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Unconventional Applications of the Magnetohydrodynamic Effect in Electrochemical Systems

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Abstract

The theory of electromagnetism has made countless contributions to the understanding of physico-chemical phenomena and continues to be relevant in many scientific fields. The use of electromagnetism to induce physical flux in a fluid containing ions is known as magnetohydrodynamics (MHD). In electrochemistry, MHD has been explored, among others, to produce for example chiral structures on the surface of electrodes. However, the Lorentz force responsible for inducing hydrodynamic convection in an electrochemical system also allows bypassing mass transport limitations in novel and unconventional processes. This review aims to highlight the principles of MHD convection and to discuss recent and unconventional approaches to the integration of magnetic fields in electrochemical systems.

Keywords: Magnetic field, magnetohydrodynamic effect, microfluidics, electrocatalysis, self-propelled swimmers.

1. Introduction

The synergy between electric and magnetic fields goes well beyond Maxwell's electromagnetism theory, since such an interplay can be extended to many different areas of science, and among others also to electrochemistry [1-5]. Due to the electric and ionic currents involved in an electrochemical process, it is possible to assume that the presence of a magnetic field can affect the overall kinetics of an electrochemical conversion. From magnetic hyperthermia [6] to magnetic attraction [7], magnetochemistry has passed from an experimental curiosity to a well-established field of research. For example, in recent years the study of spin polarization effects has become a hot topic of research [8-10]. Among the different magnetochemistry approaches, the most popular one is probably the induction of hydrodynamic convection by the Lorentz force, as it is a powerful tool to overcome mass transport limitations [2,3]. Briefly, when an homogeneous magnetic field (\vec{B}) is imposed perpendicular to an ion flux (\vec{J}), associated to an electrochemical process, a Lorentz force ((\vec{F}_L)) perpendicular to both \vec{B} and \vec{J} is produced. This is a well-known phenomenon, the so-called magnetohydrodynamic effect (MHD). One specific type of research related to the MHD effect is focused on the electrodeposition of metals, alloys or conducting polymers, in order to induce surface chirality [11,12]. However, since the MHD convection is a surface localized phenomenon, it has gained considerable attention also in different other fields of research, such as redox microfluidics or electrocatalysis. The aim of this review is to describe the physico-chemical principles governing the MHD effect and to discuss some unconventional trends in this field.

2. Fundamental concepts behind the magnetohydrodynamic effect

In electrochemistry, mass transport is governed by the diffusion of electroactive species towards the electrode surface. Under classic conditions, when a potential difference is applied between two electrodes a concentration gradient is formed, due to the continuous consumption of the analyte (Figure 1a). The thickness of this so-called Nernst diffusion layer (δ_D) is a function of time, the electrode potential, the concentration (C^*) and the diffusion coefficient of the analyte (D). Thus, as the size of the diffusion layer increases, the current obtained by the electrolysis of the electroactive species is governed by the δ_D value. However, in the presence of a homogeneous $B \vec{}$, a local Lorentz force is induced on the ion flux associated to the electrochemical process (Equation 1).

$$F \vec{}_L = J \vec{} \times B \vec{} \quad (1)$$

Under these conditions a hydrodynamic boundary layer (δ_0), with an ion flux velocity gradient across the layer ($U(y)$) is formed (Figure 1b). This leads to a decrease of the thickness of δ_D with a concomitant acceleration of mass transport, which causes a continuous supply of electroactive species towards the electrode surface, improving the mass-transport limited current. Willner et al. proposed a theoretical model that describes the influence of the $(F_L) \vec{}$ on the limiting current in the presence of a homogeneous magnetic field oriented parallel to the electrode surface (equation 2) [13-15].

$$i_L = 0.63 \left[(\rho R) \right]^{-1/3} D^{8/9} \nu^{-2/9} \left[(nFC^*) \right]^{4/3} B \vec{}^{1/3} \quad (2)$$

where n is the number of electrons, F is the Faraday constant, R is the size of the electrode, ρ is the specific density of the fluid, ν is the kinematic viscosity, D and C^* are the diffusion coefficient and bulk concentration of the electroactive species, respectively. Since the obtained Lorentz force acts only on moving charged species and strongly depends on their velocity and the strength of $B \vec{}$, its maximum value is obtained when $J \vec{}$ and the magnetic field are perfectly orthogonal to each other. For example, the magnitude of a Lorentz force produced by a magnetic field of 1 T, orthogonal to a current density of 100 mA/cm², is comparable to the buoyancy force (≈ 103 N/m³), but inferior to mechanical stirring (≈ 105 N/m³). However, the limiting current obtained in the presence of a homogeneous magnetic field is comparable to the one produced by electrochemical measurements performed with a rotating disk electrode [16,17]. For example, Hinds et al. observed that a limiting current of 9.4 A/m² produced in the presence of a 0.5 T magnetic field, is roughly equivalent to the current obtained with a rotation rate of 10 rpm [17].

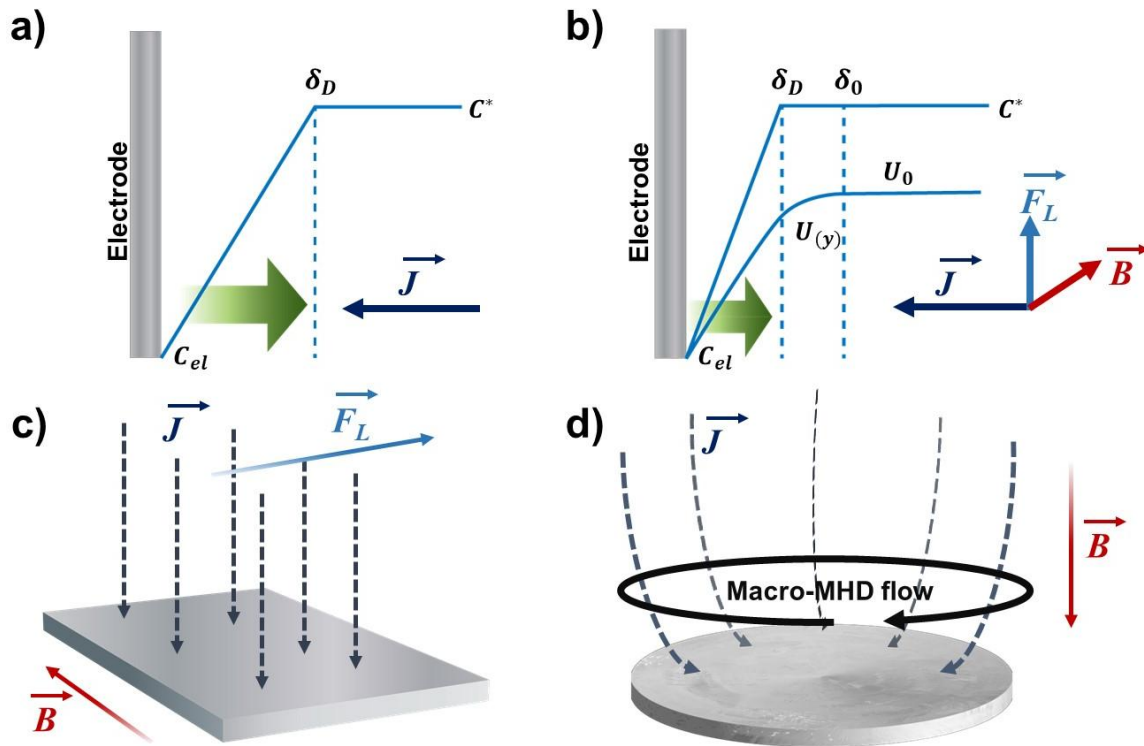


Figure 1. Illustration of the mechanism of the mass transport of electroactive species (a) in the absence and (b) in the presence of an applied magnetic field. $J \rightarrow$ is the diffusional flux of the electroactive species, c_{el} is the concentration at the electrode surface, δ_D and δ_0 are the Nernst diffusion layer and the hydrodynamic boundary layer thickness, respectively, and U_0 is the fluid velocity at the outer edge of the hydrodynamic boundary layer. (c) Illustration of the MHD flow under the influence of a homogeneous magnetic field when $B \perp J$ and (d) when $B \parallel J$.

The hydrodynamic convection induced by the Lorentz force is intimately related to the spatial arrangement of the magnetic field with respect to the position of the electrode. This interaction can cause two different types of MHD flow in the presence of a homogeneous magnetic field. When the magnetic field is placed parallel to the surface of the electrode, the entire ion flux is perpendicular to $B \rightarrow$ ($B \rightarrow \perp J \rightarrow$), producing a macro-MHD flow (Figure 1c). Under these conditions, it is possible to predict the limiting current according to the above mentioned theoretical model. However, when $B \rightarrow$ is placed perpendicular to the electrode surface ($B \rightarrow \parallel J \rightarrow$), only the ionic current at the electrode edges is able to generate a Lorentz force, causing the formation of macro-MHD flow along the electrode edges (Figure 1d). In this case, the effect of the magnetic field on the overall electrochemical kinetics is strongly related to the intensity of $B \rightarrow$ and the distance between the electrode and the magnet [18]. In addition, in such a configuration, due to the presence of local inhomogeneities, micro-MHD vortices are formed at the electrode surface (Figure 2a). This micro-MHD flow pattern induces localized high current density spots at a considerably smaller scale, impossible to produce via mechanical stirring. Finally, it is important to highlight that a magnetic field can induce other forces in addition to $(F_L) \rightarrow$, such as a paramagnetic force ($(F_p) \rightarrow$), a field gradient force ($(F_B) \rightarrow$) and an electrokinetic force ($(F_E) \rightarrow$) (Table 1) [16,17]. From a physical point of view, their origin is intimately related to the properties of the electrolyte ($(F_p) \rightarrow$) and the applied magnetic field ($(F_B) \rightarrow$) or the dynamic behavior of the charged species at the electrode/electrolyte interface ($(F_E) \rightarrow$, $(F_L) \rightarrow$). However, $(F_B) \rightarrow$ is absent in the presence of a homogeneous magnetic field, and $(F_p) \rightarrow$ is negligible compared to the driving force of diffusion ($(F_D) \rightarrow$). Therefore, it is

possible to assume that the hydrodynamic convection is driven by the Lorentz force in the vicinity of the electrode surface, and on a smaller scale, in the diffuse layer, by the electrokinetic force.

Table 1. Typical forces acting in aqueous electrolytes.

Force	Typical value	Physical origin	Definition
Driving force for diffusion ((F_D))	1010		Force due to the presence of a concentration gradient.
Paramagnetic force ((F_P))	104	Magnetic properties	Paramagnetic susceptibility of species in the diffusion layer
Field gradient force ((F_B))	101	Magnetic properties	Force due to the presence of a magnetic field gradient.
Lorentz force ((F_L))	103	Motion of charges	Motion of charges orthogonal to a magnetic field.
Electrokinetic force ((F_E))	103	Motion of charges	Force on charge carriers under the influence of a magnetic field in the diffuse layer.

3. Unconventional applications based on the magnetohydrodynamic effect

After these theoretical considerations, we would like to present in this paragraph a series of rather unconventional applications of the MHD concept. The enhancement of the faradic current due to MHD induced convection has been quite well-studied over the last 40 years, but some studies have also focused on more exotic phenomena, such as the electro-induction of surface chirality. In the present contribution, all the different examples of the MHD effect involving electro-deposition or dissolution of metals and polymers are considered as conventional applications. [1-4,6,8,12] However, recent attempts have been made to couple the MHD effect with alternative electrochemical systems, where this magnetochemical phenomena is the driving force to produce dynamic behavior. For example the hydrodynamic convection induced by the Lorentz force has been successfully used to boost the directional pumping and mixing of fluids (redox magnetohydrodynamic microfluidics), to promote mass transfer and facilitate bubble ejection (electrocatalysis) and to generate motion (Lorentz force-driven swimmers). In the following, we summarize the fundamental concepts concerning the above mentioned electrochemical processes.

Electrocatalysis

A first and rather straightforward idea is to benefit from the MHD effect in the context of the design of highly efficient electrocatalysts, which are of importance to develop novel energy conversion and storage devices, i.e., water electrolyzers, fuel cells or metal-air batteries. Commonly the efficiency of an electrocatalytic process can be fine-tuned by increasing the exposed amount of active sites or by enhancing the intrinsic activity of the catalysts. However, an interesting alternative is to enhance the overall performance of an electrocatalytic system also by improving the efficiency of local mass transport. In this context, the MHD effect is presented as a powerful tool in electrocatalysis, due to the surface-localized mixing of the solution and the efficient gas bubble removal (Figure 2a) [19]. It is important to highlight that also alternative magnetic field-based methods have been developed to enhance the electrocatalytic performance such as hyperthermia, Maxwell stress effect or spin

selectivity [19]. Nonetheless, in this section we focus exclusively on the basic concepts of MHD-enhanced electrocatalysis.

The macro- and micro-MHD convection in electrocatalysis impacts primarily the removal of gas bubbles from the electrode surface [20]. In a first order approximation, at the macroscopic level, the theoretical residence time and the size of the bubbles released from the electrode surface, is intimately related to the net magnetic force ($(F_{net})^{\rightarrow}$). For example, when $(F_L)^{\rightarrow}$ and the buoyancy force ($(F_B)^{\rightarrow}$) are in the same direction, the upward pumping effect of the hydrodynamic convection theoretically reduces the average release size and residence time (Figure 2b), whereas the opposite effect is expected when $(F_L)^{\rightarrow}$ and $(F_B)^{\rightarrow}$ are opposite to each other (Figure 2c) [21-23]. However, at the microscopic scale, the micro-MHD vortex triggers a more complex scenario, since the release of bubbles depends on the spatial distribution of low pressure regions within the bubble [24]. According to Liu et al., at a microelectrode, the hydrodynamic convection induced by the Lorentz force generates low pressure regions at the lower half part of the bubble [24]. This is caused by the interplay between the macro- and the micro-MHD vortex. When using a microelectrode ($d \approx 0.2$ mm), the macro-MHD vortex produces a more violent convection at the electrode interface, in comparison to the micro-MHD flow, which decreases the pressure at the bottom part of the bubble. Such a different fluid flow induces a pressure difference around the bubble, which nails it to the electrode surface. However, at a macro-electrode the edge induced MHD flow enhances the upper part of the flow, producing a low pressure region above the top of the bubble, thereby enabling its release. In addition, the low pressure regions can evolve over time, due to the possible formation of neighboring bubbles on the electrode surface. In such a case, the formation of a high pressure region between the bubbles acts like an external force to push them apart [25]. Furthermore, the influence of the micro-MHD flow can be fine-tuned by changing the low pressure regions, either by using porous [26] or fringed electrodes [27,28] and arrangements of multiple magnets [29].

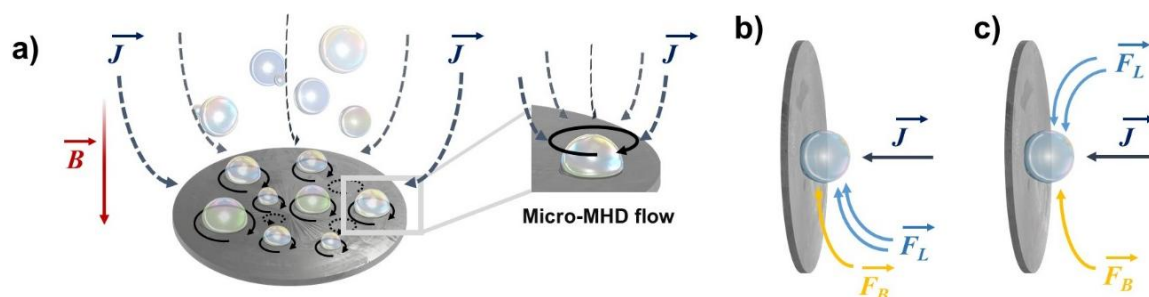


Figure 2. (a) Illustration of the micro-MHD flux generated around bubbles when $B // J$. (b) Illustration of the forces involved in the bubble release mechanism under the influence of a homogeneous magnetic field when F_L and F_B are oriented in the same direction and (c) in the opposite direction. Adapted from reference [19].

The hydrodynamic convection driven by the induced Lorentz force, impacts the overall performance of an electrocatalytic reaction in three different ways: (i) by reducing the ohmic voltage drop, (ii) by decreasing the activation potential, due to the continuous release of bubbles that otherwise would inhibit the active surface of the electrode, and (iii) by partially decreasing the thickness of the diffusion layer, which leads to an enhanced mass transport. Such advantages have been explored to enhance the performance of alternative electrocatalytic systems. For example, Kenis et al., observed a decrease of the overall cell potential during the electrolysis of CO_2 , in the presence of an external magnetic field [30]. Furthermore, Wilmer et al., demonstrated the possible enhancement of the conversion rates of bioelectrocatalytic reactions. [14,15] A 3-fold increase of the bioelectrochemical

conversion rate of oxygen, lactate and glucose, in the presence of a constant magnetic field, was observed.

Redox magnetohydrodynamic microfluidics

A second and less obvious field of research using the MHD effect is the design of microfluidic systems for directional pumping and mixing, which has gained considerable attention due to potential applications ranging from biochemical analysis to chemical synthesis [31]. Microfluidics is an interesting field of research since it allows the handling of fluids in small quantities in an automated manner. This leads to different advantages such as the production of less waste, reduced energy consumption, portability, and enhanced sensitivity. Different well-established microfluidic pumping methods have been developed i.e., electrokinetic, mechanical, centrifugal and redox-MHD [31,32]. In particular, the latter presents the unique opportunity to control microfluidics, that means to stop, start, reverse and fine-tune the flow without the need of an external valve or redesigning the device. It is important to highlight that magnetic forces can be used in a wide range of microfluidics applications, i.e. mixing, transport and trapping or for responsive valves. However, for such sophisticated applications, hybrid ferromagnetic materials are required [33].

The redox-MHD microfluidic approach takes advantage of the synergy between the ionic flux associated to a faradaic process and a magnetic field orthogonal to the electrode surface [34-36]. In this approach, the redox reactions are triggered by applying a relatively low potential difference between two electrodes (of the order of millivolts), using 1:1 molar ratio solutions of the corresponding electroactive species. Under these conditions, electrodisolution of the electrodes and bubble formation at their surface can be avoided. As stated above, the magnitude of the induced Lorentz force is given by equation 1, thus in a first order approximation, under the influence of a constant $B \vec{}$, relatively low faradic currents may already induce a MHD-flux at the surface of the electrodes. Different reversible redox probes with relatively low electrode potentials and good electron transfer kinetics can be used. Various attempts to visualize the redox MHD flow patterns triggered by the oxidation/reduction of classic redox couples, i.e., $Fe(CN)_6^{4-}/3-$ and $Ru(NH_3)_6^{2+}/3+$ or by using the ionic exchange involved in the charging/discharging process of π -conjugated polymers, have been developed [37-39]. In the majority of the studies, the redox-MHD microfluidic device is based on a microarrangement of parallel band electrodes (Figure 3a). Furthermore, coupling redox MHD with microscopy techniques, such as dark-field microscopy or light sheet microscopy [36,39], facilitates single-particle tracking at the nanoscale range, which allows quantifying the fluid velocity produced by relatively low currents (Figure 3b). Commonly, it has been demonstrated that the MHD flow speed, between the band electrodes, is in the 5 – 50 $\mu m/s$ range [40]. It is important to highlight that the intensity of $(F_L) \vec{}$ and its concomitant fluid velocity depends on additional experimental factors, such as the viscosity of the solution, the border effect, the distance between the electrodes and counteracting flows or shear stress. However, the most outstanding feature of the redox MHD microfluidic approach is the possible control of the directionality of the flux, by changing the orientation of the magnetic or the electric field. For example, Fritsch et al., took advantage of such a direction control in order to develop a semi-continuous, unidirectional microfluidic pumping device [41]. In this work, a multi-pulse galvanostatic perturbation scheme was efficiently coupled to an automatic switching of the $B \vec{}$ orientation to produce a semi-continuous unidirectional flux. In addition, the flux velocity produced by $B \vec{}$, which is orthogonal to the microfluidic device, can be influenced by the presence of magnetic field gradients $((\nabla B) \vec{})$, positioned close to the electrodes [42]. The force produced by $(\nabla B) \vec{}$ is able to generate a crossflow that overlaps with the classic MHD flux, causing a slight redirection of the main flow. This is

associated to a localized change of the magnetic flux density, which induces an additional field gradient force (F_B)⁻.

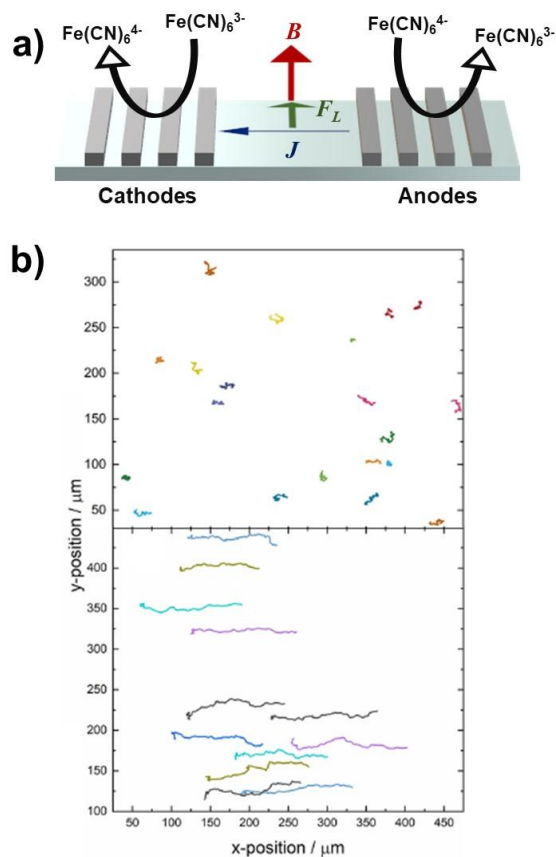


Figure 3. (a) Schematic illustration of the electrochemical processes involved in a RMHD device with a representation of the microarrangement of parallel band electrodes, the redox reactions, the ionic current, and the magnetic field which produces the MHD force and the fluid flow. (b) Trajectory plots of mixed NP suspensions at a constant applied current (50 μ A) in the absence (top) and presence of a magnetic field (bottom). Adapted from reference [36].

Lorentz force-driven swimmers

A complementary approach with respect to the one described in the previous paragraph is to generate MHD not in a stationary device like a microfluidic channel, but around mobile objects, in order to induce a reciprocal effect. This should greatly affect their dynamic features. A family of objects, which is interesting in this context, are macro-, micro- and nano swimmers. These are devices that harvest energy from an external input and transform it into motion. Such systems have gained considerable attention due to their numerous potential applications ranging from sensing to biomedicine [43-45]. Different propulsion mechanisms based on asymmetric release of bubbles, self-generation of local chemical gradients and electric fields (self-diffusiophoresis and self-electrophoresis, respectively) or applying external stimuli i.e., light, sound and electric or magnetic fields, have been extensively studied [46-51]. However, developing novel propulsion mechanisms that enhance the swimmer speed and enable trajectory control is still a challenge. For example, recently self-propelled magnetotactic swimmers, where motion is triggered by a physico-chemical processes and the trajectory is guided by a magnetic field, have been designed [51]. However, such devices require the presence of ferromagnetic components, which makes their design more complicated and limits the degree of freedom in terms of materials choice. In this context, the MHD

effect is an interesting alternative to boost the dynamic behavior of these moving particles. Theoretical studies show that the MHD acceleration of fluid surrounding an object enables propulsion, since the produced thrust is comparable to the weight of the swimmer [52]. Recently, the synergy between the self-electrophoretic mechanism and an external magnetic field was explored. In this work, an external $B \vec{}$ acts on the ionic fluxes produced by the spontaneous oxidation of Mg and the reduction of H^+ at the extremities of a Janus Mg/Pt swimmer (Figure 4a) [53]. Such reactions cause a continuous flux of ions towards or away from its surface, due to the consumption of electroactive species, which produces, by placing a magnetic field orthogonal to the surface of the self-electrophoretic object, a MHD vortex at each extremity of the device. Under these conditions two well-defined macro-MHD flow patterns along the edges of the electrodes were observed. In addition to the MHD vortices, an ion flux parallel to the surface of the device is formed to compensate the electron flow from the anode to the cathode. Such a horizontal ionic current, in the presence of the external $B \vec{}$, generates an additional Lorentz force, inducing a torque that causes a predictable clockwise or anticlockwise motion as a function of the orientation of the magnetic field (Figure 4b). As stated above, the generated hydrodynamic flow improves the reaction kinetics due to a better mixing of the solution in the vicinity of the swimmer surface, and by removing efficiently gas bubbles. Thus, this novel propulsion mechanism, based on the synergy between the magnetic field and ionic flux, is an interesting alternative to boost the propulsion speed by up to two orders of magnitude. Finally, it is important to point out, since the ionic flux is triggered by a self-electrophoretic mechanism, that this concept is generic. Therefore, it is possible to envision the use of this approach for the design of a vast variety of Lorentz force-driven swimmer with alternative functionalities.

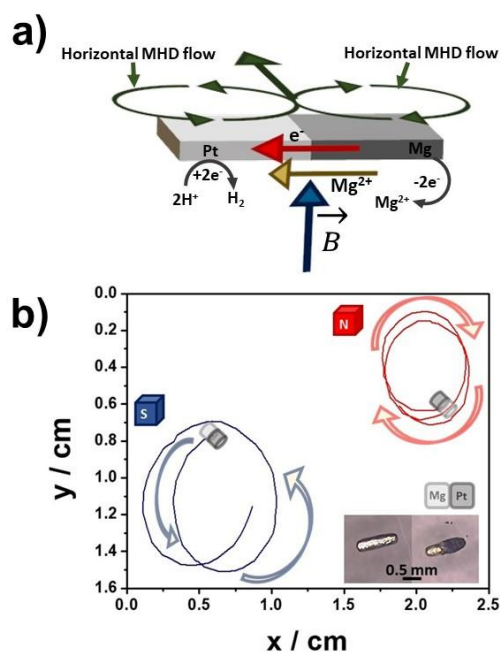


Figure 4. (a) Illustration of the Janus Mg/Pt swimmer with a representation of the associated chemical reactions, the spontaneous ionic currents, the magnetic field and the resulting Lorentz force. (b) Trajectory plots of two micro-Janus swimmers as a function of the magnetic field orientation. Inset: optical pictures of the Mg rods before and after asymmetrical modification. Adapted from reference [52].

4. Conclusions and perspectives

This short review allows a comprehensive in-depth look into the principles governing electrochemically triggered MHD convection. We specifically aim to draw attention to current and unconventional applications of the MHD effect, which illustrate its potential interest for optimizing electrocatalysis, redox magnetohydrodynamic microfluidics, and Lorentz force-triggered motion in microswimmers. The MHD effect can be strategically used to control with precision the flow of ionic currents, by adjusting the strength and spatial position of the magnetic field. In addition, the Lorentz force promotes fluid circulation and ultimately diminishes the impeding influences of the diffusion layer and bubble formation. Furthermore, this concept allows also a precise control of microswimmer propulsion, in terms of trajectory and speed of the object. The described Mg/Pt hybrid microswimmers enable bypassing the integration of ferromagnetic components, and this could inspire the development of alternative Lorentz force-driven Janus swimmers composed of a wide range of materials. Although the MHD effect is very well-known, its application to alternative electrochemical research topics, such as electroanalysis, water remediation and electrosynthesis, is promising and merits further exploration. For instance, the straightforward optimization of the intensity and spatial position of the magnetic field, allows fine-tuning the MHD convection, and thus maximizing the efficiency of the electrochemical systems. In addition, since the induced Lorentz force is a phenomenon localized close to the surface, it can be influenced by the physico-chemical properties of the working electrode, allowing eventually the on purpose generation of micro-MHD vortices. For example, the use of highly ordered macroporous electrodes or well-positioned nanoparticles can lead to the formation of original localized micro-MHD vortices that may enhance the overall reaction kinetics (Figure 5). This type of hydrodynamic convection at very small scales may lead to a decrease of mass transfer resistance at such well-structured surfaces, which is not possible by conventional convection approaches (rotating disk electrodes or classic stirring). Finally, it is important to highlight that the interdisciplinarity of the concept and the resulting applications of the MHD effect should stimulate collaborative approaches to further study its universal impact on electrochemistry and also on more technologically orientated innovations.

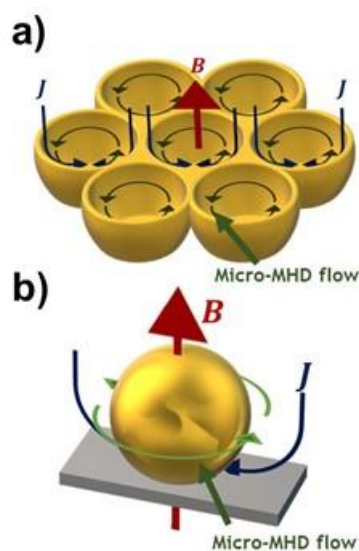


Figure 5. Illustration of micro-MHD vortices under the influence of an orthogonal homogeneous magnetic field; (a) inside the pores of a macroporous electrode, (b) around a metallic nanoparticle.

6. Acknowledgements

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· · First example of the possible use of MHD for optimizing the propulsion of self-electrophoretic macro- and micro Janus swimmers.

Graphical abstract

