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Metal oxide ion-gated transistors: A perspective on in operando characterizations and emerging Li-ion-based applications

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METAL OXIDE ION-GATED TRANSISTORS: A PERSPECTIVE ON *IN OPERANDO* CHARACTERIZATIONS AND EMERGING Li-ION-BASED APPLICATIONS

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Abstract

In this Prospective, we discuss how ionic media interfaced to metal oxide (MO) semiconducting thin films can modulate their electronic conductivity. From *in situ* diagnosis tools to monitor the State-Of-Health of Li-ion batteries, to synaptic transistors where ion diffusive dynamics governs short-term and long-term plasticity, technologies based on ionic medium/MO interfaces are emerging, strongly benefitting from advanced nanoscale resolved scanning probe techniques and computational chemistry.

Keywords

Computation/computing; Devices; Electrical properties; Energy storage; Neuromorphic; Operando.

Introduction

Metal oxides (MOs) feature a myriad of chemical compositions, morphologies and structures to tailor their functional properties [1–3]. The chemical diversity in the oxides can be exemplified by the simultaneous presence of multiple cations and/or cations of a single element with several oxidation states [4–10]. MOs have been extensively used in areas such as energy conversion and storage, catalysis, water and soil remediation, sensing and biocompatible and/or biomedical materials [1,3,10–14].

Semiconducting MOs have been used in thin film electronics (e.g., WO_3 , MoS_2 , In_2O_3), including Ion-Gated Transistors (IGTs) [15–19]. Besides their intrinsic technological interest, IGTs are platforms to study processes such as mixed ionic and electronic (*iontronic*) transport, charge density modulation to control the electronic properties of materials, and charge carrier transport at high charge density. IGTs have low driving voltages ($<4\text{ V}$) and permit to achieve charge carrier density as high as

$10^{14} - 10^{15} \text{ cm}^{-2}$, due to the high capacitance of the ionic medium/transistor channel interface (ca. $1 - 10 \mu\text{F cm}^{-2}$) [20,21].

Factors such as the nature of the transistor channel material, ionic medium and ionic medium/channel interface bring about different mechanisms of doping modulation in IGTs. In *electrical double layer* (EDL) IGTs, the doping results from an electrostatic process at the ionic medium/channel interface, similar to field-effect transistors. Upon application of a gate electrical bias, a redistribution of ions at the ionic medium/gate and ionic medium/channel interfaces takes place, paralleled by accumulation (depletion) of charge carriers in the transistor channel, which is impermeable to ions (e.g., the doping is 2D, *two-dimensional*). On the other hand, in electrochemical IGTs the channel material undergoes a reversible Faradaic doping (dedoping) that can extend into the bulk of the channel (e.g., the doping is 3D, *three-dimensional*). Here, the application of a gate electrical bias induces a reversible redistribution of ions within the transistor channel and the ionic medium that, together with charge injection from source and drain, results in electrochemical doping (dedoping) of the channel, which is permeable to ions [22–24]. Oxygen vacancies created by strong electric fields between the ionic liquid and the channel have also been invoked to explain the working mechanism of IGTs making use of MO channels [21,25–27]. Considering the high capacitance (on the order of $10^{-6} \text{ F cm}^{-2}$) at the transistor channel/ionic medium interface, due to the nanoscale thickness of EDL (2-4 nm-thick), IGTs are also considered for fundamental studies, e.g., on metal-insulator transitions.

In our research groups, we used IGTs to study fundamental charge carrier transport aspects in organic and inorganic semiconducting films. We reported, for the first time in n-type organic semiconductors, potential regions of high conductivity in IGTs of the fullerene derivative phenyl-C61-butyric acid methyl ester (PCBM) [28]. This required a choice of ionic liquid leading, for its wide electrochemical

stability, to surface-confined, electrostatic (*two-dimensional*, 2D) film doping. We studied the doping mechanism in nanostructured MO-based ionic liquid-gated transistors and proposed a combination of electrochemical doping mechanisms whereby the injected electrons are compensated by the insertion of protons from water and by large cations of the ionic liquid packing at the interface with the MO [29]. Using Atomic Force Microscopy (AFM) force-distance profiling on EDLs forming at ionic liquids/ WO_3 interfaces, we followed, *in operando*, the evolution of the arrangement of the ions, and its effect on the doping [30]. More recently, considering that IGTs (i) can be operated at biases close to those of biological action spikes in neurons, (ii) can feature the processing rate of the human brain, and that neuromorphic computing requires both long-term and short-term potentiation capabilities within the same device, we proposed a methodology to control the IGT response times. Such methodology is based on gate-source voltage (V_{gs}) pulses' frequency, number of applied V_{gs} pulses and pulse duration [31,32]. In addition, we have expanded the use of the IGT concept to the growing field of lithium-ion batteries. Indeed, we demonstrated that it is possible to monitor *in operando* the evolution of the electronic transport properties of MO-based Lithium-Ion Battery (LIB) cathodes during their de-lithiation/lithiation, which represents a powerful LIB diagnostic tool [33].

In this Prospective, we will discuss a few cases of study in the field of MO-based IGTs, with the aim to show the relevance and potential of this class of devices at the fundamental and technological levels. After a brief discussion on the fabrication of MO films for IGT channels, we introduce the use of IGTs to follow *in operando* the electronic properties of LIB electrode materials and, in principle, the state-of-health of LIBs. Afterward, we critically present nanoscale observations pertaining to electrified interfaces in MO-based IGTs and how computational chemistry can help the interpretation of such observations. Finally, we include a discussion on possible developments on the use as synaptic transistors of IGTs based on MO channels and gated in presence of Li ions.

Preparation of Ion-Gated Transistors based on metal oxide channels

Several methods have been used to deposit MO thin film channels for IGTs [1,11,34]. Generally speaking, solution-based methods (e.g., sonochemical, co-precipitation, solvothermal/hydrothermal, sol-gel, and microwave-assisted) are preferred over vacuum-based methods (e.g., thermal deposition, sputtering, pulsed-laser, and atomic layer deposition) when high surface area materials and low-cost routes are sought [5,34–41]. High surface area enhances adsorption capacity, increases the number of active sites for reactions, and facilitates mass transport by minimizing diffusion limitations. Meanwhile, vacuum-based methods enable high-quality films on different substrates, offering control over composition, morphology and structure. Nonetheless, these methods often require sophisticated equipment and high-purity chemicals [1,2].

In solution combustion synthesis (based on self-sustained exothermic reactions (Fig. 1)), metallic precursors (e.g., $\text{Cu}(\text{NO}_3)_2$, $\text{Zn}(\text{NO}_3)_2$, $\text{In}(\text{NO}_3)_3$) react with oxidizer agents (e.g., urea, citric acid, alanine, glucose) at relatively low temperatures (100 – 250 °C) to produce crystalline materials with large surface areas [34,35]. Thin film transistors of In_2O_3 (Fig. 1(b)) [34,42], ZnO [43], $\text{In}_{0.7}\text{Zn}_{0.3}\text{O}_{1.35}$ [34], $\text{Zn}_{0.3}\text{Sn}_{0.7}\text{O}_{1.7}$ [34] and $\text{In}_{0.9}\text{Sn}_{0.1}\text{O}_{1.55}$ [34] showed the versatility of combustion-based methods to fabricate transistors on different substrates with reduced cost [34,42–45].

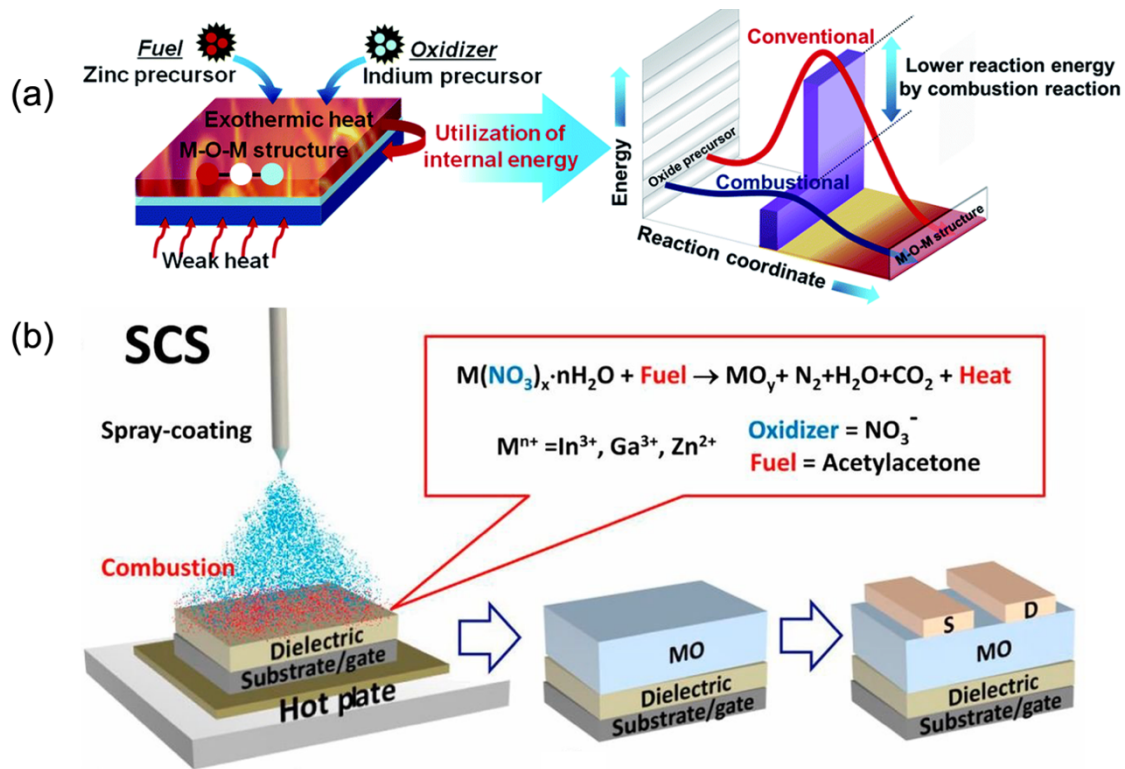


Fig. 1. Schematic diagram of (a) MO film synthesis via self-combustion synthesis (metal precursors bearing coordinated fuel and oxidizer ligands) and (b) process used to grow MO films under ambient conditions and corresponding bottom-gate top-contact transistor structure. Reprinted with permission from references [42,44].

Printing is a highly versatile process that enables both film deposition and patterning at ambient temperatures. Hong et al. reported flexible ZnO-based IGTs on polyimide (Kapton) [46], using as precursor for the ink formulation a $Zn(OH)_2$ aqueous ammonia solution, then thermally treating the films at 250 °C [46]. TiO_2 and SnO_2 films have also been explored in IGTs using printing on flexible substrates [47–50]. The authors considered flexible polyamide and polyethylene terephthalate substrates, demonstrating devices that can withstand mechanical bending [47–50].

In hydrothermal synthesis, parameters such as pH, temperature, and presence of additives are used to form MO materials with controlled morphological and structural characteristics [51]. We conducted comparative studies on IGTs based on WO_3 materials fabricated by different hydrothermal experimental protocols to understand the influence of morphology (e.g., granular, nanobelt, and nanoplate) on the IGT performance [52].

It could be interesting to consider biological syntheses (enzymatic reactions) for MOs to be applied in IGTs [11,53]. The applications of this approach are presently focused on biomedical, environmental, and energy conversion (antibacterial activity, wastewater treatment, catalytic reactions, and solar cells) [11,53]. This approach could also be considered for the sustainable production of metallic oxides for IGTs.

Exploring the use of Ion-Gated Transistors to follow *in operando* the electronic properties of Lithium-Ion Battery electrode materials as a function of their State-Of-Charge

LIBs are among the major components in the portable electronics revolution, as well as in the transition to a more sustainable society based on electric vehicles. Despite considerable progress, we still face challenges to the improvement of energy density, cycle life, reliability, and safety of LIBs. Electrochemical Impedance Spectroscopy (EIS) has indicated that the electronic properties of MO electrodes change with the degree of lithiation during charge-discharge cycles [54–57]. However, EIS does not permit clear discrimination between ionic (resistance to Li-ion transfer between the electrolyte and the electrode, through the Solid Electrolyte Interphase) versus electronic (electronic conductivity of the battery electrode material) contributions to the overall electrode impedance.

In operando strategies are needed to investigate the dependence of the electronic properties of LIB electrode materials on their State-Of-Charge (SOC), associated with their State-Of-Health (SOH, e.g., the specified performance and expected life of a used battery compared with a pristine battery of the same type). We studied IGTs incorporating the MO of interest as LIB electrode material, to understand their electronic properties at different SOCs, for their optimal use in LIBs.

We carried out the electrochemical characterization of the MO battery electrode materials deposited over patterned transistor substrates in a three electrode configuration, using the battery material included between the source and drain electrodes as the working electrode, activated carbon paper as the counter and quasi-reference electrode, and organic electrolytes with lithium salts as the gating medium [33]. We studied the battery electrode material as transistor channel material by measuring output and transfer characteristics to deduce conduction onset voltage, charge carrier density, charge carrier mobility and ON/OFF ratio. With $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ (NMC532)- and $\text{LiMn}_{1.5}\text{Ni}_{0.5}\text{O}_4$ (LNMO)-based composite materials formulated like in conventional LIB cathodes and operated in the organic electrolyte LP30 (1M LiPF_6 in ethylene carbonate:dimethyl carbonate 1:1 v/v), we observed that NMC532- and LNMO-IGTs work in depletion mode and the transistor (electronic) current can be reversibly modulated by fast lithiation/de-lithiation processes. Further, with NMC532-and LNMO-based cathode materials, we found that the electronic conductivity increases with the decrease of the lithiation in the transistor channel (battery cathode) material.

At present, we focus on the exploration of the electronic properties exhibited by lithium titanate oxide (LTO, $\text{Li}_4\text{Ti}_5\text{O}_{12}$), which is commonly employed at the anode in LIBs. The transistor configuration, with activated carbon as gate and an ionic liquid containing Li^+ ions as the gating medium (e.g., LiTFSI

in [EMIM][TFSI]) allows to effectively operate the LTO channel to achieve fully lithiated and delithiated states, while monitoring the current going through the LTO channel (Fig. 2.).

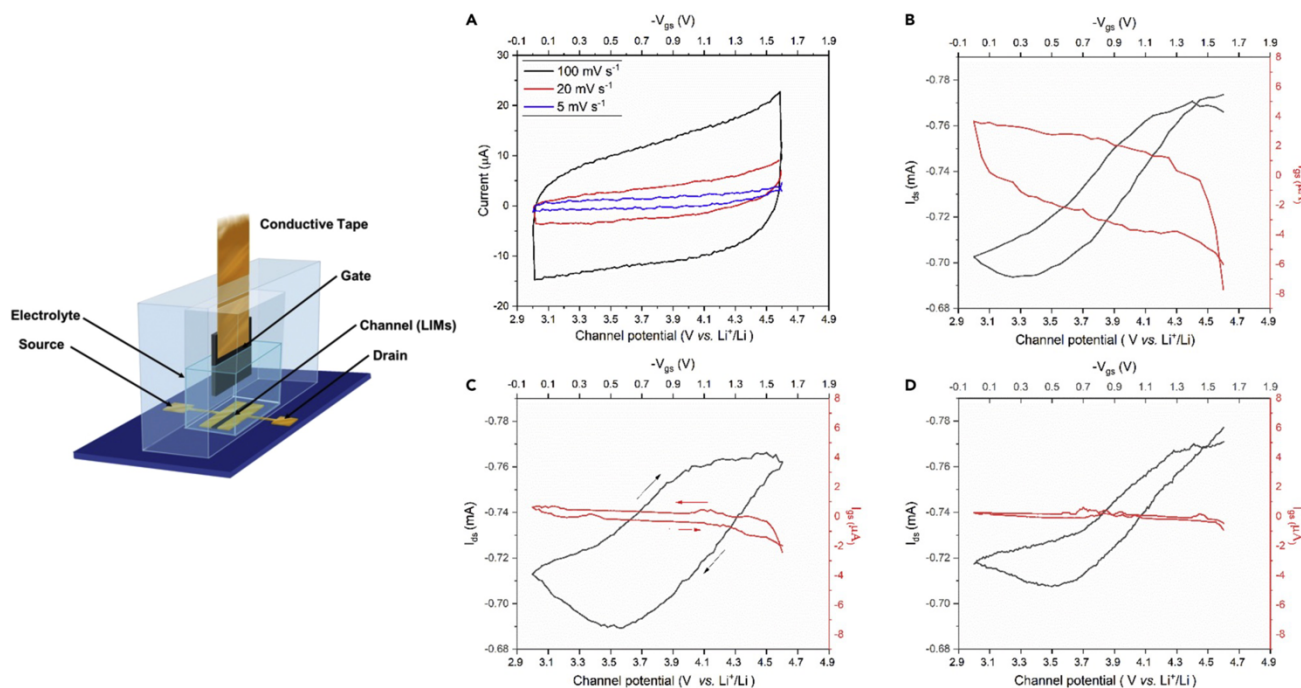


Fig. 2. Left: Scheme of an Ion-Gated Transistor configuration for the *in operando* exploration of the electronic transport properties of Lithium-Ion Battery electrode materials (Lithium-ion Insertion Material, LIM) as a function of their State-Of-Charge. **Right:** Characterization of IGTs making use of NMC532-based composite materials: (A) Cyclic voltammograms of the NMC532-based composite channel at different V_{gs} scan rates. Transfer curves (I_{ds} versus V_{gs} , at fixed V_{ds}) at V_{gs} scan rates of: (B) 100 mV s^{-1} , (C) 20 mV s^{-1} and (D) 5 mV s^{-1} , with $V_{\text{ds}} = -200 \text{ mV}$. Reprinted with permission from reference [33].

We are working to try to establish the correlation between the electronic conductivity of LIBs electrodes (both cathode and anode) and the degree of lithiation for each electrode during battery operation. These insights will serve as parameters in the determination of the SOC of the entire battery device and, more significantly, facilitate a precise evaluation of the SOH of LIBs.

We plan to carry out X-Ray Diffraction (XRD) and X-ray Photoelectron Spectroscopy (XPS) *in operando* investigations to shed light onto possible phase transitions and changes in surface chemical composition and bonding in the MO-based IGT channels.

Investigation of nanoscale phenomena pertaining ion-gating of metal oxide channels

The development of materials and devices for ion-gated electronics relies on the understanding of the mechanisms responsible for the characteristic high charge carrier density and mobility achievable at low gate biases [58]. In the case of MO materials gated with ionic media, the gating processes are direct consequences of transformations at the nanoscale interface between the solid and ion conductor [20]. This is true both in electrostatic mechanisms pertaining to the EDL capacitance and electrochemical mechanisms such as redox induced formation of defects, intercalation or insertion of ions [21]. Therefore, experimental investigations often rely on high spatial resolution material characterizations performed at the interfaces of the systems.

One of the most significant challenges for the study of ion-gated devices is that investigations must be performed in the presence of the electrochemical bias (*in operando*). Characterizations on the state of oxidation, phase transitions and formation/migration of oxygen vacancies must be performed *in operando*. This is indicated by the numerous reviews recently published on the issue [24,59,60]. Reviews are available on *in operando* scanning-probe microscopies (SPMs) [61], Transmission Electron Microscopy (TEM) [62,63], XRD [64], X-ray Absorption Spectroscopy (XAS) [65,66] and XPS [67].

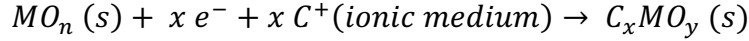
Phase transitions at ionic medium/metal oxide interfaces

Electrochemically induced oxygen vacancies formation and electromigration underlie the operation of IGTs based on perovskite-structured oxides including ZnO, In₂O₃, TiO₂, SrTiO₃ and SrCoO_{3-x} [21,25,26]. Devices based on oxygen vacancy triggered phenomena are investigated for sensor applications, neuromorphic devices [26], spintronics and gate-switchable magnetic materials [15,68]. One strategy available for studies of gate-induced oxygen vacancy dynamics is through monitoring changes in the materials' structure by *in operando* XRD (long-range) or TEM (local range). Both techniques can provide insights on deformations of the crystalline structure, formation of new phases, and change in oxygen content at the interface of the channel material. Recent examples include the study of phase shifts on SrCoO_x [69], SrTiO₃ [70] and SrFeO_{2.5} [71] perovskite oxide films interfaced with ILs by electrochemical TEM. *In operando* electrochemical XRD has been used for the dynamic investigation of oxygen-vacancy induced phase shifts and structural changes induced by electromigration via ion gating in devices based on BiO₃ [72], CdO [73], WO₃ [74] and many of the previously mentioned perovskite oxides [75,76].

XAS techniques performed at synchrotron radiation beamlines *in operando* can be used to probe local coordination geometry and electronic structure oxidation state and chemical environment of elements in ion-gated MO. Therefore, *in operando* XAS studies reveal in-depth insights about the electrochemical mechanisms related to oxygen vacancies. Noticeable recent examples in the field of iontronics include the ion-gating induced formation of CoO_{2.75} - SrCoO_{2.9} phases in SrCoO_x devices [77] and the study of electrochemically induced transition between conductive VO₂ and insulating V₂O₅ phases [78].

Ion-gating induced intercalation of cations at the interface of metal oxide channels

Some of the leading MO materials studied for neuromorphic IGT applications rely on the electrochemical double injection mechanism of ions and electrons [22,23]:



In which C^+ is a small radius cation such as H^+ and Li^+ , MO_n is the transition metal oxide used as IGT channel, and y is $((n-x)/2)$. Significant examples include devices based on WO_3/H_xWO_y interfaced with aqueous electrolytes, ionic liquids, and ion-gel media [52,79], WO_3/Li_xWO_y [80], $MoO_3/LiMoO_y$ [81] interfaced with ion gels, TiO_2/LTO ($Li_4Ti_5O_{12}$) materials interfaced with Li^+ ionic liquid media [82] and VO_2/H_xVO_y with ionic liquids [83]. Reversible intercalation/deintercalation of Li^+ coupled with redox reactions are also the fundamental mechanism responsible for charge storage in Li-ion battery electrodes [24].

In the case of WO_3 and MoO_3 , the double injection yields the formation of high conductivity bronze phases at the surface of the materials, resulting in an insulator to metal transition [51]. Although these phenomena are widely explored for gas sensors and electrochromic devices [51], neuromorphic applications require precise control for the switching process [81]. Therefore, advanced characterization investigations of the nanoscale processes that happen during electrochemical gating by double injection are currently being pursued, often *in situ* and *in operando* configuration.

Thin films of WO_3 deposited over lanthanum aluminate (LAO (1)) gated with Diethylmethyl(2-methoxyethyl)ammonium bis(trifluoromethylsulfonyl)imid (DEME-DFSI) ionic liquid have been investigated by combination of *in operando* XRD and *ex situ* Time-of-flight Secondary Ion Mass spectroscopy (TOF-SIMS) (Fig. 3) [84]. Authors were able to show that the films presented significant structural change only at gate biases above 3.5 V, by monitoring shifts to the (400) peaks with $2\theta-\omega$

scans. TOF-SIMS analysis indicated the presence of hydrogen at the gated films at depths up to 60 nm, with increasing concentration for films biased with higher gate potentials. Authors concluded that the double injection mechanism promoted by H^+ plays a significant role in the device mechanism. At lower biases, the intercalation is limited to the surface of the films, but at $V_{gs} > 3.5$ V the potential barrier for ionic diffusion of the protons in the film is overcome, resulting in structural transformation observed in the XRD results. The presence of protons was attributed to water in the ionic liquid, in accordance with other works on ionic liquid gated MO transistors [52,75,83,85].

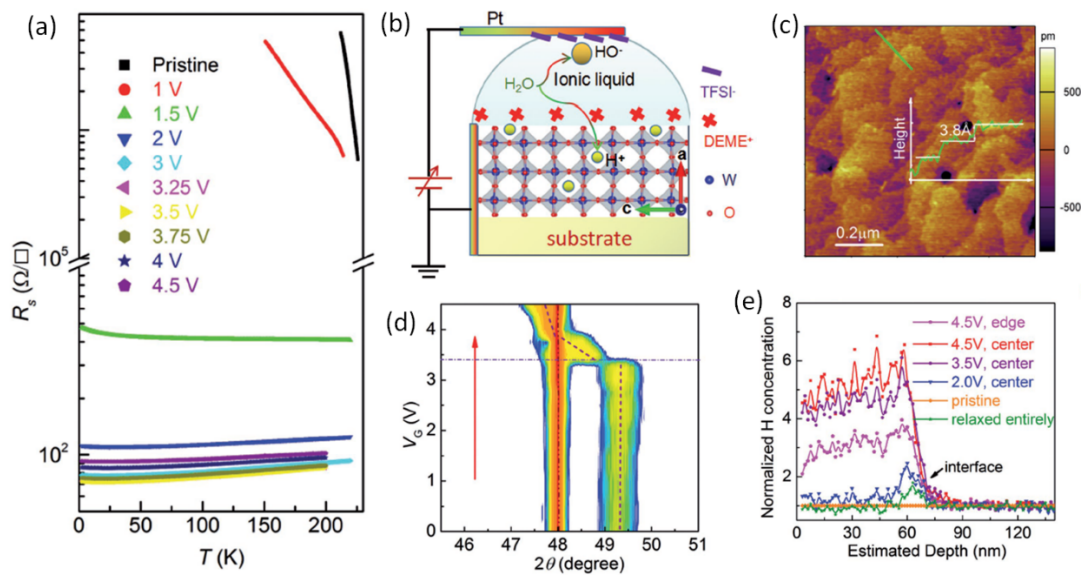


Fig. 3. WO₃-based DEME-TFSI-gated transistors: (a) sheet resistance as a function of temperature and gate bias (b) schematic representation of the device (c) AFM image of the film (d) *in situ* XRD 2θ - ω scans around WO₃ (400) peaks, as function of gate bias (d) Depth profiles of the H⁺ ion in a series of WO₃ samples obtained by TOF-SIMS. Reprinted with permission from reference [84].

XRD *in situ* was also used to study lattice structure and doping level of WO₃ with Li⁺ in IGT [86] and a similar proton-induced-doping double injection mechanism was proposed for devices interfaced with polyethylene glycol + sodium fluoride ion gels, with the authors attributing the source of protons to

water traces found in the ion-gel [87]. Devices based on MoO_3 gated by Li^+ intercalation were investigated by XRD [73], TEM [73] and TOF-SIMS [20]. All these works can advance the present understanding on the scale of the ion permeation and structural changes that the material undergoes in relationship with the insulator-metal transitions. The case of vanadium oxide is interesting in the sense that earlier works attributed the insulator-to-metal transitions observed in ion-gated devices to oxygen defects [27,88], but more recent studies based on in situ Nuclear Magnetic Resonance [89], tip-induced SPM hydrogenation tests [85] and XAS studies [83] suggest that the formation of H_xVO_2 phases at the surface plays a significant role, with significant implications for the proposed practical uses. The new findings highlight the importance of probing the interfaces with complementary *in operando* techniques, as the gating mechanism of MO channels by electrochemical bias might be the result of simultaneous processes.

Computational studies at Ion-Gated Transistor interfaces

Ion arrangements in the EDL result from interactions such as long-range electrostatic and short-range van der Waals interactions or hydrogen bonding, and brings about distinct microstructures, in turn affecting IGT device physics (doping) and performance [30,90–98].

Classical (Molecular Dynamics (MD)) or quantum (Density Functional Theory (DFT)) computational methodologies have been used to study the properties of ionic liquid at polarized surfaces (electrified) [91,98–103]. Black et al. investigated the 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ([HMIm][TFSI])/amorphous indium gallium zinc oxide (a-IGZO) interface by AFM [91]. MD was used to figure out the structure of the layers and the distances of the layers from the MO surface. MD suggested that the conductivity of the films is controlled by the shrinking of the EDL thickness via cation reorientation and anion expulsion [91].

Zhang et al. utilized MD simulations to investigate the interfacial structure at a mica surface exposed to hydrophobic ([EMIM][TFSI]) and hydrophilic ([EMIM][BF₄]) ionic liquids in the presence of both water molecules and K⁺ ions (Fig. 4). By increasing the water content, the K⁺ can be found at higher distances from the interface for both ionic liquids; the interfacial water layer is denser in the hydrophobic ionic liquid than then in the hydrophilic one. [101]

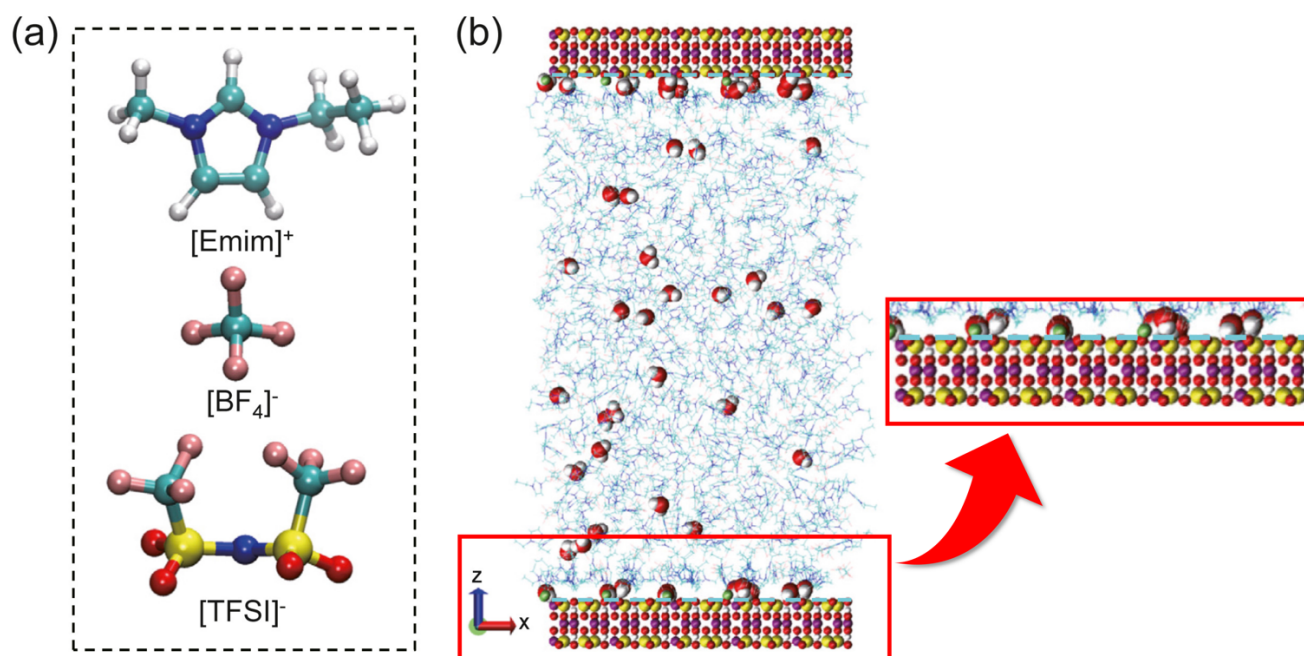


Fig. 4. MD modeling of water-RTIL mixture between two mica surfaces. (a) All-atom model of ions [EMIM]⁺, [BF₄]⁻ and [TFSI]⁻. (b) Snapshot of the MD system. At the edges of the theoretical box, the colored part (yellow, purple, and red) indicates mica (silica/alumina) (insertion Fig. 4(b)), lines represent ionic liquids, dark green spheres represent K⁺ ions and red/white spheres represent water molecules. The water molecules were enlarged for clarity. The cyan dash lines represent the position of oxygen atoms on mica surface. Reprinted with permission from reference [101].

At the 1-methyl-3-butylimidazolium fluorophosphate ([BMIM]/[PF₆])/rutile TiO₂ interface, MD simulations revealed the formation of a dense layer of [BMIM], oriented parallel to the TiO₂ surface [104]. The interaction energy calculated by MD, suggested an adsorption of [BMIM] at the TiO₂ surface featuring low ion mobility and stronger affinity between the [BMIM] and TiO₂ with respect to [PF₆] [104].

Future developments in the field of Ion-Gated Transistors

IGTs based on MO channels have shown promising results as biological synapses, mimicking synaptic functions such as potentiation, depression, spike-timing-dependent plasticity, and learning (Fig. 5) [105–107].

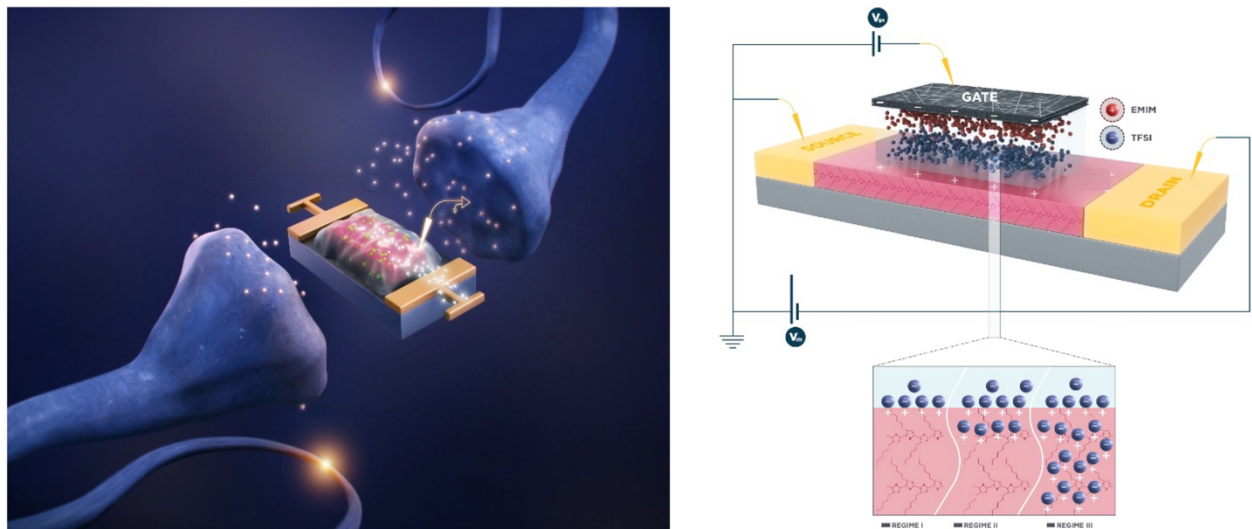


Fig. 5. (a) Synaptic IGT transistor mimicking a biological synapse. (b) Device configuration and potential doping scenarios (I to III) when a gate-source voltage is applied to an IGT. Reprinted with permission from reference [31].

Adding Li^+ to the ion gating medium can change the IGT response time significantly and, consequently, the time scale of synaptic activities. When Li^+ ions are electrostatically adsorbed at the surface of the semiconducting channel, they can easily diffuse back into the ionic medium once the gate bias is removed, resulting in short-term plasticity. Interestingly, we can modulate the synaptic function and subsequent Li^+ diffusive dynamics in IGTs. For instance, by using high V_{gs} pulse amplitudes, a high number of pulses, or a channel material featuring a high degree of order, once Li^+ ions intercalate into the channel they cannot easily leave the channel after V_{gs} removal, leading to long-term plasticity [106].

Last but not least, work is in progress to study by *in operando* AFM force-distance profiling the dynamics of the arrangements of the ions at IGT electrified interfaces as a function of the advancement of the doping degree, in semiconducting metal oxide channels.

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Declarations

Funding and Conflicts of interests

The authors have no financial or competing interests to declare that are relevant to the content of this article.

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