ION-BEAM ENGINEERING AS A PHYSICAL STRATEGY FOR ELECTROCHEMICAL MATERIALS DESIGN

DOI: 10.66951/GFAW9691; https://doi.org/10.66951/GFAW9691

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Abstract

Ion-beam modification represents a versatile physical strategy for tailoring the electrochemical properties of materials without the need for conventional chemical functionalization. This work demonstrates the potential of multi-energy Cu and Ag ion implantation for engineering two distinct

classes of energy storage materials: polymer interfaces and multilayer graphene. A combination of ion-beam simulations, structural and chemical characterization, theoretical calculations, and electrochemical analyses revealed that implantation induces controlled defect formation, local bond rearrangement, and stabilization of implanted atoms, resulting in modified electrochemical behavior. In polymer materials, these changes enhanced interfacial polarization while preserving the predominantly insulating character of the matrix. In multilayer graphene, ion implantation introduced controlled structural modifications, affected the interlayer arrangement, and tuned the electrochemical response of the anode material. The results demonstrate that multi-energy ion implantation provides a transferable physical approach for engineering electrochemical materials and highlights its potential as an alternative to conventional chemical modification strategies for future energy storage applications.

Introduction

The development of advanced materials for electrochemical and energy storage applications is commonly based on chemical functionalization, which enables tuning of structural, electronic, and interfacial properties through the introduction of new functional groups or chemically active species [1]. Although highly effective, these approaches are often material-specific, require multistep synthesis procedures, and may be difficult to transfer between different classes of materials [1].

Ion-beam modification offers an alternative physical strategy for tailoring material properties without altering the overall architecture of the material. During ion implantation, energetic ions transfer energy to the target through electronic excitation and nuclear collisions, inducing controlled defect formation, bond rearrangement, local structural modifications, and stabilization of implanted atoms [2]. Unlike conventional chemical methods, these processes can be achieved using the same implantation approach across chemically distinct materials while preserving their macroscopic integrity [2,3].

In this work, the potential of multi-energy Cu and Ag ion implantation is demonstrated using two representative classes of electrochemical materials: polymer interfaces and multilayer graphene. Although these materials differ fundamentally in their chemical composition, structure, and function, both benefit from controlled ion-beam-induced modification. In polymers, implantation promotes the formation of electrochemically active interfacial regions with enhanced polarization behavior, whereas in graphene it enables controlled structural tuning of the electrode architecture for lithium-ion storage [3,4]. The presented results demonstrate that multi-energy ion implantation represents a versatile physical strategy for engineering electrochemical materials and highlights its potential as an alternative to conventional chemical modification for future energy storage technologies.

4 Experimental

Polyimide (PI), poly (methyl methacrylate) (PMMA), and cyclic olefin copolymer (COC) films (thickness 50 μm) were obtained from Goodfellow (UK) and Polyplastics Co. (Japan). A commercially available graphene sheet (35 μm thick, 10 × 10 cm) supplied by Nanografi (Turkey) was used as a representative multilayer graphene material. Cu^+ and Ag^+ ions were implanted into all materials using a Tandetron 4130 MC accelerator. Multi-energy implantation was performed sequentially at 2.8, 2.0, and 1.2 MeV with ion fluences of 1×10^{12}, 1×10^{13}, and 1×10^{14} cm^{-2}. The beam current density was maintained at 15–45 nA cm^{-2} to minimize beam-induced heating and preserve the integrity of the implanted materials.

The implantation process was designed to produce an extended subsurface modified region by overlapping the ion ranges corresponding to the three implantation energies. Structural, chemical, and electrochemical changes induced by ion implantation were subsequently evaluated using

complementary characterization techniques, including SRIM simulations, DFT calculations, RBS/ERDA, FTIR, XPS, Raman spectroscopy, X-ray diffraction (XRD), electrochemical impedance spectroscopy (EIS), galvanostatic charge–discharge measurements, and electrical characterization, depending on the investigated material.

5 Results and Discussion

5.1 A Common Physical Strategy for Materials Engineering

The first step of this study was to verify whether the same ion-beam approach could be applied to chemically and structurally different electrochemical materials. Stopping and Range of Ions in Matter (SRIM) simulations were employed to model the ion–solid interactions and predict implantation profiles. As shown in **Figure 1.**, SRIM simulations performed for PI implanted with Cu and Ag ions indicated that sequential implantation at 2.8, 2.0, and 1.2 MeV produced overlapping implantation profiles, resulting in an extended subsurface modified region rather than a modification confined to the immediate surface. The projected ion ranges depended on the implanted ion species and the target material, while the combination of three implantation energies enabled the deposited energy and implanted atoms to be distributed over a broader depth range.

Unlike conventional chemical functionalization, which is generally limited to surface-accessible regions, multi-energy ion implantation provides a physical strategy for tailoring materials through controlled subsurface modification. During ion implantation, energy is transferred through electronic excitation and nuclear collisions, leading to gradual structural rearrangement, defect formation, and local physicochemical activation of the material [2,3]. To complement the SRIM depth profiles, DFT calculations were used to clarify how implantation-induced dehydrogenation can create active sites capable of stabilizing Cu and Ag atoms in the modified polymer structure, as illustrated in **Figure 2**. Together, the SRIM and DFT results show that the same implantation strategy can be used not only to control the depth of modification, but also to support the chemical stabilization of implanted species in electrochemically relevant materials.

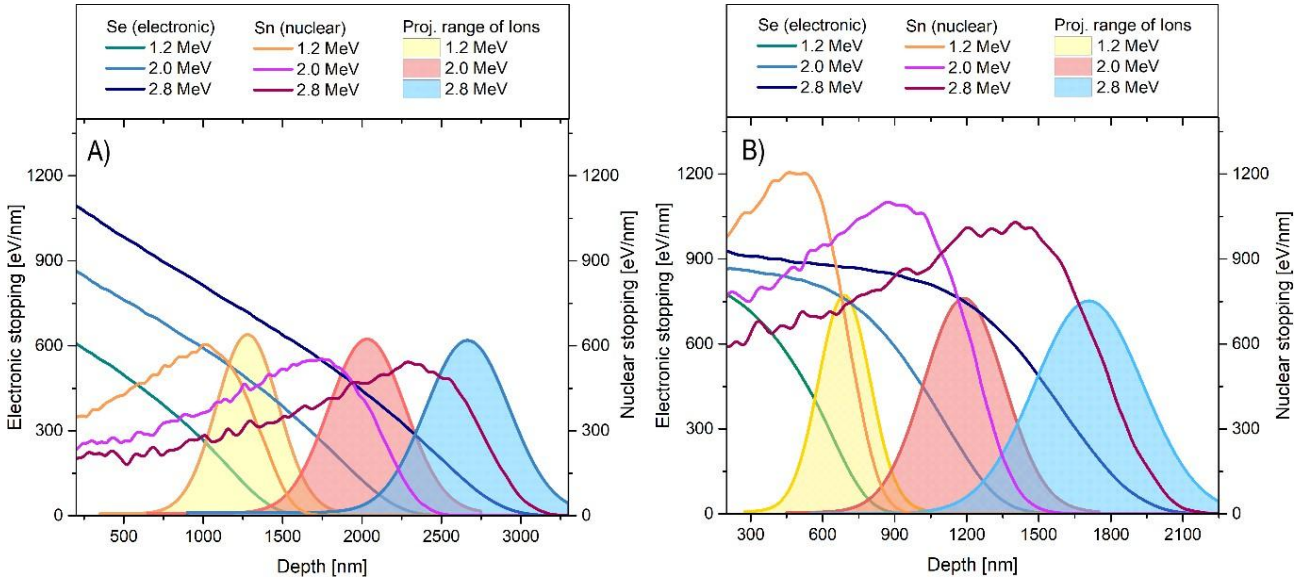


Figure 1. SRIM simulation of multi-energy ion implantation into polyimide (PI): implantation profiles for Cu ions (A) and Ag ions (B). The simulations demonstrate the formation of overlapping ion distributions produced by sequential implantation at different energies, leading to an extended subsurface modified region.

5.2 Engineering the Material–Electrolyte Interface

Although the investigated materials differ in their structure and function, both are relevant for energy storage applications because their electrochemical behavior is strongly influenced by the material–electrolyte interface [7]. Polymer materials can act as functional interfacial, protective, dielectric, or polarization-active layers, where the key requirement is not high electronic conductivity, but controlled interaction with ions and electrolyte species [7,8]. In contrast, multilayer graphene represents a conductive carbon-based electrode material, where structural order, defect density, surface chemistry, and interlayer spacing directly affect lithium-ion storage and transport [4,6]. In both systems, multi-energy ion implantation modifies precisely the region that is essential for electrochemical performance. The implanted ions do not affect only the outermost surface but create an extended near surface and subsurface region with altered physicochemical properties. In polymers, this modification leads to defect formation, dehydrogenation, local oxidation, and stabilization of implanted metal atoms, which together contribute to enhanced interfacial polarization. In multilayer graphene, implantation introduces controlled disorder into the graphitic structure, increases the interlayer spacing as confirmed by XRD, and changes the surface chemistry of the electrode material. This concept is illustrated in **Figure 2.** for the COC polymer system, showing the pristine COC structure and the corresponding changes in electrochemical response after ion implantation. Although this example focuses on COC, it illustrates the broader mechanism observed in this work: ion-beam-induced defects and chemical rearrangements can transform an originally passive or weakly active polymer interface into a more electrochemically responsive region [2,3]. Therefore, the role of ion implantation is not limited to introducing radiation damage. Instead, controlled defect formation can be used as a functional tool for designing material–electrolyte interfaces [5,7]. This makes multi-energy ion implantation a versatile physical strategy for tailoring electrochemical materials across different material classes, from polymer-based interfacial layers to graphene-based electrodes [2,5].

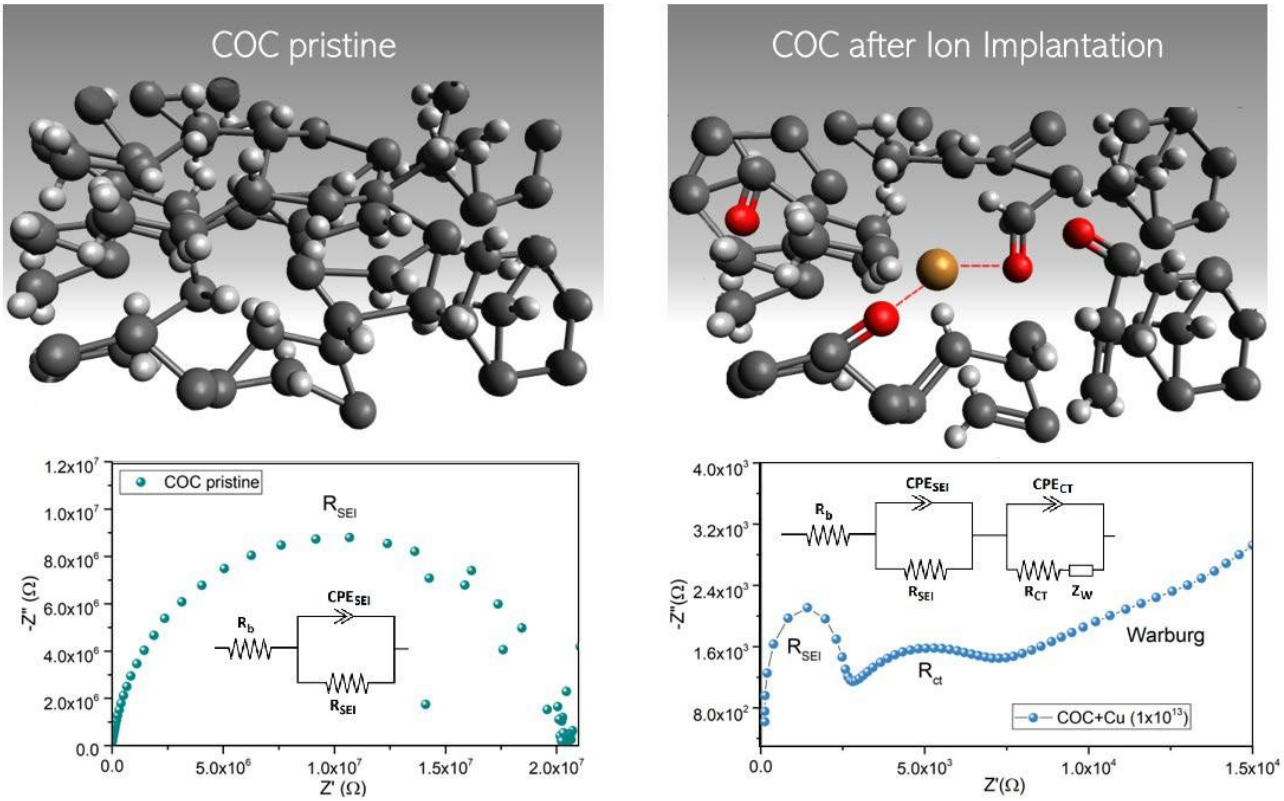


Figure 2. Schematic representation of the COC polymer system and its electrochemical modification after ion implantation. The figure shows the pristine COC structure and illustrates how implantation-induced defects and chemical rearrangements alter the electrochemical response of the polymer interface.

Conclusion

This work demonstrates that multi-energy Cu and Ag ion implantation can be used as a transferable physical strategy for tailoring electrochemical materials with fundamentally different structures and functions. Sequential implantation energies produced an extended near-surface and subsurface modified region, enabling controlled defect formation, local bond rearrangement, and stabilization of implanted metal atoms. In polymer materials, these changes promoted electrochemically responsive interfacial regions with enhanced polarization behavior, while in multilayer graphene, ion implantation introduced controlled structural disorder, increased the interlayer spacing confirmed by XRD, and modified the electrochemical response of the anode material. Overall, the comparison of polymer interfaces and multilayer graphene highlights that the same physical principle can be applied across distinct material classes, offering a versatile route for the design of future energy storage materials without relying solely on conventional chemical functionalization.

5 Acknowledgement

The instrumental development has been carried out at the CANAM (Centre of Accelerators and Nuclear Analytical Methods) infrastructure LM2015056. The scientific results were obtained with the support of the GACR Project No. 22-10536S and University of J. E. Purkyně project UJEP-SGS-2023-53-007-2. The authors acknowledge the assistance provided by the Advanced Multiscale Materials for Key Enabling Technologies project, supported by the Ministry of Education, Youth and Sports of the Czech Republic, Project No. CZ.02.01.01/00/22_008/0004558, co-funded by the European Union.

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