Fábio Fedrizzi Vidor · Gilson Inácio Wirth
Ulrich Hilleringmann

ZnO Thin-Film Transistors for Cost-Efficient Flexible Electronics

Springer

ZnO Thin-Film Transistors for Cost-Efficient Flexible Electronics

Fábio Fedrizzi Vidor • Gilson Inácio Wirth
Ulrich Hilleringmann

ZnO Thin-Film Transistors for Cost-Efficient Flexible Electronics

Springer

Fábio Fedrizzi Vidor
Escola de Engenharia
Federal University of Rio Grande do Sul
Porto Alegre, RS, Brazil

Gilson Inácio Wirth
Escola de Engenharia
Federal University of Rio Grande do Sul
Porto Alegre, RS, Brazil

Ulrich Hilleringmann
Fakultät EIM-E, Sensorik
University of Paderborn
Paderborn, Germany

ISBN 978-3-319-72555-0 ISBN 978-3-319-72556-7 (eBook)
https://doi.org/10.1007/978-3-319-72556-7

Library of Congress Control Number: 2017961120

© Springer International Publishing AG 2018
This work is subject to copyright. All rights are reserved by the Publisher, whether the whole or part of the material is concerned, specifically the rights of translation, reprinting, reuse of illustrations, recitation, broadcasting, reproduction on microfilms or in any other physical way, and transmission or information storage and retrieval, electronic adaptation, computer software, or by similar or dissimilar methodology now known or hereafter developed.
The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.
The publisher, the authors and the editors are safe to assume that the advice and information in this book are believed to be true and accurate at the date of publication. Neither the publisher nor the authors or the editors give a warranty, express or implied, with respect to the material contained herein or for any errors or omissions that may have been made. The publisher remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Printed on acid-free paper

This Springer imprint is published by Springer Nature
The registered company is Springer International Publishing AG
The registered company address is: Gewerbestrasse 11, 6330 Cham, Switzerland

Mas não basta pra ser livre
Ser forte, aguerrido e bravo
Povo que não tem virtude
Acaba por ser escravo

(Hino Rio-Grandense)

Preface

This work would never be possible without the guidance and support of many people. Therefore, before we start with the book itself, we would like to express our sincere thanks to everybody who was involved.

We would like to start thanking the Sensor Technology Department from Paderborn University, Germany, from the former to the current researchers and staff for the support and for the excellent working environment. Special thanks to Thorsten Meyers, who has considerably contributed to the developing and revision of the work presented in this book, and to Julia Reker, Thomas Hett, Dmitry Petrov, Marcel Schönhoff, André Kleine, Sabine Schleghuber, Sebastian Lappe, Werner Büttner, Karsten Wolff, Fabian Assion, Torsten Frers, Benjamin Ohms, Jumir Vieira de Carvalho, and Sebastian Meyer zu Hoberge. Conjointly, we must highlight the established TFT team, whose participants have contributed to this work in diverse ways during the last years. To all involved students, especially Kathrin Müller and Leon Lenk, we are deeply grateful for your work. For the support with SEM images and PL measurements, we are indebted with the groups AG Lindner and AG Meier from Paderborn University, respectively.

We extend our acknowledgments to LAPROT (Prototyping and Testing Laboratory) from UFRGS (Federal University of Rio Grande do Sul), Brazil, specially Thiago Hanna Both, who has carefully read this book contributing with suggestions and insights. Moreover, we would like to express our gratitude to our friends in Brazil and in Germany for their friendship over the years, specially to Fernanda and Guilherme, Tarek, Erika, Lizoel and his family, Lara and Jonas, and the people from the P8-building (Paderborn University).

We are really grateful to the cooperation CAPES/CNPq/DAAD (BEX 12399/12-4), FAPERGS (1011092), and DFG (GRK 1464 and HI 551/63-1) for the financial support throughout the project, which was substantial for the accomplishment of this work. Such partnerships are important for widening the horizons for subsequent researches as well as for intensifying the cooperation between Brazil and Germany.

This book is dedicated to our families, whose love and support have been a continual inspiration, especially to Jéssica, who has actively contributed to the book reading the first drafts and employing her designer skills to enhance the figures' quality, and to Humberto and Joceli, Mariana and Helena, and Anja, Vanessa, and Desirée.

Abstract

Flexible and transparent electronics enable the integration of innovative cost-efficient products. One of the outstanding aspects of this technology is its wide range of applications, from flexible and transparent displays to wearable electronics and RFID (radio-frequency identification) tags for sensor networks employed, for instance, in health monitoring systems. In this area, thin-film transistors (TFTs) are the key elements which drive the electrical currents in the devices. Conjointly, hybrid systems, combining high-performance silicon-based transistors for data processing and thin-film transistors for enhanced user interactivity, emerge profiting from the synergy of both technologies.

In this study, ZnO-based TFTs for flexible and transparent electronics were integrated and characterized. The fabrication processes were limited to cost-efficient and low-temperature methods compatible to large-area flexible substrates; therefore, solution-based techniques were primarily applied. For the active semiconductor, ZnO precursors and dispersions containing nanostructures of the material were evaluated, the latter depicting better compatibility with the integration process as well as higher performance and reliability. As gate dielectric, poly(4-vinylphenol) (PVP) and a high-*k* nanocomposite were employed. The transistors were structured in both inverted staggered and inverted coplanar setups. On the one hand, the staggered structures depict larger contact area between the drain/source electrodes and the active semiconducting layer, hence higher charge carrier injection. On the other hand, their coplanar counterparts profit from the late semiconductor deposition, which enables an effective analysis of the instabilities concerning the transistor. To investigate the performance metrics and reliability issues, an extensive characterization of the transistors was performed. After the main instability effects were identified and mitigated, the TFTs were also integrated on polymeric substrates. Aiming at the fabrication of compact and energy-efficient devices, optical photolithography was used for layer patterning instead of shadow mask technique. Along with the resolution of around 1 μm achieved for multiple-layer definition, the employment of freestanding PET substrates reproduces a more realistic scenario for a later large-scale production. Different methods, namely, spin- and spray-coating and doctor blade technique, for the semiconductor dispersion deposition were investigated, leading only to minor variations on the TFT electrical performance. The metrics of the integrated ZnO nanoparticle TFTs are among the best reported for nanoparticle-based transistors up to date. Additionally, they are comparable to those of TFTs fabricated using cost-intensive techniques or high-temperature processes.

In order to evaluate the ZnO TFTs in electronic circuit applications, inverters employing an active transistor in the pull-down network and a load transistor in the pull-up network were integrated on rigid and on flexible substrates. Furthermore, the dynamic characteristics of such inverters were analyzed in ring oscillator circuits. Finally, by an adaptation of the photolithography, self-alignment processes were used to reduce the transistor's parasitic capacitances as well as to pattern the semiconducting layer in order to avoid cross-talk effect between devices. Furthermore, a complementary design using *n*-type inorganic and *p*-type organic TFTs is evaluated.

Contents

List of Symbols, Abbreviations, and Acronyms

Symbols

A^{**}	Effective Richardson constant
C	Capacitance
C_{ins}	Gate capacitance per unit area
C_{G}	Effective gate capacitance per unit area
C_{sem}	Capacitance of the semiconductor per unit area
d_{T}	Threshold depth
f	Oscillation frequency
e, e^{-}	Electron, elementary charge (1.602×10^{-19} C)
$\mathscr{E}$	Electric field
E_{C}	Conduction band
E_{F}	Fermi level
E_{V}	Valence band
g_{m}	Transistor transconductance
h	Thickness of the accumulation layer in the semiconductor
$I_{\mathrm{DS}}, I_{\mathrm{D}}$	Drain-source current
$I_{\mathrm{GS}}, I_{\mathrm{G}}$	Gate-source current
I_{OFF}	Transistor off-state current
I_{ON}	Transistor on-state current
J	Current density
J_{o}	Saturation current density
k	Boltzmann constant
k	Relative permittivity
L	Channel length of the transistor
n	Charge carrier density
N	Number of inverters in the chain of the ring oscillator circuit
NM	Noise margin of inverter circuits
NM_{L}	Noise margin for low levels
NM_{H}	Noise margin for high levels

q	Elementary electric charge ($1.602 \cdot 10^{-19}$ C)
Q	Total electric charge
R_a	Roughness average
R_C	Contact resistance
R_D	Contact resistance at the drain electrode
R_S	Contact resistance at the source electrode
S	Subthreshold swing
T	Temperature
T	Period of the oscillation
t_p	Propagation time
t_ins	Thickness of the insulator
t_int	Position of the semiconductor/gate dielectric interface
t_sem	Thickness of the semiconductor
V	Applied voltage
V_DD	Supply voltages
$V_\mathrm{DS}, V_\mathrm{D}$	Drain-source voltage
$V_\mathrm{GS}, V_\mathrm{G}$	Gate-source voltage
V_IH	Input high voltage of inverter circuits
V_IL	Input low voltage of inverter circuits
V_IN	Input voltage
V_OH	Output high voltage of inverter circuits
V_OL	Output low voltage of inverter circuits
V_ON	Turn-on voltage
V_OUT	Output voltage
V_T	Threshold voltage
W	Channel width of the transistor
β	Geometry ratio of inverter circuits
W_{ϕ_Bn}	Width of the metal-semiconductor barrier
$\Delta\phi$	Image-forced lowering of the barrier height
ϵ_0	Vacuum permittivity
ϵ_ins	Relative permittivity of the insulator
ϵ_s	Relative permittivity of the semiconductor
η	Ideality factor
λ	Wavelength
μ	Charge carrier mobility
μ_avg	Average mobility
μ_eff	Effective mobility
μ_FE	Field-effect mobility
μ_inc	Incremental mobility
μ_sat	Saturation mobility
ϕ_Bn	Barrier height on a n-type semiconductor
ϕ_m	Work function of a metal
χ	Electron affinity

Chemical Elements and Compounds

Ag	Silver
Al	Aluminum
AlN	Aluminum nitride
Al_2O_3	Aluminum oxide
Ar	Argon
Au	Gold
Cd	Cadmium
CdS	Cadmium sulfide
CH_3-COOH	Acetic acid
Cl	Chlorine
CH_3F	Fluoromethane
Cu	Copper
Ga	Gallium
GaAs	Gallium arsenide
GaZnO	Gallium zinc oxide
He	Helium
HNO_3	Nitric acid
H_2O	Water molecule
H_2O_2	Hydrogen peroxide
H_3PO_4	Phosphoric acid
I	Iodine
In	Indium
In_2O_3	Indium oxide
InGaO	Indium gallium oxide
InGaZnO	Indium gallium zinc oxide
InZnO	Indium zinc oxide
NaOH	Sodium hydroxide
N_2	Nitrogen
NH_4F	Ammonium fluoride
NH_4OH	Ammonium hydroxide
OH^-	Hydroxide
O_2	Oxygen
Si	Silicon
a-Si	Amorphous Si
a-Si:H	Hydrogenate amorphous silicon
poly-Si	Polycrystalline silicon
SiO	Silicon monoxide
SiO_2	Silicon dioxide
Sn	Tin
SnO_2	Tin dioxide
Ti	Titanium
TiO_2	Titanium dioxide

V_o	Oxygen vacancies
Zn	Zinc
Zn_i	Zinc interstitials
$Zn(Ac)_2$	Zinc acetate
$Zn(NO_3)_2$	Zinc nitrate
ZnO	Zinc oxide
$ZnO \cdot xH_2O$	Zinc oxide hydrate
$Zn(OH)_2(NH_3)_x$	Ammine-hydroxo zinc
ZnSnO	Zinc tin oxide

Abbreviations and Acronyms

2-ME	2-Methoxyethanol
ALD	Atomic layer deposition
ALU	Arithmetic logic unit
AMOLED	Active-matrix organic light-emitting diode
AZO	Aluminum zinc oxide
C_8–BTBT	2,7-Dioctyl[1]benzothieno[3,2-b][1]benzothiophene
CCD	Charge-coupled device
CYTOP	Poly(perfluorobutenylvinylether)
D	Drain electrode
DLE	Deep-level emission
DNTT	dinaphtho[2,3-b:2,3-f]thieno[3,2-b]thiophene
FE	Field-emitted charge carriers
GIZO	Indium gallium zinc oxide
GZO	Gallium zinc oxide
HOMO	Highest occupied molecular orbital for organic materials
HVPE	Hydride or halide vapor-phase epitaxy
IEEE	Institute of Electrical and Electronics Engineers
IGO	Indium gallium oxide
IoE	Internet of Everything
IoT	Internet of Things
ITO	Indium tin oxide
IZO	Indium zinc oxide
LCD	Liquid-crystal display
LUMO	Lowest unoccupied molecular orbital for organic materials
MBE	Molecular-beam epitaxy
MOCVD	Metal-organic chemical-vapor deposition
MOSFET	Metal-oxide-semiconductor field-effect transistor
NBE	Near-band edge emission
NMP	N-Methyl-2-pyrrolidone
PC	Polycarbonate
PDMS	Polydimethylsiloxane

PECVD	Plasma-enhanced chemical-vapor deposition
PEEK	Polyether ether ketone
PEN	Polyethylene naphthalate
PET	Polyethylene terephthalate
PES	Polysulfone
PGMEA	Propylene glycol methyl ether acetate
PI	Polyimide
PL	Photoluminescence
PLD	Pulsed-laser deposition
PMCF-m	Poly(melamine-co-formaldehyde)-methylated
PMMA	Poly(methylmethacrylate)
PP	Polypropylene
PS	Polystyrene
PTFE	Poly(tetrafluoroethene)
PVA	Poly(vinylalcohol), polyvinyl alcohol
PVC	Polyvinyl chloride
PVP	Poly(4-vinylphenol)
RF	Radio-frequency
RFID	Radio-frequency identification
RH	Relative humidity
RIE	Reactive ion etching
RTS	Random telegraph signal
S	Source electrode
SRAM	Static random-access memory
TE	Thermionic-emitted charge carriers
TFE	Thermionic-field-emitted charge carriers
TFT	Thin-film transistor
UV	Ultraviolet
UV-Vis	Ultraviolet-visible
VLSI	Very-large-scale integration
VTC	Voltage transfer characteristic
ZTO	Zinc tin oxide

Chapter 1
Introduction

Products and applications using transparent and flexible electronics are widely connected to a futuristic scenario. They were explored by novels, such as the *Shape of Things to Come* from H.G. Wells in [TSTC33], and by films, such as "Barbarella" [Bar68] based on Jean-Claude Forest's comics in 1968. The animation studio Hanna-Barbera also explored some aspects of future daily artifacts through "the Jetsons" [TJ60] cartoons in the 1960s and 1980s. Some of the recent Hollywood productions, such as Minority Report ([MR02]) and Ironman ([IM08], [IM10], and [IM13]), also give us insights of future applications for flexible and transparent electronics and how they can be integrated in our lives. These futuristic visions and ideas motivate the scientific community as well as companies to develop and to employ this technology.

Flexible and transparent electronics enable the fabrication of innovative products making use of different aspects of the applied materials and compounds. Some concepts of applications which take advantages of these characteristics are shown in Fig. 1.1. The integration of transparent displays, for example, increases the interactivity of the user with the surrounding environment. Moreover, by employing a flexible substrate, integrated sensor networks can be used as wearable electronic skins enabling, for instance, collection and analysis of body functions for sports and medical applications. The food industry can also profit from this technology: cost-efficient radio-frequency identification (RFID) tags can be employed to monitor food quality and storage conditions in real time. For these applications, thin-film transistors (TFTs) are commonly used as active circuit elements. The advantages of this type of transistors are the integration process almost independent of the substrate (generally used just as mechanical support) and the opportunity to apply a wide range of materials in its structure. Nevertheless, the TFT technology is not to be seen as a substitute for crystalline silicon (Si)-based transistors in the high-performance market. This technology should act promoting new products and applications, being implemented in most cases in hybrid systems, to improve data acquisition and user interface.

F.F. Vidor et al., *ZnO Thin-Film Transistors for Cost-Efficient Flexible Electronics*,
https://doi.org/10.1007/978-3-319-72556-7_1

Fig. 1.1 Concept applications of transparent and flexible electronics in the field of displays, sensor networks for medical monitoring and for sport activity, and RFID tags for the food industry

As this technology avails a wide range of products, the development of new systems itself can be time- and cost-intensive; one of the main issues is dealing with the feasibility and with the complexity of the whole project. Therefore, the use of abstraction levels leads to a more effective development of the technology. Figure 1.2 shows an example of a schematic design of abstraction levels for transparent and flexible systems. In this particular case, the main focus has been given to its electronic part; nevertheless the same method can be also directed to the mechanical and aesthetic aspects of the product. Each abstraction level can use the previous one as a black box or as a model avoiding unnecessary internal complexity and focusing on the developing of the current level elements. In the example of Fig. 1.2, the system was divided into different levels in order of increasing abstraction: materials, devices, circuits, modules, and the system itself. The focus of this study is placed in the device level; however, a merge with both frontiers (materials and circuits) is also covered.

The purpose of this book is the development of cost-efficient inorganic-based thin-film transistors and circuits on flexible and transparent substrates employing low-temperature processes. Aiming at reduced costs, solution-based materials are preferred for the integration of the TFTs. The use of solution-based techniques fulfills the large area integration and flexible substrates requisites while exhibiting advantages when compared to cost-intensive vacuum-based processes. Among inorganic materials, metal oxides dominate the sector with different compounds and deposition methods, which can be selected depending on the system requirements [WS09, FBM12, PMV$^+$16]. Zinc oxide (ZnO) has emerged as a primary compound in this field, possessing outstanding electrical and chemical characteristics as well as being transparent to the visible light spectrum. Therefore, solution-based processes employing either ZnO precursors or a dispersion containing nanostructures of the

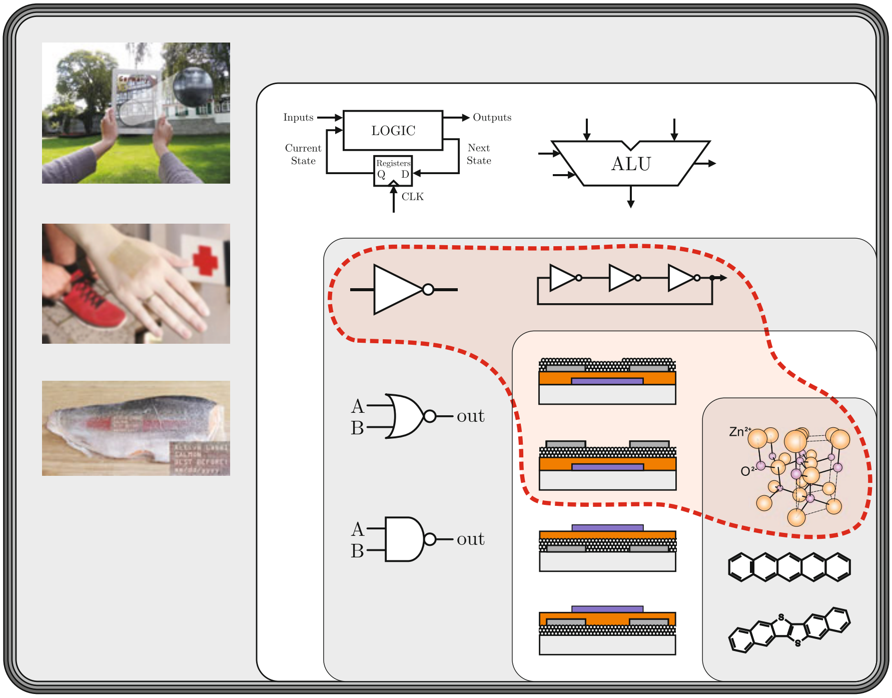

Fig. 1.2 Schematic design of abstraction levels for flexible and transparent systems. The selection in red depicts the levels covered in this book

material are chosen for the formation of the active semiconducting layer. Besides the investigation of different gate dielectric materials as well as deposition methods for the semiconductor, it is also an objective of this study to identify and minimize the agents responsible for instability effects in the transistor operation. Further studies should be conducted to analyze the TFTs characteristics upon different transistor structures on either rigid (oxidized Si or glass wafer) or on polymeric substrates. Moreover, the integration and evaluation of the transistors in digital circuits, e.g., inverter circuits and ring oscillators, are also sought.

1.1 Structure of the Work

Initially, the fundamentals concerning thin-film transistors and flexible electronics are presented in Chap. 2. The discussion comprises the intrinsic attributes of the active semiconducting material employed in this study; important aspects related with TFTs, such as its history and a comparison with high-performance Si-based transistors; as well as their operation properties, modeling, and electrical characterization.

Chapter 3 is devoted to the integration process of the transistors on rigid and on flexible substrates. Therefore, TFT basic structures and each of its components are addressed. Although the focus is given to the processes and methods applied in this study, a general overview of the most used techniques and materials found in the literature is given.

The electrical characterization of the TFTs is mainly presented in Chap. 4. Transistors integrated employing ZnO precursor or nanoparticles as well as a discussion concerning the electron flow mechanism in the nanoparticulated semiconducting film and its effect on the transistor's current are described. Along with the I–V curves, qualitative models representing the TFT behaviors are given and analyzed.

Chapter 5 addresses the performance of circuits integrated employing the ZnO-based TFTs. Whereby, inverter circuits integrated using load-transistors in the pull-up network and active-transistors in the pull-down network on rigid and on flexible substrates are analyzed. Additionally, their dynamic characteristics (ring oscillator circuits) are evaluated.

Improvements for the integrated circuits and devices are presented in Chap. 6. Approaches such as the reduction of parasitic capacitances and of cross-talk effects, implementation of a complementary design applying both *n*-type ZnO-based TFTs and *p*-type organic-based TFTs, and evaluation of further deposition methods for the active semiconducting material are addressed.

Finally, in Chap. 7, the main conclusions of this book as well as future perspectives for ZnO-based TFTs and flexible electronics are discussed.

References

[Bar68] Barbarella: http://www.imdb.com/title/tt0062711/ (1968)

[FBM12] Fortunato, E., Barquinha, P., Martins, R.: Oxide semiconductor thin-film transistors: a review of recent advances. Adv. Mater. **24**(22), 2945–2986 (2012). http://dx.doi.org/10.1002/adma.201103228

[IM08] Iron Man: http://www.imdb.com/title/tt0371746/ (2008)

[IM10] Iron Man: http://www.imdb.com/title/tt1228705/ (2010)

[IM13] Iron Man: http://www.imdb.com/title/tt1300854/ (2013)

[MR02] Minority Report: http://www.imdb.com/title/tt0181689/ (2002)

[PMV$^+$16] Petti, L., Münzenrieder, N., Vogt, C., Faber, H., Büthe, L., Cantarella, G., Bottacchi, F., Anthopoulos, T.D., Tröster, G.: Metal oxide semiconductor thin-film transistors for flexible electronics. Appl. Phys. Rev. **3**(2), 021303 (2016). http://dx.doi.org/10.1063/1.4953034

[TJ60] The Jetsons: http://www.imdb.com/title/tt0055683/ (1960s and 1980s)

[TSTC33] The Shape of Things to Come: Wells's Novel, 1st edn. Hutchinson & Co., London (1933). https://www.penguin.co.uk/books/60361/the-shape-of-things-to-come/ or https://en.wikipedia.org/wiki/The_Shape_of_Things_to_Come

[WS09] Wong, W.S., Salleo, A.: Flexible Electronics: Materials and Applications. Springer, New York (2009). ISBN: 978–0–387–74362–2. http://www.springer.com. Electronic Materials: Science & Technology