University of Groningen

Organic Materials in Silico

Sami, Selim

DOI:
[10.33612/diss.146910127](https://doi.org/10.33612/diss.146910127)

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2020

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):
Sami, S. (2020). *Organic Materials in Silico: From force field development to predicting dielectric properties*. [Thesis fully internal (DIV), University of Groningen]. University of Groningen.
<https://doi.org/10.33612/diss.146910127>

Copyright

Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: <https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment>.

Take-down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): <http://www.rug.nl/research/portal>. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.

Organic Materials in Silico

*From Force Field Development
to Predicting Dielectric Properties*

Selim Sami

university of
 groningen

faculty of science
 and engineering

stratingh institute
 for chemistry

Nederlandse Organisatie voor Wetenschappelijk Onderzoek

DIFFER

Dutch Institute for
 Fundamental Energy Research

This project was carried out in the research groups Theoretical Chemistry and Chemistry of (Bio)organic Materials and Devices of the Stratingh Institute for Chemistry and Zernike Institute for Advanced Materials of the University of Groningen, the Netherlands. This work is part of the research programme of the Foundation of Fundamental Research on Matter (FOM), which is part of the Netherlands Organisation for Scientific Research (NWO). This is a publication of the FOM-focus Group 'Next Generation Organic Photovoltaics', participating in the Dutch Institute for Fundamental Energy Research (DIFFER).

An electronic version of this dissertation is available at:

<http://www.rug.nl/research/portal>

Printed by Ridderprint, www.ridderprint.nl

Cover design by Randy Wind, Native Development

Thesis design by Riccardo Alessandri

Copyright © 2020 Selim Sami

university of
 groningen

Organic Materials in Silico

From Force Field Development
to Predicting Dielectric Properties

PhD thesis

to obtain the degree of PhD at the
University of Groningen
on the authority of the
Rector Magnificus Prof. C. Wijmenga
and in accordance with
the decision by the College of Deans.

This thesis will be defended in public on

Friday 11 December 2020 at 11:00 hours

by

Selim Sami

born on 8 October 1990
in Şişli, Turkey

Supervisors

Prof. R. Broer

Prof. J.C. Hummelen

Co-supervisor

Dr. R.W.A. Havenith

Assessment Committee

Prof. J.N. Harvey

Prof. E.L. von Hauff

Prof. P.R. Onck

To my family, for their unwavering support.

Contents

1	Introduction	1
1.1	Climate change and the road to carbon neutrality	2
1.2	Photovoltaics	5
1.3	Organic electronics	8
1.4	High dielectric constant organic electronics	12
1.5	Multiscale Modeling of organic electronics	14
1.6	Aim and Outline of this Thesis	17
2	First-Principles Computation of the Electronic Dielectric Constant for Organic Semiconductors	21
2.1	Introduction	22
2.2	Results and Discussion	27
2.2.1	Method validation on ethylene carbonates.	27
2.2.2	Obtaining accurate densities for fullerene derivatives	28
2.2.3	Dielectric constant of fullerene derivatives.	29
2.3	Conclusions.	31
2.4	Theory	32
2.5	Methods	33
2.6	Appendix: Method Details	35
3	Q-Force: Quantum Mechanically Augmented Molecular Force Fields	41
3.1	Introduction	42
3.2	Methods	46
3.3	Results and discussion	54
3.4	Conclusions.	61

4	How Ethylene Glycol Chains Enhance the Dielectric Constant of Organic Semi-conductors: Molecular Origin and Frequency Dependence	63
4.1	Introduction	64
4.2	Results and Discussion	66
4.3	Conclusions.	70
4.4	Methods	71
4.5	Appendix: Parametrization and validation of the force field.	74
5	Strategies for Enhancing the Dielectric Constant of Organic Materials	81
5.1	Introduction	82
5.2	Results and discussion	85
5.3	Conclusions.	91
5.4	Methods	92
5.5	Appendix: Parametrization and validation of the force fields	93
6	Resolving the Crystal Structure in Thin Films of Fullerene Derivatives Functionalized with Ethylene Glycol Side Chains	99
6.1	Introduction	100
6.2	Results and discussion	102
6.2.1	Case study 1: Fullerene derivatives with ethylene glycol side chains as n-type organic thermoelectrics	102
6.2.2	Case study 2: Charge transport and structure in thin films of fullerene derivatives with ethylene glycol side chains	106
6.3	Conclusions.	113
6.4	Computational methods	114
7	Outlook	119
	Bibliography	123
	Summary	143
	Samenvatting	147
	Curriculum Vitæ	151
	Acknowledgments	155