



Cairo University

CURRENT FLOW AT THE INTERFACE OF COPPER AND POLYETHYLENE IN HIGH-VOLTAGE APPARATUS

By

Eng. Mohammed Ahmed El-Shahat Abo-Saleh

A Thesis Submitted to the
Faculty of Engineering at Cairo University
In partial Fulfillment of the
Requirements for the Degree of

DOCTOR OF PHILOSOPHY
In
Electrical Power and Machines Engineering

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Prof. Dr. Hussein I. Anis

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Title of Thesis:

Current flow at the interface of copper and polyethylene in high-voltage apparatus.

Key Words:

Polyethylene – copper – interface –barrier to injection – dielectric constant.

Summary:

Computational quantum mechanics in the framework of density functional theory (DFT) is used to develop the interface model between Cu (111) and PE (001). The “bulk plus band line up” method and the projected density of states (PDOS) analysis are combined to calculate the actual barrier to charge injection. This combination of methods lead to finding new states appearing between the Fermi level of Cu and the conduction band of PE, which lowered the barrier to electrons injection. These states are due to morphological deformation that takes place in both the metal and the polymer at the interface. The most common chemical impurities that are normally found in PE are studied in this thesis to understand their impact on conduction mechanism. These impurities, such as carbonyl, vinyl, and conjugated double bond, produce trap states between the conduction and valence bands of PE, which reduce the barrier height to around the experimental value of 1 eV. The lowered barrier height increases the charge injection, which then facilitates the conduction mechanism. Variations in the dielectric constant at the interface are also a significant factor. The present study investigates the change in dielectric constant of PE at the interface with Cu using the microscopic polarization theory. It is concluded that the calculated current density using Schottky injection mechanism under a certain electric field and temperature does not only depend on the barrier height, but also on the relative dielectric constant at the interface, which in turn changed the current density and the conduction process.

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LIST OF SYMBOLS

λ	Trap separation
q	Electronic charge
E	Electric field
J	Current density
A	Richardson constant
T	Temperature
V_b	Barrier to charge injection height
K	Boltzmann's constant
ϵ	Dielectric constant
ϵ_0	Free space dielectric constant
ϵ_r	Relative dielectric constant
h	Plank's constant
m_T	Tunneling effective mass
E_a	Electron affinity
a, b, c	Lattice constant
E_g	Band gap
V_b^e	Electrons barrier height
V_b^h	Holes barrier height
WF	Work function
m	Particle mass
∇^2	Laplacian operator
Ψ	Wavefunction
E_T	Total energy
E_p	Potential energy
\hat{H}	Hamiltonian operator
E_k	kinetic energy
Z	Charge number
r_i	Locations of electron i
R_j	Locations of nucleus j
$\delta(\sigma_i, \sigma_j)$	Kronecker-Delta function
H_{HF}	Hartree-Fock Hamiltonian
E_{P_x}	Exchange potential
$E_{P_{Hartree}}$	Hartree potential
E_{P_c}	Correlation potential
$n(r)$	Electron density
$E_{P_{ext}}$	External potential energy
$F[n]$	Global functional
E_{xc}	Exchange-correlation energy
H_{KS}	Khon and Sham Hamiltonian
$E_{P_{KS}}$	Effective potential of KS
ϕ_k	Basis set functions
C_k	Constant coefficients
L	Size of a basis sets
r_c	Cutoff radius

E_{vac}	Vacuum level
E_{f}	Fermi level
\mathbf{P}	Polarization
ρ_{ind}	Induced charge density
E_{loc}	Localized field
E_{ext}	External field

LIST OF ABBREVIATIONS

PDOS	Projected density of states
DFT	Density Functional Theory
CBM	Conduction band minimum
VBM	Valence band maximum
QE	Quantum Espresso
PE	Polyethylene
Cu	Copper
Ag	Silver
Al	Aluminum
Au	Gold
Pt	Platinum
Pd	Palladium
Si	Silicon
SiO₂	Silicon oxide
SCLC	Space-Charge-Limited Conduction
XPS	X-ray Photoelectron Spectroscopy
EELS	Electron Energy Loss Spectroscopy
LDA	Local density approximation
GGA	Generalized gradient approximation
HSE	Heyd-Scuseria-Ernzerhof
HF	Hybrid functionals
MCSCF	Multi configuration self-consistent field
CI	Configuration interaction
CC	Coupled cluster
MP	Perturbation theory by Møller and Plesset
HK	Hohenberg and Kohn
KS	Kohn and Sham
SCF	Self-consistent field
LCAO	Linear Combination of Atomic Orbitals
PW	Plane Waves
AS	Atomic Sphere
CG	Conjugate gradient
BFGS	Broyden- Fletcher-Goldfarb-Shanno
MD	Molecular Dynamics
DOS	Density of states
GDIIS	Geometric direct inversion in the iterative subspace
CP	Car-Parrinello
PWSCF	Plane-Wave Self-Consistent Field
PAW	Projector Augmented Waves
PBE	Perdew-Burke-Ernzerhof
FCC	Face centered cubic
PEA	Pulsed electro-acoustic

ABSTRACT

Polymers are increasingly becoming the insulator of choice in various high voltage power apparatus, such as capacitors and cables. Under high operating fields conduction (leakage) currents in polymers contribute to the eventual breakdown of the insulator, thus determining its active life time. A barrier to charge injection at polymer/metal interface is a key to understanding the high field conduction in cables and supercapacitors. The conduction mechanism of injected currents is influenced by barriers to charge injection and also by possible variations in the dielectric constant at the interface. The present work investigates barriers to charge injection at the atomic level at the interface of copper and polyethylene (PE), one of the most dominant material combinations in the power industry. Emphasis is carried out in this work on morphological deformation.

Computational quantum mechanics in the framework of density functional theory (DFT) is used to develop the interface model between Cu (111) and PE (001) in the light of previous DFT studies on the interface of PE with Pt, Au, and Ag in terms of the absolute barriers to holes and electrons injection. The “bulk plus band line up” method and the projected density of states (PDOS) analysis are combined to calculate the actual barrier to charge injection, which was not well identified in similar studies. This combination of methods lead to finding new states appearing between the Fermi level of Cu and the conduction band of PE, which lowered the barrier to electrons injection. These states are due to morphological deformation that takes place in both the metal and the polymer at the interface. The present work stresses the importance of considering morphological deformation in lowering the barriers to charge injection in soft metal which is less considered as compared to chemical impurities.

The most common chemical impurities that are normally found in PE are studied in this thesis to understand their impact on conduction mechanism. These impurities, such as carbonyl, vinyl, and conjugated double bond, produce trap states between the conduction and valence bands of PE, which reduce the barrier height to around the experimental value of 1 eV. The lowered barrier height increases the charge injection, which then facilitates the conduction mechanism.

This work recognizes the fact that the barrier height is not the sole factor affecting conduction. Variations in the dielectric constant at the interface are also a significant factor. The present study investigates the change in dielectric constant of PE at the interface with Cu using the microscopic polarization theory. It is concluded that the calculated current density using Schottky injection mechanism under a certain electric field and temperature does not only depend on the barrier height, but also on the relative dielectric constant at the interface, which in turn changed the current density and the conduction process.