. To reduce the operational voltage of OFETs, low number of semiconductor-insulator interface states which could act as traps for charge carriers during the transport in the transistor channel are required. Our polymer FETs using almond gum as the gate dielectric exhibit $SS < 270$ mV/decade which reflects a good quality of the interface between AG and DPPTT-PMMA. From Figure 4b it can be seen that the gate leakage current (I_{GS}) increases up to 10^{-7} A for $V_{GS} = -3$ V. However, I_{GS} can be significantly reduced by patterning of the semiconductor, as well as shrinking of the device dimensions [35]. The calculated ON/OFF current ratios of our devices is around 10^3 . This appears to be comparable with ON/OFF ratios of most low voltage polymer FETs reported previously. As can be seen in Table 2, although the used semiconductors were different, comparable OFET performances have been achieved in this study.

Amongst naturally occurring insulator materials, chicken albumen, silk fibroin, glucose and DNA nucleobases appear to be interesting natural dielectrics for OFETs. For example, chicken albumen displays a relatively high dielectric constant of 5.3 – 6.1 and water contact angle around 60 – 80° for layers baked at 160 °C [41]. The calculated threshold voltage for the recently reported pentacene and C_{60} OFETs, as well as IGZO TFTs using chicken albumen as the gate insulator was -8 V [41], 1.5 V [41] and 2.25 V [42], respectively. The on/off ratio was found to be 10^4 for both pentacene and C_{60} OFETs and 2×10^7 for IGZO TFTs. The values of the saturated field effect mobilities for the respective devices were $0.09 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, $0.13 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ and $5.64 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$. Also, L-S Tsai *et al.* [43] demonstrated C_{60} /pentacene FETs based on silk fibroin having a threshold voltage of 5.9 V, a saturated field effect mobility of $10 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ and an on/off current ratio of 10^3 in ambient air. Alternatively, fullerene OFETs using glucose as the gate dielectric ($k = 6.35$) were reported [44]. Lately, DNA nucleobases such as guanine ($k = 4.35$) and cytosine ($k = 4.65$) have also been used as the gate dielectric in organic field-effect transistors [44]. The fabricated devices exhibited subthreshold swings of 1.4 V/dec and 1.2 V/dec for guanine and cytosine, respectively. The saturated field effect mobilities were $0.12 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ for OFETs using Guanine and $0.09 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ for those based on cytosine. In comparison with our work, we note that the involved AG dielectric shows higher dielectric

constant ($k \sim 7.2$) and our OFETs display much lower threshold voltages (-0.8 V) and subthreshold swings (~ 270 mV/dec). In addition, the recorded water contact angle of almond gum films is 92° as shown in the inset of Figure 4. This confirms that AG film is more hydrophobic than chicken albumen [41] and could therefore be more suitable as gate dielectric in OFETs. It is important to point out that traditional polymer dielectrics are processed from solution and are particularly useful for large-area or flexible electronics. Unfortunately, many of these polymer insulators are only soluble in solvents that are toxic and expensive. Almond Gum has advantage of being natural, biodegradable, low-cost and water soluble. AG is easy to be processed into thin films and exhibits good dielectric properties. This includes a reasonably low leakage current, good film quality and high capacitance. We think that all the above-mentioned features make almond gum a promising dielectric for low voltage OFET devices.

4 Conclusion

In this study, we have fabricated bottom gate/bottom contact organic field-effect transistors using water soluble almond gum, a natural biodegradable material freshly collected from the almond tree, as the gate dielectric. The diffractogram recorded for almond gum polysaccharide shows that almond gum films are largely amorphous. However, the presence of broad peaks suggests that small crystallites may be present. FTIR spectra confirm the presence of hydroxyl and carboxylate groups in the chemical composition of the almond gum. The characterization of MIM structures reveals that the capacitance per unit area is 7.2 nF/cm² at 1 kHz and leakage current density below 10^{-6} A/cm². The electrical characterization showed that the AG DPPTTT-PMMA OFETs display $V_{th} = -0.8$ V which enables operation of the devices with $V_G \leq -3$ V. The electrical parameters extracted from the transfer characteristics show a relatively high saturation field-effect mobility $\mu = 0.75$ cm².V⁻¹.s⁻¹ and subthreshold swing around 270 mV/dec which confirms a good quality of the interface between AG and DPPTTT-PMMA polymer blend semiconductor. As a result, the use of the almond gum in organic transistors has a high potential to open a new promising way towards green manufacturing of the next generation of low-end sensors and electronics.

Acknowledgements

N. Mohammadian and L.A. Majewski would like to thank James Sexton for the thickness measurement of the almond gum films. M. Seck and A. K. Diallo thank the CEA-MITIC (Centre d'Excellence Africain en Mathématiques, Informatique et TIC) for the financial support.

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Figure Captions

Figure 1: (a) Almond tree (*Prunus dulcis*) with almond gum on the branch, (b) Almond gum dried, crushed and sieved, (c) solution of Almond gum in water and (d) bottom gate bottom contact OFET using almond gum as dielectric is illustrated.

Figure 2: X-ray diffraction of almond gum. Inset is the FTIR spectrum of AG.

Figure 3: (a) Capacitance per unit area vs. frequency. Inset represents leakage current density vs. voltage using MIM structure. (b) Dielectric constant vs. voltage at $f=1$ kHz. Inset is the structure of the studied MIM capacitors.

Figure 4: A representative (a) output and (b) transfer characteristics of an OFET using almond gum as the gate dielectric. Inset in (b) represents the water contact angle of AG film.

Table Captions

Table 1: Electrical parameters of a typical OFET using almond gum as the gate dielectric.

Table 2: A comparison of OFET performances using almond gum as the gate dielectric with several polymer dielectrics reported in the literature.

Figures

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Tables

Table 1: Electrical parameters of a typical OFET using almond gum as the gate dielectric.

Parameters	μ ($\text{cm}^2.\text{V}^{-1}.\text{s}^{-1}$)	V_{th} (V)	SS (mV/dec)	ON/OFF current ratio
Values	0.75	-0.80	266	10^3

Table 2: A comparison of OFET performances using almond gum as the gate dielectric with several polymer dielectrics reported in the literature.

Parameters	μ ($\text{cm}^2.\text{V}^{-1}.\text{s}^{-1}$)	V_{th} (V)	SS (mV/dec)	ON/OFF Ratio
Almond gum [This work]	0.75	-0.80	266	10^3
c-PMMA [36, 37]	0.015	-1.00	-	10^5
c-PVP [38]	0.048	-0.45	~ 300	10^3
P(VDF-TrFE) [39]	0.0004	(-1) - (-3)	~ 500	$(10^3) - (10^5)$
BST-P(VDF-HFP) [40]	0.14	-0.50	221	10^3
Chicken albumen (p-type OFETs) [41]	0.09	-8	-	10^4
Chicken albumen (n-type OFETs) [41]	0.13	1.5	-	10^4
Chicken albumen (IGZO TFTs) [42]	5.64	2.25	1150	2×10^7
Silk fibroin [43]	10	5.9	-	10^3
Glucose [44]	0.085	-	6200	-
Guanine [44]	0.12	-	1400	-
Cytosine [44]	0.09	-	1200	-