

MHD PHENOMENA IN ELECTROCHEMISTRY

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Physical phenomena underlying magnetochemical processes and classification of these processes in accordance with the influence of the magnetic field on the diffusion kinetics (magneto-electrolysis) and the phenomenon under the influence of the Lorentz force (dynamic magnetolysis) and Faraday's electromagnetic induction (static magnetolysis) are considered. Parameters for practical realization of magnetochemical processes are determined.

Key words: magnetochemical chemistry, magneto-electrolysis, magnetolysis.

Introduction. Magnetochemical chemistry is a relatively new and rapidly developing field of electrochemistry, dating back about 40 years. During this time, its main directions have been determined, including the study of the effect of magnetic fields (MF) of different nature on electrochemical (transition reaction) and diffusion kinetics (mass transfer). Convincing evidence of the MF influence on the transition reaction has not been found yet. The effect of MF on diffusion kinetics manifests itself in creating forced convection in electrolyte by the Lorentz force. This increases the current density and, hence, the productivity of the process. In addition, the MF effect can change the structure, morphology, and physical properties (for example, magnetic properties) of sediments. To determine the mechanism of complex electrochemical reactions, a magnetic impedance method (MIM) has been developed [1].

When using a gradient MF in the presence of para- and diamagnetic ions in electrolyte, it turns out to be possible to control the composition of galvanic alloys [2]. This may find a wide application in applied electrochemistry.

Many electrochemical reactions occur with gas release (for example, water electrolysis). In this case, the bubbles are electrically charged and are acted upon by a lifting force and a Lorentz force. This creates additional convection and enhances mass transfer. The phenomenon can be used for the development of MHD propulsors for sea vessels and for the separation of the gaseous phase from the liquid in the absence of gravity (for example, in the spacecraft for oxygen evolution by water electrolysis) [3].

1. Magnetochemical chemistry.

Magnetochemical chemistry is a relatively new field of electrochemistry and its terminology has not been defined yet. By magnetochemical chemistry, we mean a branch of physical chemistry that studies electrode processes which occur both when the electrodes are polarized from an external current source with the simultaneous action of an external magnetic field or without polarization from an external current source but only due to the action of the magnetic field. Due to the smallness of the effect, we neglect the influence of the geomagnetic field ($\sim 5 \cdot 10^{-5}$ T) and the intrinsic MF of electrolyzers in the electrochemistry of aqueous solutions (in melt electrochemistry, for example, at the electrolysis of aluminum, magnesium, and other metals, we cannot neglect the intrinsic MF which reaches an induction of about 10^{-2} T [4]).

2. Magneto-electrolysis and magnetolysis.

Magneto-electrolysis is the name given to the decomposition of chemicals by the electric current in the presence of a magnetic field [5]. Several works have been published on magneto-electrolysis [6–9]. In [6], a significant influence of the magnetic field on mass transfer in the near-electrode layers and hence on diffusion kinetics, a weak influence on electrochemical kinetics (transition reaction) and a noticeable effect on the structure and morphology of deposits were noted. The usefulness of numerical modeling of electrochemical processes to determine their mechanism is also noted. In [10], authors propose a method to numerically simulate magneto-electrolysis problems. In [7], an increase in the limiting current was also noted due to the creation of forced convection by the Lorentz force in proportion to $B^{1/3}$, where B is the magnetic field induction.

For paramagnetic and diamagnetic components of electrolytes, the emerging Kelvin force F_K (magnetic field–gradient force or magnetophoretic force) is considered, determined by the formula

$$F_K = \frac{1}{2\mu_0} C \chi_m \Delta B^2,$$

where μ_0 is the magnetic constant in free space, C is the concentration, χ_m is the molar magnetic susceptibility. This force initiates convection which drives paramagnetic species ($\chi > 0$) towards a high magnetic field gradient and diamagnetic species ($\chi < 0$) towards a low magnetic field gradient. The order of magnitude of this force varies 10^3 to 10^{6-7} N/m³. The influence of the alternating magnetic field (AMF) and spin-dependent electrochemistry is also considered.

In [8, 9] the influence of a uniform and a non-uniform magnetic field on electrochemical processes is briefly considered.

Naturally, the areas of application of magnetic fields in physical chemistry and, in particular, in electrochemistry, both theoretical and applied, can be much wider than those noted in [6–9]. The present study is devoted to this issue.

Magneto-electrolysis is also called magnetolysis. Magnetolysis, i.e. electrolysis in the presence of an externally applied MF is a new field [11]. This introduces confusion into the terminology.

In our opinion, the decomposition of electrolytes only due to an external MF without polarization of electrodes from the current source should be called magnetolysis. This accurately reflects the physical nature of the phenomenon. During electrolysis, the polarization of the electrodes occurs from an external current source, and during magnetolysis due to Lorentz forces or locally due to Faraday's electromagnetic induction. This is the difference between magneto-electrolysis and magnetolysis. As for the electrode reactions themselves, they proceed in all cases in full accordance with the laws of classical electrochemistry since they are electrochemical in nature.

Magnetolysis, in turn, should be subdivided into dynamic (Lorentzian) and static or induced (Faradaic). The use of magneto-electrolysis in technology makes it possible to intensify many electrochemical processes and it is quite well studied [12]. The application of an external MF initiates strong tangential electrolyte flows (forced convection) in the near-electrode regions, i.e. it affects the hydrodynamics of the process and, consequently, the diffusion kinetics (Fig. 1). For example, in copper hydro electrometallurgy, the application of MF makes it possible to increase the cathode current density by a factor of $2.5 \div 3.0$ without deteriorating the quality of copper [13]. As regards the electrode reactions directly, under the conditions of magneto-electrolysis (low ion velocities and weak magnetic fields), the magnetic field practically does not affect them.

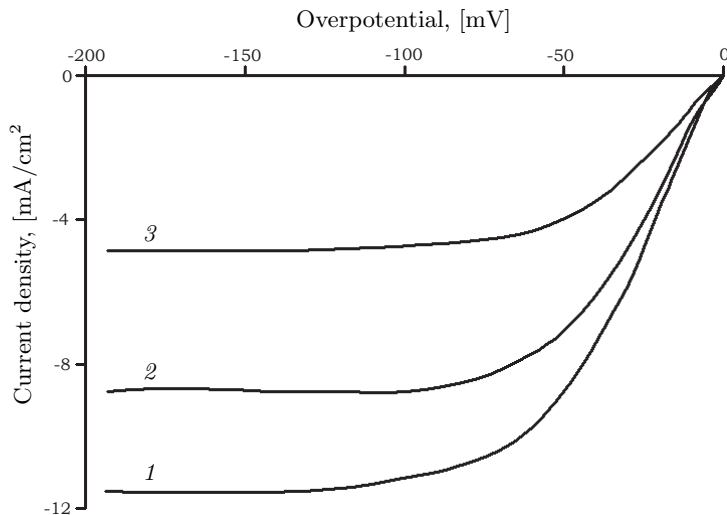


Fig. 1. Experimental current-voltage curves for a $\text{Zn(OH)}_4^{2-}/\text{Zn}$ system in KOH 7 M plotted for different B : 1 - $B = 0.6$ T, 2 - $B = 0.3$ T, 3 - $B = 0$ T, $C_{\text{Zn(OH)}_4^{2-}} = 0.2$ M, $C_{\text{KOH}} = 7$ M, $v = 20$ mV/min, overpotential vs. HgO/Hg. Adapted from [1].

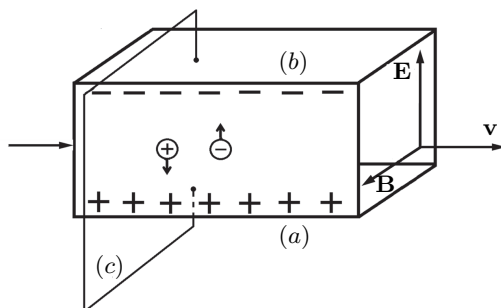


Fig. 2. Schematic of dynamic magnetolysis: a channel through which an electrolyte flows at a rate v ; a steady magnetic field of induction \mathbf{B} is applied parallel to the electrodes (a), (b); \mathbf{E} is the emerging electric field, c is the short circuit, \ominus - anions, \oplus - cations.

Dynamic magnetolysis. The idea of polarization of electrodes when an external magnetic field acts on a moving electrolyte belongs to Faraday [14]. He tried to polarize two platinum electrodes by dipping them from the bridge into the river Thames, while the geomagnetic field of the Earth served as the MF source. He had failed to detect any effect. If the experiment had been successful, electrochemistry would have probably developed further in two directions, and magnetochemistry would have reached the same perfection as classical electrochemistry.

Dynamic magnetolysis is based on the deflection of moving ions under the influence of an external magnetic field. If an electrolyte is pumped through a channel with parallel short-circuited electrodes located inside it and an external magnetic field is applied, then due to Lorentz forces, velocity components of oppositely charged ions moving towards the opposite electrodes appear (Fig. 2). The electrodes are polarized. At certain flow rates v and magnetic field induction \mathbf{B} , the electrolyte decomposition voltage is reached, and electrolysis occurs. In this case, the same goal is achieved with a stationary electrolyte

but with a moving magnetic field. The relative motion of the electrolyte and the magnetic field are important.

According to Kelly [15], the magnetic potential difference is $\Delta\varphi_{\text{mag}} = 2Bav$, where a is the distance between the electrodes. At $v = \text{const}$, the magnetic potential difference $\Delta\varphi_{\text{mag}}$ depends linearly on B . This has been confirmed in many experiments. Thus, according to [7], at $v = 0.65 \text{ m/s}$, $\Delta\varphi_{\text{mag}}/B \approx 0.12 \text{ V/T}$.

To reach a water decomposition voltage of 1.68 V, a magnetic field with the induction $B = 14 \text{ T}$ is required. Such magnetic field is difficult to induce. But already at $v = 4 \text{ m/s}$ and $B = 1.3 \text{ T}$, this is the range of really achievable MFs.

Due to the fact that it is based on the phenomenon of electromagnetic induction, static magnetolysis makes it possible to decompose electrolytes also without electrode polarization from an external current source, but, unlike dynamic magnetolysis, under static conditions [16, 17].

The idea of induced magnetolysis is in the predominant interaction of an alternating magnetic field with electrodes when electrons are charge carriers, and in a negligible interaction with the electrolyte, where ions are charge carriers.

According to the Faraday's law of electromagnetic induction, the electromotive force in the loop is $\mathbf{E} = -d\Phi/dt$, where $\Phi = \mathbf{B}\mathbf{S}$ is the magnetic flux through the surface S of the loop. At a certain value of the average MF induction, the potential difference between the loop-shaped electrodes reaches the electrolyte decomposition voltage and magnetolysis occurs. Current rectification is easily achieved by placing diodes between the electrodes or by making one of the electrodes from the valve metal, or by imposing an alternating pulsed magnetic field which is created by a coil powered by a rectified asymmetric sinusoidal current.

The current density in the loop is $J = nev = neuE$, where n is the number of charge carriers per unit volume, e is the electron charge, v is the average velocity of their ordered motion, u is the electrical mobility of the charge, E is the electric field strength.

It is known that the mobility of free electrons, for example, in copper, is 10^4 times higher than the mobility of H^+ and OH^- ions, and that the electron concentration exceeds the concentration of ions (in the case of a 35% KOH solution) by about 20 times. This causes the predominant interaction of the alternating magnetic field with the electronic conductor.

The block diagram (Fig. 3) shows the real and possible applications of MHD effects that occur in electrochemical systems when exposed to an external magnetic field of different nature.

It should be noted that the influence of an external magnetic field directly on the electrode reaction (transfer reaction) has not been strictly proven yet despite the fact that this effect was predicted back in 1973 [18]. This is the most interesting field of theoretical research. There is a small effect of up to 10 Oe MF on the processes in electrochemical systems which may take place on condition of electron transfer through the phase boundary metal–electrolyte at a microscopic level.

This instrumental method of experimental electrochemistry, which enhances the possibility of electrode reactions and double layer structure study, has not found wide application at present. Whereas this method is sufficiently simple and the magnetic field effect on electrochemical kinetics may be easily discovered, the quantization of the energy of electrons, taking part in electrochemical reactions, determines these phenomena. This influence probably may change the electrode reaction rate. The change may be both monotonic and periodic, depending on the orientation of the magnetic field.

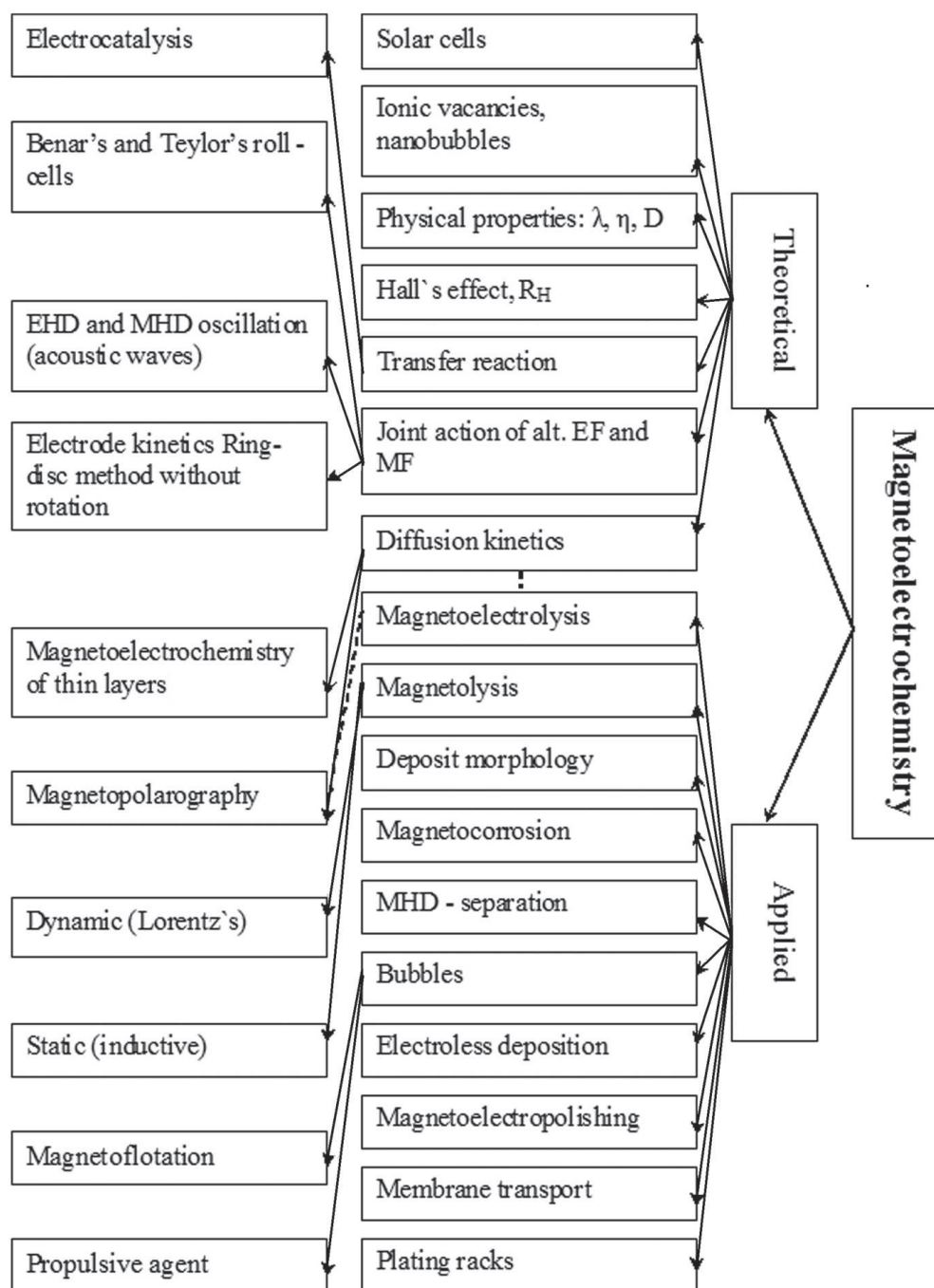


Fig. 3. Block diagram of possible use of MHD phenomena in theoretical and applied electrochemistry: electrical conductivity λ ; viscosity η ; diffusion coefficient D , Hall constant R_H ; electrohydrodynamic (EHD); electric field (EF).

In [1], a new method of electrochemical analysis (magnetic impedance method), namely, the MHD transfer function measurement has been developed with the use of the magnetic field. This technique allows us to analyze electrochemical systems with complex mechanisms, such as zinc or nickel electrodeposition, and to provide information about mechanisms occurring at operation.

3. Diffusion kinetics.

As to the influence of the magnetic field on the diffusion kinetics, morphology, structure, and various properties, for example, magnetic ones, of deposited metals, several hundred papers have been published over the past thirty years.

The magnetic field also has a macroscopic (phenomenological) action on electrochemical reactions. It greatly increases the velocity of mass transport in electrochemical processes. The magnetic field acts more strongly when the process is controlled by mass transport, i.e. in the diffusion-controlled region the MF influence may be determined by the MHD effect. This influence leads to the appearance of vortex flows in the diffusion layer. There are three regions of the MF influence.

1. Weak MF convective diffusion plus small contribution of the MHD effect.
2. Medium MF. Volume forces are generated. Mixed free and forced convection.
3. Strong MF. Mass transport in the diffusion layer is determined by MHD phenomena.

In [19–21], the reasons of the appearance of volume forces in the electrolyte at water electrolysis (30% KOH solution) under the action of a uniform magnetic field of different orientation with an induction of 0.8 T on the development of convective electrolyte flows are considered. The flows enhance the mass transfer and, as a result, the productivity of the process. Diagrams of emerging Lorentz forces in the overall balance and their comparative assessment are given.

4. Bubbles.

Also in [19–21], the significant role of the forces acting on the emerging charged gas bubbles in the near-electrode zones was found. The nature of negative charges on hydrogen bubbles due to the emission of electrons from the cathode into the bubble-electrolyte interface was explained. It was more difficult to explain the appearance of a positive charge on oxygen bubbles. It was assumed that such charge arose due to the formation, like for semiconductors, of positively charged holes. This idea was confirmed in [22]. The idea of the possible formation of ion vacancies in the near-electrode zones during the electrolysis of electrolyte solutions (like solid electrolytes), which are the cause of the formation of nanobubbles, was suggested and experimentally confirmed in that study. These are negatively or positively charged vacuum voids with a size of about 0.1 nm and a lifetime of ~ 1 s. Due to the induced Lorentz force, ion vacancies circulate together with the solution. The coalescence of nanobubbles leads to the formation of microbubbles, and their motion can be observed optically.

Regardless of the nature of charge formation on microbubbles, their role in creating forced convection of the electrolyte is in good agreement with the diagrams of emerging volume forces [19–21]. The assumption of the appearance of ion vacancies with the subsequent formation of charged nano- and microbubbles [22] serves as an additional proof of the significance of charged bubbles in the development of forced convection.

5. Applied electrochemistry.

The diagram (Fig. 3) presents the most important practical processes.

It is dynamic magnetolysis. Boese convincingly proved in the patents “Magnetodynamische autoelectrolyse” [23–25] the possibility of realization of dynamic magnetolysis. Boese had developed very original equipment for water splitting. He showed that a voltage of 10 V could be realized in a channel, where the sea water flows with a velocity of 20 m/s, and a magnetic field induction of 0.5 T is used.

The prime cost of the hydrogen production by magnetolysis is lower than by electrolysis. It has specific advantages of good seal.

It should be noted that under magnetolysis conditions, hydrogen and oxygen overpotentials are decreased by removing gas bubbles from the electrodes in the electrolyte flow. The specifications of Boese the electrolyzer are the following:

- electrolyte – 35% KOH,
- flow velocity – 4.0 m/s,
- current density – 0.75 kA/m²,
- magnetic induction – 1.3 T,
- efficiency – 69%.

Dynamic magnetolysis also includes the development of MHD propellers [26, 27]. The main difficulty in this project is the selection of the anode material. Boissonneau [26–27] showed that even platinized titanium used as an anode material breaks down. The author did not take into account the role of charged nano- and microbubbles of gas, formed during electrolysis, in creating propulsion thrust.

Static (inductive) magnetolysis is discussed above.

6. Other applications of MHD phenomena.

The joint action of alternating electric and magnetic fields results in undamped EHD and MHD oscillations. This effect can further intensify the processes. Due to the vortex flows of the electrolyte, there appears a possibility to investigate the electrode kinetics by the ring-disk method without mechanical rotation of the electrode [28].

Magnetopolarography is a physicochemical investigation method, implying that the cell is influenced by both polarizing voltage and external magnetic field. It may be used for the investigation of electrode layers, reaction kinetics, analytical purposes, etc.

Alternating pulsed fields may be used in magnetopolarography as an impedance meter method without external polarizing voltage when the modulation of the double layer capacity is implemented by an alternating magnetic field.

In *magnetocorrosion*, the magnetic field can both stimulate corrosion when $\mathbf{B} \perp \mathbf{E}$ and inhibit it when $\mathbf{B} \parallel \mathbf{E}$. The action of the magnetic field on corrosion is schematically illustrated in Fig. 4. Corrosion in liquid Pb17Li exposed to a magnetic field is discussed in [29].

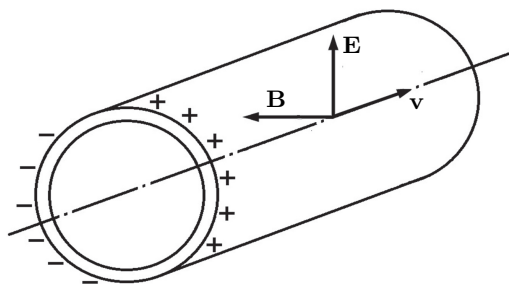


Fig. 4. Corrosion of the anodic sections of the pipeline through which an electrically conducting liquid flows at a rate \mathbf{v} under the action of a magnetic field $\mathbf{B} \perp \mathbf{v}$.

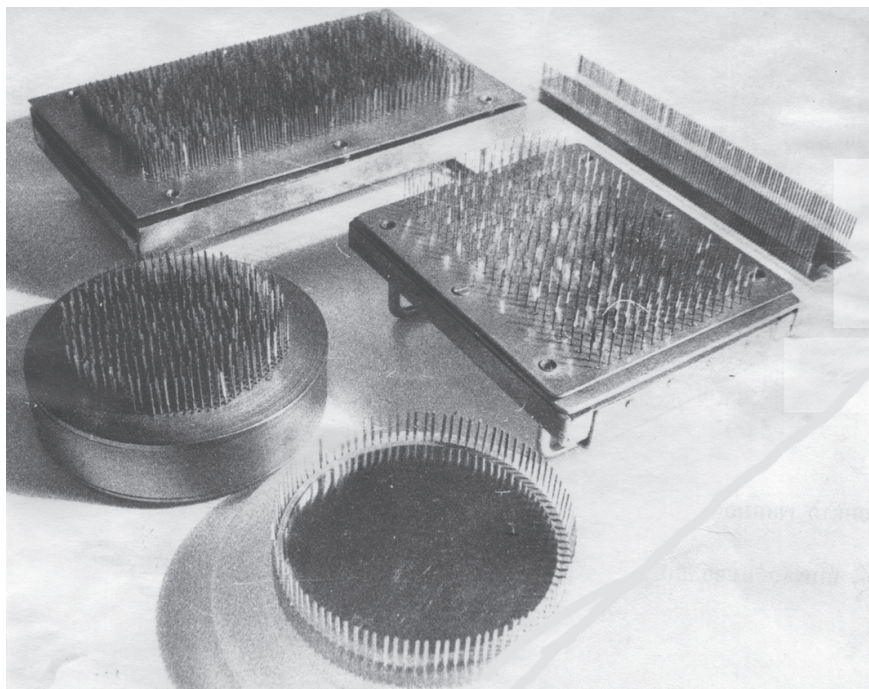


Fig. 5. Plating racks.

Electroless deposition. Also the magnetic field influences electroless deposition. In [30, 31] it is shown that the action of a magnetic field of 0.5 T on the chemical nickel-plating process increases the hardness of the coating due to the higher content of phosphorus. In addition, the morphology and microstructure of the sediment changes. However, no theory of this effect has been worked out yet.

Plating racks. We have developed several magnetic plating racks [32–35] (Fig. 5).

They make it possible to introduce automation into the process of assembling and dismantling ferromagnetic and nonferromagnetic parts and are very convenient in operation. Besides, magnetic fields pass along the ferromagnetic parts to the electrode reaction zone and favorably affect the process.

Conclusions.

The most important problem in magnetochemistry is the problem of determining the action of the magnetic field directly on the electrode reaction, i.e. on the transition reaction. However, neither convincing evidence of this action has been obtained. For interpretation, the magnetic field influence on the diffusion kinetics of electrochemical processes by initiating forced convection in the electrolyte and enhancing mass transfer in the near-electrode layers is important. This contributes to the increase of the current density and process productivity. The main attention was paid to the magnetohydrodynamic processes occurring in the electrolyte due to emerging Lorentz volume forces. However, when studying the nature of such forces, insufficient attention has been paid to the effect of charges on gas microbubbles that appear during electrolysis. Particularly fruitful can be the idea and experimental confirmation of Japanese scientists on the occurrence of ion vacancies in the electrolyte solution, which form nanobubbles upon

coalescence, which also carry an electric charge and can be the cause of emerging MHD flows.

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