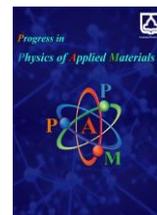




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# Nonferritic Metallic Oxide / Polyvinyl Alcohol Polymer Nanocomposite as a Biocompatible Dielectric Material for Future Generations of Clean Energy Systems

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## ARTICLE INFO

### Article history:

Received: 11 June 2025

Revised: 23 July 2025

Accepted: 23 July 2025

Published online: 2 September 2025

### Keywords:

Clean energy systems;

Nonferritic material oxide;

Polyvinyl Alcohol Polymer (PVA);

Eco-friendly.

## ABSTRACT

In the last decades, there has been much research in the manufacture of clean energy systems such as display components and chips due to the large-scale production and economical synthesis conditions. For getting low dependence on fossil fuel consumption, it has led researchers to use, as an example of organic field effect transistors (OFETs), eco-friendly gate dielectric materials which are biocompatible with nature. For this reason, among a large number of metal oxides, polymers and organic materials, as a novelty of the present work, the electrical and dielectric characteristics of nonferritic metallic- lithium oxide (NFLiO<sub>x</sub>) an eco-friendly metal oxide and polyvinyl alcohol polymer (PVA) are investigated and tested as a possible alternative biocompatible dielectric material for the next OFET generations.

## 1. Introduction

In recent years, various energy devices such as supercapacitors, energy storage, and especially field effect transistors, have been the focus of attention of engineers in the optoelectronic, and medical application industries [1-3]. With the advancements in technology, the demand for these electronic chips has increased significantly. Traditional energy devices are mainly composed of non-biodegradable materials, which lead to destructive environmental issues. The indestructibility of these energy devices after burying leads to environmental pollution. In addition, these devices often lead to wastage of valuable metal resources [1]. Thereby, the use of polymers in various industries and energy devices seems inevitable. It means that people should find a solution route for these environmental

concerns and the challenges of electronic chip. The Intergovernmental Panel on Climate Change has stated that the temperature of the earth's climate has reached a level of 1.09 degrees Celsius (in 1850-2020 years) [1].

The main reasons for Climate change are the emission of greenhouse gases, atomic waste and electronic chips. In fact, in these years, the global average concentration of carbon dioxide in the atmosphere has increased to about 419 ppm (a polluting particle out of a metallic particle in the earth's atmosphere) [2, 3]. In particular, according to this report, 72% of greenhouse gas emissions in the world were caused by the production of electricity and energy systems in total [4]. These data clearly indicate the need to reverse this trend. Many studies have been done to determine a strategy and a new approach in reducing this alarming trend.

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### Cite this article as:

Bahari, A., Fallah Hamidabadi, V., Farhadi Koutenaeei, A., and Moradbeigi, N., 2026. Nonferritic Metallic Oxide / Polyvinyl Alcohol Polymer Nanocomposite as a Biocompatible Dielectric Material for Future Generations of Clean Energy Systems. *Progress in Physics of Applied Materials*, 6(1), pp.27-34. DOI: [10.22075/ppam.2025.38068.1150](https://doi.org/10.22075/ppam.2025.38068.1150)

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However, a sustainable process must be taken so that the next generations should have the least negative impact on the environment and nature [5-8]. Therefore, to discover and implement innovative and sustainable materials and production methods, one needs to conduct fundamental research and develop new strategies.

On the other hand, the use of polymers and conjugated polymers in flexible electronic industries, fuel cells and transistors due to their mass production and large scale, simple and economical synthesis conditions should be considered with these concerns. To reduce the negative environmental effects of these materials, it is necessary to go for materials that are friendly to nature and include in the field of green synthesis [8-15]. The fact is that biodegradable polymers can be regenerated into hydrogels, fibers, foams, films, tubes, particles, and other forms [14]. Polymers generally originate from renewable or biological sources such as animals, plants, marine organisms, and microorganisms, while synthetic polymers are chemically synthesized [15].

First polymers include polysaccharides such as starch, cellulose, chitosan, and chitosan derivatives, as well as protein-based polymers such as silk fibroin, collagen, and silk. Biodegradable synthetic polymers usually contain ester, amide or ether linkages and include polylactic acid, polycaprolactone, polyurethane, polyethylene glycol, poly(lactic-co-glycolic acid), polybutylene, and polyvinyl alcohol [15-22]. There are two ways to solve the problems and challenges related to environmental issues and greenhouse effects: in the first way, one can use biocompatible solutions such as water, hybrids, precursors and ligands. They are not going to use toxic solvents, non-removable and non-degradable pollutants, and the best solvent for them is water solvent. In the second way; Biodegradable polymers should be used that both maintain the stability of their characteristics during operation conditions. It means that they should be easily destroyed using chemical reactions or environmental microorganisms into carbon dioxide [23-32]. In addition, electronic chips, and here OFETs, can meet the demand for versatility, efficiency in energy supply [12] and electrical devices [13], metamaterials [10] and smart wearables [9].

For this purpose, lithium metal oxide was synthesized from lithium nitride with the help of a water solvent and under environmental conditions with PVA polymer as a possible gate dielectric of OFET transistors. By measuring the electrical transfer and output characteristics of OFETs; such as carrier mobility (there is a direct relationship between carrier mobility and the output current density), threshold voltage (lower threshold voltage, lower electricity consumption) and higher on-off current ratio and lower subthreshold voltage (SS) for faster OFET operations, compared to the current dielectric gate materials of chips and electronic components, NFLiOx/PVA nano composite is a more desirable gate dielectric and is also promised to be used in the future production of environmentally friendly electronic and optoelectronic chips.

## 2. Experimental Procedures and Details

It is known that there are at least four configurations of OFETs in the field of electronic industries, depending on the purpose of the manufacturers, they choose a specific configuration. The configuration is depicted in Figure 1. Firstly, the aluminum metal gate is placed under the silicon substrate. Then 2 ml nonferritic metallic; lithium oxide (which produced from 1 g lithium nitride in 1000 ml pure water) and 2 ml polyvinyl alcohol (which produced from 2 g PVA in 1000 ml pure water) with purity: 99.94% from Merck company, mixed together and poured dropwise on the P-type Si (100) substrate. Silicon (Si) wafer has  $1 \times 1 \times 0.2$  cm<sup>3</sup> dimensions, resistivity of 5 ohm-cm, was rinsed in sulfuric acid and then in acetone on the ultrasonic bath at room temperature. It has been transferred in the furnace for 2 more hours of drying time. Finally, NFLiOx/PVA gate dielectric with 5 nm thickness could be formed on the silicon substrate and on top of gate dielectric material, two source and drain aluminum electrodes with  $w = 500$   $\mu$ m are placed at a distance of 500  $\mu$ m from each other. Such a configuration where the gate is placed at the bottom of substrate and the two source- drain electrodes are on top of gate dielectric for Prova probe's contacting, is known bottom-gate top- contact (BGTC) configuration (which is shown in Figure 1 along with the composition of the chemical structure of the materials used).

As shown in Figure 1 one of the materials used in this research is lithium which cannot be found freely in nature. Monovalent lithium, is one of the lightest metals that easily dissolves in water, due to its high reactivity.

At the same time, the electron of Li last layer is easily released and becomes a cation. This feature makes this material a good conductor of electricity and suitable analyte for biosensors in medical instruments. In addition, lithium is widely used in various industries, including rechargeable batteries, mobile phones, electronic chips, rectifiers, and smart coatings. Moreover, it does not have a destructive greenhouse effect. It also can form lithium hydroxide in reaction with water and steam. In addition, when lithium nitrate dissolves in water, nonferritic metallic; lithium oxide is formed, which is capable of being a good electrical conductor, and due to electrostatic forces, it is weak and fragile and is not able to hold ions in its network. In other words, the ions are easily mobile in its structure and transfer electric charge.

Another material used is polyvinyl alcohol (PVA), which is made from polyvinyl acetate by hydrolysis method and is easily destroyed by biological organisms. This polymer can be decomposed in water in conducting electrical and ionic charges and characterized by biodegradation in a green synthesis route [32-35]. It is therefore a biodegradable polymer in a wide range of combinations with other polymer compounds such as biopolymers and other hydrophilic polymers, and its degradability increases through hydrolysis due to the presence of hydroxy groups in carbon atoms. In addition, the crystal structure of PVA, is connected by modifying the chemical composition of the control groups (OH) and with the atoms (H) between the hydroxyl groups and connects the hydrogen atoms together.

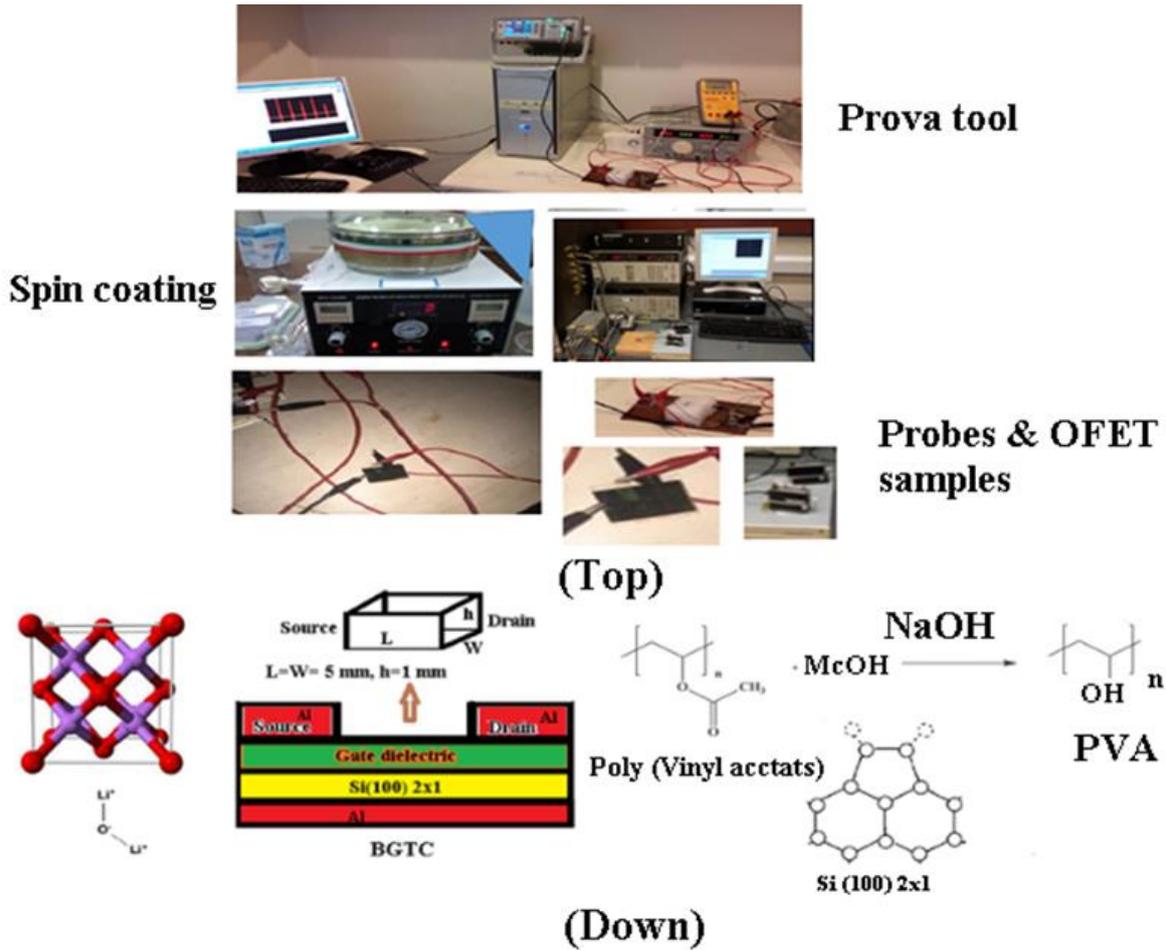


Fig. 1. (Up): Equipment (Prova, Spin coating and fabricated OFETs); (Down) Schematic view of an OFET with Al gate on the bottom and the source-drain electrodes on top of p-type Si(100) 2x1, known as BGTC transistors.

Table 1. Some characteristics of nonferritic metallic; lithium oxide and polyvinyl alcohol (PVA) [7,8].

"Properties"	"PVA (Purity: 99.94%)"	"Nonferritic metallic; lithium oxide" (Purity: 99.9%)"
"Molecular formula"	C <sub>4</sub> H <sub>6</sub> O <sub>2</sub> C <sub>2</sub> H <sub>4</sub> O <sub>2</sub>	NFLi <sub>2</sub> O
"Average molecular (g/mole)"	Wight: 44.05	Molar mass: 29.88
"Degree of polymerization"	1.400	N/A
"Degree of hydrolysis"	88%	N/A
"Density (g/cm <sup>3</sup> )"	1.19	2.01

The chemical structure of the materials used in the present work, includes non-ferritic metallic (NF)- Lithium oxide (LiO<sub>x</sub>), labelled as NFLiO<sub>x</sub>, PVA and silicon 2x1. Table 1 shows some specifications of cited samples.

### 3. Results and Discussion

Here, by referring to our previous works [6-10, 35] and also from Figures 1 and 2, after applying voltage to the gate-source electrodes (V<sub>GS</sub>), the hole carriers are excited and then drawn from the electrodes into the gate dielectric channel of the composite material, where with the increase of (-V<sub>GS</sub>), the output voltage (I<sub>DS</sub>) increases, until it reaches the saturation regime, where V<sub>GS</sub>-V<sub>th</sub> = V<sub>DS</sub> (the threshold voltage was indicated by V<sub>th</sub>). In this context, several relationships between the output current (I<sub>DS</sub>) and the applied gate voltage (VGS) assist us in plotting electrical

transfer characteristic curves. For example, by replacing the charge relationship with the dielectric capacity in the form of the below relationship, we can find carrier charges in stated capacitor (Q):

$$Q = C_i \frac{V_{DS} - V_{th}}{WL} \tag{1}$$

where C<sub>i</sub> is the capacitance per unit area (W: Width, L: Length of the channel between source and drain electrodes).

To obtain the mobility of carriers (μ) in the linear regime before the saturation regime, the following relationship can be used [8]:

$$I_{DS} = \mu_{Linear} \frac{WC}{L} (V_{DS} - V_{th})^2 \tag{2}$$

$$\mu = \frac{I_{DS}L}{WQV_{DS}} = \frac{I_{DS}L^2}{C(V_{DS} - V_{th})V_{DS}} \tag{3}$$

and/or in a saturated regime:

$$\mu_{sat} = \frac{2l}{wC} \left( \frac{\partial \sqrt{I_{DS}}}{\partial V_{GS}} \right)^2 \tag{4}$$

Therefore, by drawing the slope of the  $\sqrt{I_{DS}} - V_{GS}$  curves, the carrier mobility can be determined. And finally subthreshold voltage can be estimated from slopping of  $V_{GS} - \text{Log } I_{DS}$  curves and Eq. (5):

$$SS = \frac{dV_{GS}}{d \text{Log } I_{DS}} \tag{5}$$

$C_i$  (which is measured with GPS132A tool) in equations (1-3), shows that there is an inverse relation between  $C_i$  and carrier mobility.

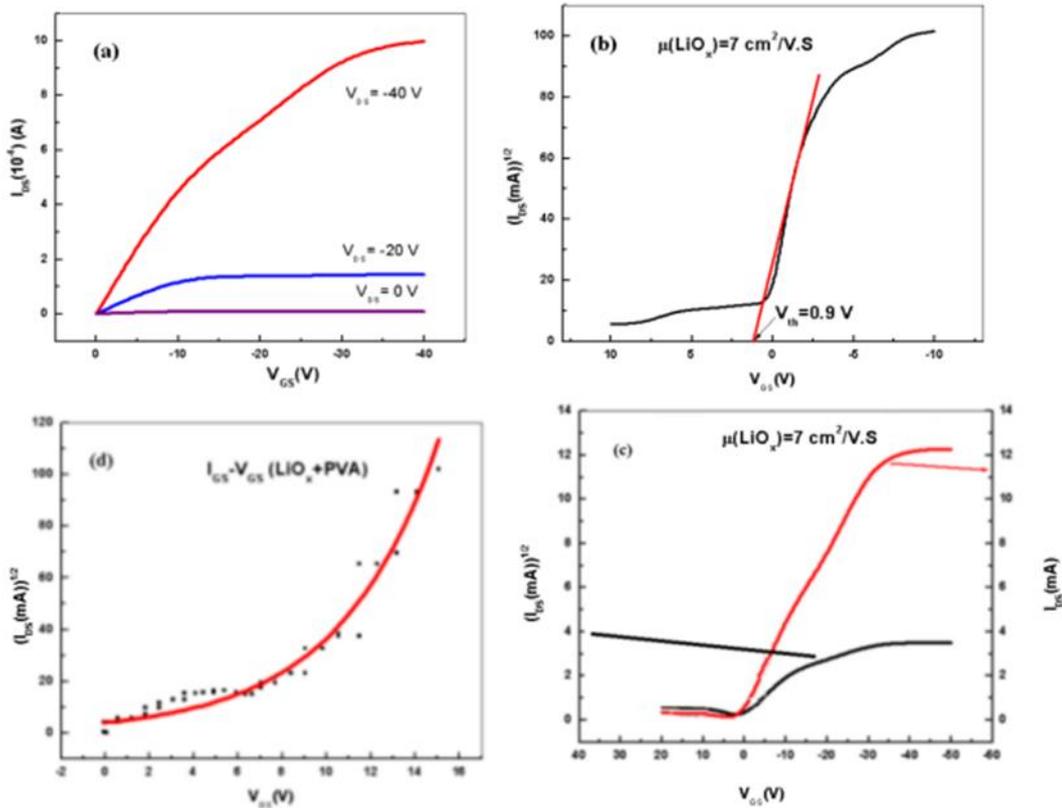
In Figures 3-5, the square root of the source-drain current ( $\sqrt{I_{DS}}$ ) at a constant drain voltage ( $V_{DS}= 20 \text{ V}$ ) of the OFET transistor for NFLiO<sub>x</sub>, PVA and composite gate dielectrics, give us carrier mobility and the threshold

voltage. The logarithm of the output current ( $\text{Log } I_{DS}$ ) determined subthreshold voltage (SS). SS suggesting that how the rate of transition from OFF state to the ON state of the device can be improved with gate dielectric material.

The measured with the help of Prova IV measuring device and plotting the measurement results;  $I_{DS}-V_{GS}$ ,  $(I_{DS})^{1/2}-V_{GS}$ ,  $\text{Log } I_{DS}-V_{GS}$  and shown in Figures 2-5. It specified the transition and output characteristics of OFET.

In addition to the above parameters, the ratio of ON to OFF current ( $I_{on}/I_{off}$ ) or the ability of the transistor to change from ON to OFF current values has been estimated by considering the maximum output current,  $(I_{DS})_{Max}$  in the accumulated layer to its minimum in the not accumulated layer, of- current,  $(I_{DS})_{Min}$  in the  $I_{DS} - V_{GS}$  curve.

Figs. 3-5 display the findings, and Table 2 lists them. By comparing the findings of the present work and the findings of other researchers in this area, presented in the Table 3, it is clear that NFLiO<sub>x</sub>/PVA nano composite has not only acceptable and desirable values for being gate dielectric material of OFET devices, but also it is biocompatible due to the use of water solvent.



**Fig. 2.** (a) exhibits the effect of applied voltages of  $V_{DS}= 0, 20, 40 \text{ V}$  on the electrical transfer characteristics. (b) shows the carrier mobility and threshold voltage. (c) The output electric current ( $I_{DS}$ ) and  $\sqrt{I_{DS}}$  in terms of applied voltage to the aluminum gate ( $V_{GS}$ ) of the OFET transistor. (d) shows the effect of applied voltage on the source-drain (at a constant voltage of  $V_{DS}= 20 \text{ V}$ ), for the NFLiO<sub>x</sub> gate dielectric.

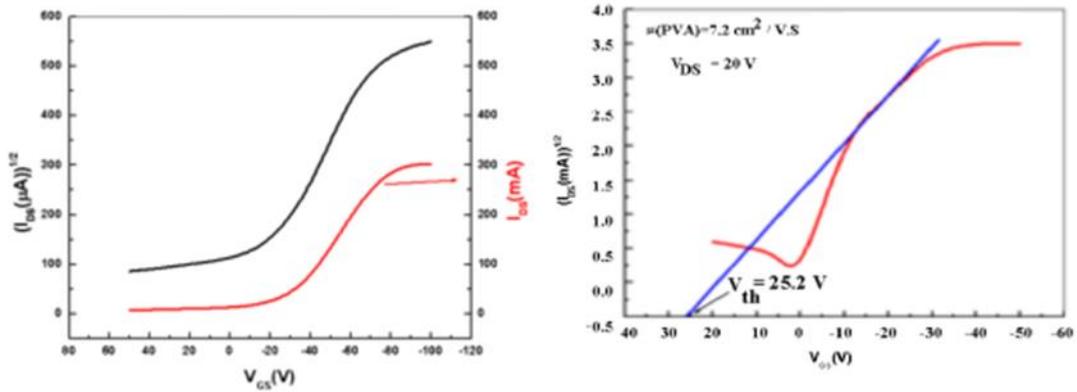


Fig. 3. (Left): The output electric current and its root in terms of applied voltage to the aluminum gate of the OFET transistor. (Right): The curve shows the carrier mobility and threshold voltage for the PVA gate dielectric.

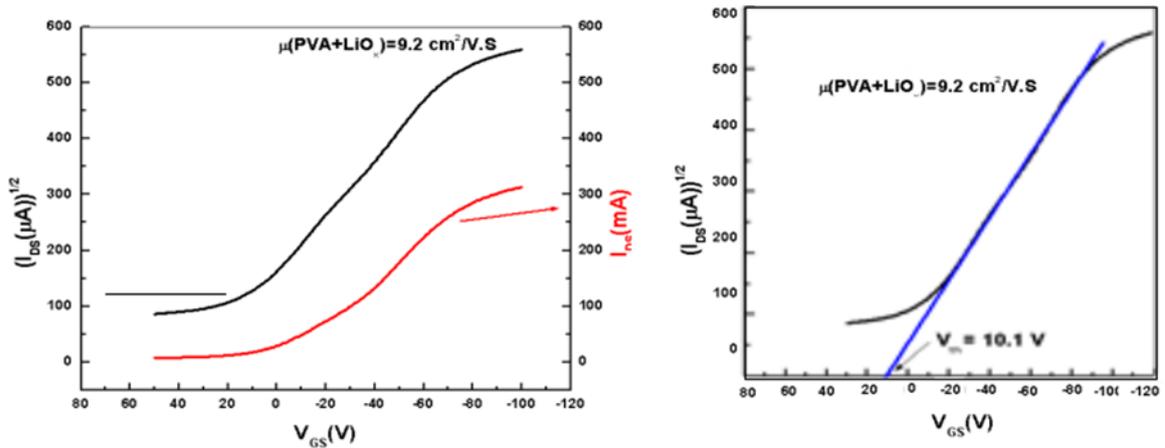


Fig. 4. (Left): The output electric current and its root in terms of applied voltage to the aluminum gate of the OFET transistor. (Right): The curve shows the electrical transfer characteristic of the carrier mobility and the threshold voltage for the gate dielectric of the PVA+ NFLiO<sub>x</sub> nanocomposite.

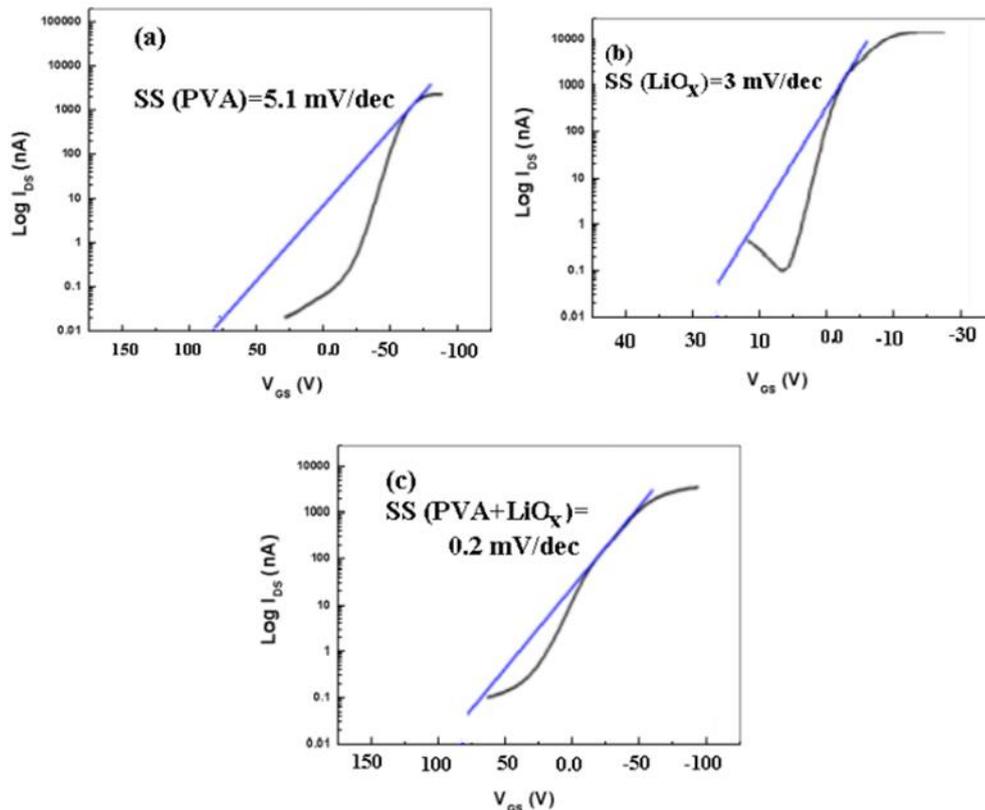


Fig. 5. The logarithm of the output electric current in terms of the applied voltage to the aluminum gate (at a source-drain voltage of 20 V) of the OFET transistor gives the SS in terms of mV/decade (mV/dec.) of (a): PVA, (b): NFLiO<sub>x</sub>, and (c): NFLiO<sub>x</sub>+PVA according to equation (3).

**Table 2.** Measured values of carrier mobility, threshold and subthreshold voltage, ratio of ON to OFF current with 300  $\mu\text{m}$  length and width of OFET transistor channel.

Gate dielectric	$C_i$ (F/ $\text{m}^2$ )	$\mu$ ( $\text{cm}^2/\text{V}\cdot\text{s}$ )	SS (mV/dec.)	$V_{th}$ (V)	$I_{on}/I_{off}$ ( $10^5$ )
NFLiO <sub>x</sub>	17	7.0	3.0	0.9	1.2
PVA	52	7.2	5.1	25	0.3
NFLiO <sub>x</sub> +PVA	22	9.2	0.2	10.1	32.0

**Table 3.** Comparing the findings related to OFET electrical characteristics of other researchers with the findings of the present work.

Device	Dielectric	$\mu$ ( $\text{cm}^2/\text{V}\cdot\text{s}$ )	$V_{th}$ (V)	SS (mV/dec.)	$I_{ON}/I_{OFF}$	Ref.
BCBG	SiO <sub>2</sub>	0.0094				25
BCBG	TiO <sub>2</sub>	0.013	-1			26
BCTG*	PEO:EV(ClO <sub>4</sub> ) <sub>2</sub>	8	-1.87	177	10 <sup>7</sup>	34
BGTC	In <sub>2</sub> O <sub>3</sub> /NFLiO <sub>x</sub>	2.64	-0.16	220	10 <sup>4</sup>	32
TGTC	Ge (Sn)			72	3×10 <sup>6</sup>	32
BGTC	DPP-DTT**	0.184	1.12	186	10 <sup>4</sup>	20
BGTC	InAs/GaSb			60	10 <sup>7</sup>	31
BGTC	povidone/silica	34	0.4		2.2 × 10 <sup>3</sup>	21
BGTC	NFLiO <sub>x</sub>	7	0.9	3	1.2 × 10 <sup>5</sup>	Present work
BGTC	PVA	7.2	25	5.1	3 × 10 <sup>4</sup>	Present work
BGTC	NFLiO <sub>x</sub> / PVA	9.2	10.1	0.2	3.2 × 10 <sup>6</sup>	Present work

\*BCTG: Bottom-contact Top gated, BCBG: Bottom- contact Bottom- gated.

BGTC: Bottom- gate Top -contacted.

\*\* DPP-DTT: self-assembled poly [2,5-(2-octyldecyl)-3,6-diketopyrrolopyrrole-alt-5,5-(2,5di (thien-2-yl)thieno [3,2-b]-thiophene)].

## 4. Conclusions

In the present work, by fabricating three devices of OFET with NFLiO<sub>x</sub>, PVA, and NFLiO<sub>x</sub>/PVA gate dielectric materials and measuring their electrical characteristic parameters, a suitable gate dielectric and eco-friendly material for OFET transistors is found in a biocompatible synthesis method without using any toxic materials. According to the curves and comparing the findings of other researchers with the present work, it has been determined that the NFLiO<sub>x</sub>/PVA gate dielectric material has favorable electrical characteristics: higher carrier mobility ( $\mu$ ) of 9.2  $\text{cm}^2/\text{V}\cdot\text{s}$ , higher carrier mobility ( $\mu$ ) 9.2  $\text{cm}^2/\text{V}\cdot\text{s}$ , higher on-off current ratio  $2.3 \times 10^6$ , minimum threshold voltage ( $V_{th}$ ) equal to 10.1 V and with lower subthreshold swing (SS) around 0.2 mV/dc and also with eco friend characteristic, which can be considered for future OFET devices.

## Acknowledgements

This research was financed by a research grant from the university of Mazandaran.

## Funding Statement

This research received no specific grant from any funding agency.

## Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Authors Contribution Statement

Ali Bahari: Conceptualization, Methodology, Writing—original draft, Formal analysis, Validation, Project administration, Funding acquisition, Investigation, Writing—original draft, Supervision.

Vaheed Falah Hamidabadi: Methodology, Investigation, Abbas Farhadi: Investigation,

Nasrin Moradbeigi: Investigation, Formal analysis.

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