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Mini Review

Research Progress of Magnetic Field Techniques for Electrodeposition of Coating

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Recent advances in electrodeposition techniques under magnetic fields are introduced. The mechanism of magnetic fields to electrochemical deposition technology, including mass transfer, coating structure modification, electrochemical reaction and crystal orientation are reviewed. Current research results show that MHD effect induced by the Lorenz force and the magnetization effect are the main factors. The research situation of single metal layer, alloy layer, composite coatings and high entropy alloy layer under magnetic fields is summarized. Finally, and the problems existing in current research of magnetic electrochemistry are pointed out, and the development prospect of controllable magnetoelectrodeposition is also proposed.

Keywords: Electrodeposition; Magnetic field; MHD effect; Magnetization effect; Prospect

1. INTRODUCTION

Electrodeposition technology is to take the substrate material as the cathode in aqueous solution, or non-aqueous solution or molten salt, and use the discharge and reduction of metal ions on the cathode surface to obtain a metal coating. At present, it is a relatively mature and widely used material preparation method, and can significantly improve the wear resistance, corrosion resistance, high temperature oxidation resistance, conductivity, catalysis and other functions of the matrix material[1-2]. At the same time, electrodeposition technology has a series of advantages, such as simple operation, low process cost, good process flexibility, etc., so it is widely used in surface engineering, nanowires, part forming, new material preparation and many other fields[3-4].

In recent years, as an important material preparation method, the introduction of magnetic field in the electrodeposition process has become a new branch of electrochemical science, and it is called magneto-electrodeposition, which can be widely used in electrodeposition of metals, alloys and composite coatings[5-9]. At present, the influence of magnetic field on electrodeposition is mainly attributed to the interaction of magnetic field and current, which promotes solution convection, namely, magnetohydrodynamic(MHD) effect[10-11]. The MHD effect will affect the mass transfer, adsorption desorption, electron transfer and other processes in the electrodeposition process. In addition, MHD effect has significant influence on the properties of electrolyte, chemical reaction, physical and chemical properties of solution, current distribution on electrode surface, structure and properties of coating [12-13]. Since the 1990s, the electrodeposition under high magnetic fields has developed rapidly due to the application of superconducting magnetic field[14-15]. Magnetic field assisted electrodeposition has attracted more and more attention of researchers due to its advantages of easy control, non pollution, non-contact energy transfer, high energy density and high selectivity for the prepared materials[16].

In this paper, the research results of magnetic field on electrodeposition are summarized, and the correlated mechanism of magnetic field assisted electrodeposition process is clarified. Simultaneously, the existing problems in its application at present stage are expounded, and the further research of magnetoelectrochemistry is prospected.

2. MECHANISM OF ELECTRODEPOSITION IN MAGNETIC FIELDS

When electrodeposited in magnetic fields, ions or particles in electroplating solution are affected by multiple forces, such as diffusion force, natural convection force, Lorentz force and magnetic field gradient force, which has a significant impact on the mass transfer, grain nucleation and coating morphology, and then the comprehensive performances of coatings can be significantly improved(Figure 1).

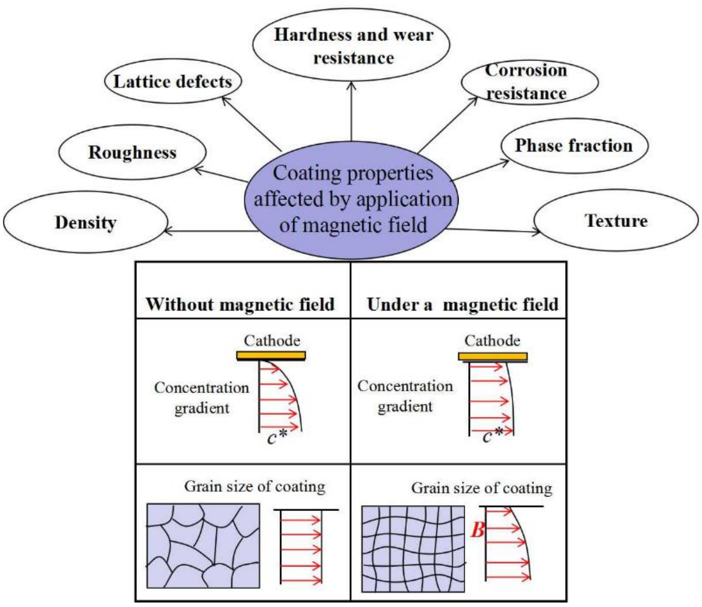

Figure 1. Influence of magnetic field on some properties of electrodeposited coatings

Hinds[17] analyzed the influence of magnetic field on the mass transfer process of various forces of ions in the plating bath under an 1T magnetic field intensity, as shown in Table 1. It indicated that the application of magnetic field, especially strong magnetic field, will have a significant impact on the electrodeposition process. The main influence mechanism of magnetic field on electrodeposition process is listed as following.

Table 1. Typical forces acting in aqueous electrolytes[17]

Force	Expression	Typical value(N/m^3)
Driving force for diffusion ($\vec{F}_D$)	$RT\vec{\nabla}c$	10^{10}
Driving force for migration	$zFc\vec{\nabla}V$	10^{10}
Driving force for forced convection	$\rho(r\omega)^2/2\delta_0$	10^5
Driving force for natural convection	$\Delta\rho\vec{g}$	10^3
Viscous drag	$\eta\nabla^2 v$	10^1
Paramagnetic force（$\vec{F}_P$）	$\chi_m B^2\vec{\nabla}c/2\mu_0$	10^4
Field gradient force（$\vec{F}_B$）	$\chi_m cB\vec{\nabla}B/\mu_0$	10^1
Lorentz force（$\vec{F}_L$）	$\vec{j}\times\vec{B}$	10^3
Electrokinetic force（$\vec{F}_E$）	$\sigma_d\vec{E}_{\parallel}/\delta_0$	10^3
Magnetic damping force（$\vec{F}_M$）	$\sigma\vec{v}\times\vec{B}\times\vec{B}$	10^1

B=1T，T＝298K, c=10^3 mol/L, δ=10^{-4} m, z=2，V=1V，ρ=10^{3}kg/m^3, d=10^{-2}m，ω=10^{2}rad/s，δ_0=10^{-3}m，$\Delta\rho$=10^2 kg/m^3, η=10^{-3}Ns/m^2，v=10^{-1}m/s，χ_m=10^{-8} m^3/mol，∇B=1T/m，j=10^{3}A/m^2，σ_d=10^{-1}C/m^2，$E_{//}$=10V/m，σ=10$^2\Omega^{-1}$/m^{-1}

2.1 Magnetohydrodynamics(MHD) effect

According to the different arrangement of magnetic field and electric field, it can be divided into parallel magnetic field electrodeposition (the direction of magnetic field is parallel to the current, B□J), perpendicular magnetic field electrodeposition (the direction of magnetic field is perpendicular to the current, B□J), and gradient magnetic field electrodeposition.

2.1.1 Effect of parallel magnetic field on Electrodeposition

Electrodeposition in parallel magnetic field means that the direction of magnetic field is parallel to the direction of current in the process of electrodeposition. Aogaki[18] found many mysterious "rings" with a diameter of about 1 mm on the coating surface in the process of Cu electroplating under a parallel magnetic field. It is thought that the "vortex" effect was caused by the fluctuation of the composition on the surface of the cathode. However, the deep-seated cause of the fluctuation of the composition was not explained. Asai[19] studied the electrodeposition of Ni-Al_2O_3 in a parallel magnetic field, and found that Al_2O_3 was distributed in a honeycomb shape in the coating, and it was believed that only particles with a particle size greater than 1 μm could have a honeycomb shape distribution on the coating under a high magnetic field. However, wang et al.[20] found that the nano-sized Al_2O_3 particles also can be formed a honeycomb distribution under an 10T magnetic field.

According to Faraday's law, when a parallel magnetic field is applied, there is no interaction between magnetic field(B) and current(J), that is, Lorenz force will not be generated. However, the cathode surface is not perfectly smooth, there exists micro unevenness, resulting in current distortion and a component(J_P) along the perpendicular direction of the magnetic field, and a Lorenz force (f_L) will be induced and can be expressed as:

$$f_L = B \times J_p \qquad (1)$$

According to formula(1), under the interaction of current and magnetic field, it will cause solution disturbance at the micro scale of cathode surface, forming the hydrodynamic effect, namely micro MHD effect, and its mechanism is shown in Figure 2. The Lorenz force on different grain surfaces was simulated by finite element method of COMSOL software, it is found that Lorenz force at the grain tip of metal convex structure is the largest, as shown in Figure 3[21]. At the same time, Zhou et al.[22] studied the composite electrodeposition behavior of Fe/nano-Si particles in parallel magnetic field, the distribution of Si particles on coating surface was a “ridge” structure. It was considered that there exsited a macro MHD effect near the edge of the electrode surface.

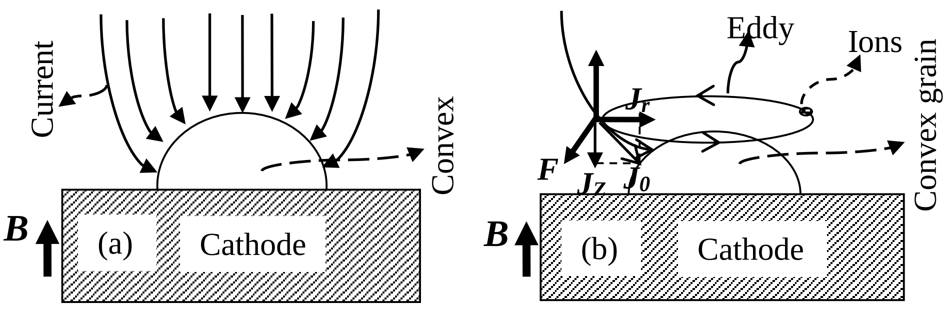

Figure 2. Schematic view illustrating the MHD effect in a parallel magnetic field. (a) the current distributions in front of the convex grain, (b) formation of micro electrolyte flows

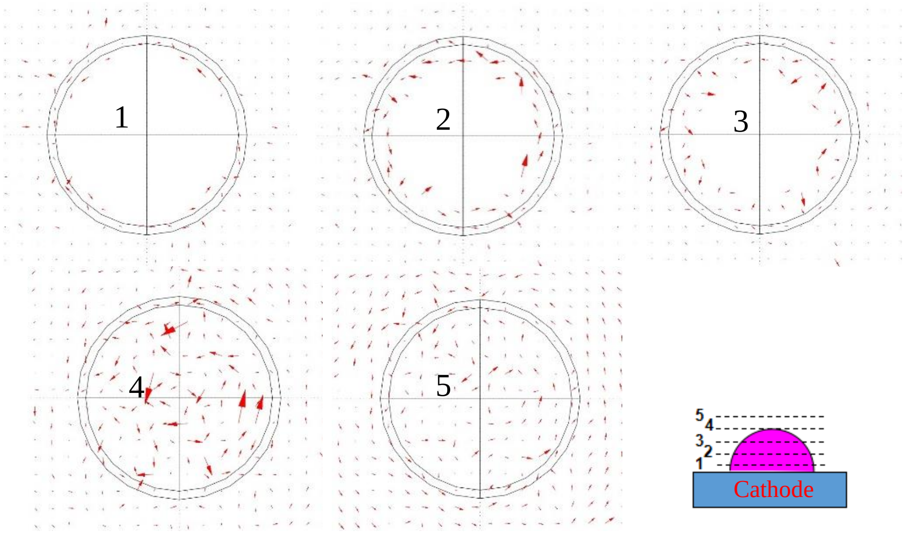

Figure 3. The MHD effect simulated by COMSOL on different depth planes(grain size) by electrodeposition in parallel magnetic field[21]

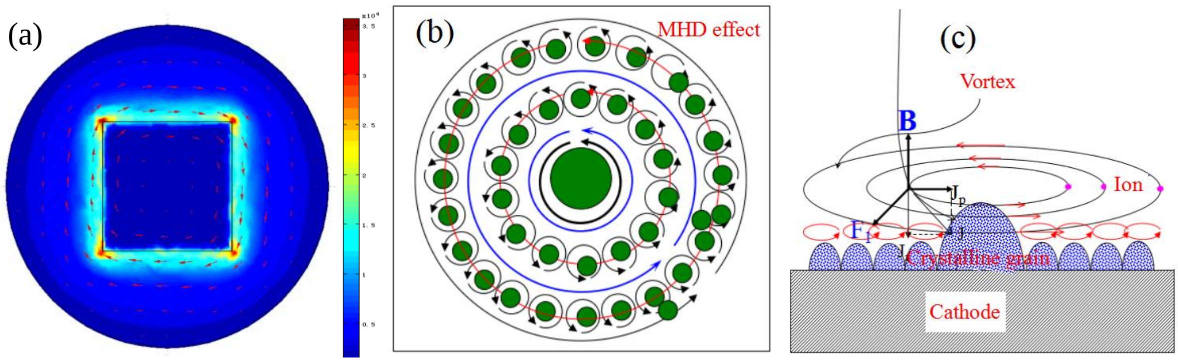

Figure 4. MHD effect induced by electrodeposition in parallel magnetic field (a) software simulates MHD effect simulated by COMSOL under 10T magnetic field; (b) A schematic diagram of MHD effect on cathode surface; (c) A schematic diagram of MHD effect on cathode side

At the same time, the results of simulation analysis by COMSOL software also show that there exist macro MHD effect in the process of electrodeposition, as shown in Figure 4. Morimoto[23] also theoretically clarified the nucleation process of electrodeposition, the suppression of three-dimensional nucleation by microscopic magnetohydrodynamic flow(micro-MHD flow) and the formation of stream pattern of macroscopic MHD flow by the multiple nucleation of three-dimensional nuclei.

2.1.2 Electrodeposition in perpendicular magnetic field

When in a perpendicular magnetic field, that means the direction of magnetic field is perpendicular to the current during the electrodeposition process. According to Formula(1), the Lorenz force induced by the interaction of magnetic field and current now is up to the largest, which will form a strong macro MHD effect(Figure 5). Both the micro MHD effect and the macro MHD effect can significantly improve the mass transfer and reduce the thickness of the diffusion layer near the cathode surface.

Therefore, micro MHD effect will affect the nucleation and growth of the grains during the electrodeposition process. Wen[24] found that strong MHD effect can significantly refine the grain size of the coating when the Ni-Fe films were electroplated in a high magnetic field. Long et al. [21,25] Found that the micron Si particles or Fe-Si alloy particles were distributed in a striped structure on the Fe-Si coating surface. At the same time, current efficiency also can be significantly affected, Matsushima[26] found that the current efficiency for electrodeposition of iron decreased significantly, from approximately 60% without magnetic field to 30% under a 5T magnetic field. It was thought that the external magnetic field also had a catalytic effect on hydrogen evolution reaction, which aggravated the hydrogen evolution.

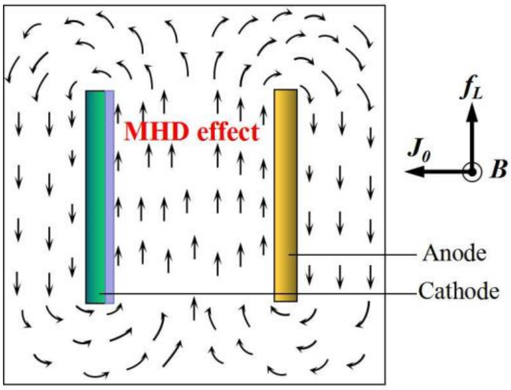

Figure 5. MHD effect induced by perpendicular magnetic field

2.1.3 Electrodeposition in gradient magnetic field

In the process of electrodeposition, when the applied magnetic field is an uneven magnetic field or the magnetizable material is used in the uniform magnetic field, there will be a magnetic field gradient(∇B) near the electrode, which will generate a magnetic field gradient force(F_b) on metal ions or plating solution particles, and the F_b can be expressed as[27]:

$$F_b = \chi_m \frac{B\nabla B}{\mu_0} C \qquad (2)$$

At the same time, the concentration of the local magnetic field will lead to a significant increase of magnetic field intensity, and the Lorentz force generated by the interaction between magnetic field and current will increase, which will greatly improve the convection of electroplating solution. As the consumption of metal ions, there exist a concentration gradient near the electrode surface when the diffusion convection near the electrode is relatively weak, and a concentration gradient forces(F_P) similar to the magnetic field gradient force will be generated[28-29]:

$$F_p = \frac{\chi_m B^2}{2\mu_0} \nabla C \qquad (3)$$

Where χ_m, μ_0, B, C, ∇B and ∇C are the molar susceptibility, vacuum permeability, magnetic field strength, body concentration of electroplating solution, magnetic field gradient and concentration gradient respectively.

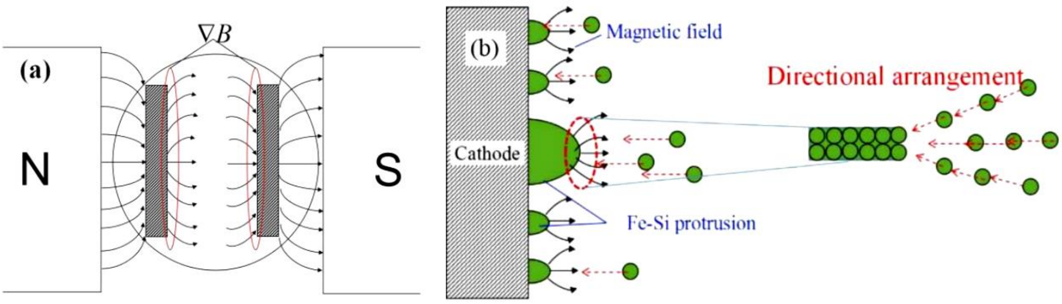

Figure 6. region of □B near the electrode surface(a) and directional arrangement of magnetic field particles(b)[21]

During the process of electrodeposition in magnetic fields, paramagnetic ions (or particles) will move to the areas with high magnetic field intensity under the action of magnetic field gradient force, while diamagnetic ions(or particles) will be excluded from these zones, which will also change the concentration of hydrated particle near the electrode. Long[21, 30-31] studied the migration behavior of $Fe_{\chi}Si$ in the preparing of Fe-Si composite coatings in magnetic field. It was found that the magnetic field gradient force plays a leading role in the electroplating under an relatively weak magnetic field, and the Fe-Si alloy particles are arranged in "needle" structure in the coating. When the magnetic field is higher than 0.5T, the Fe-Si alloy particles are arranged in "dome" structure in the coating surface due to the synergistic effect of magnetic magnetic field gradient force and MHD effect(Figure 6). In addition, Arghavanian et al.[32] modified ZrO_2 particles by electroless plating with a layer of Ni and increased the magnetic properties of the particles, thus significantly increasing the particle content in the Ni coating.

2.2 magnetization effect

In the environment of external magnetic field, during the electrodeposition process, the material will also be affected by the magnetizing force. The magnetizing force F_M can be expressed as [33]:

$$F_M = \mu_0 (M\cdot\nabla)H = \chi\mu_0(M\cdot\nabla)H = (\chi/\mu_0)(B\cdot\nabla)B \propto B^2 \quad (4)$$

Where μ_0 is the vacuum permeability, M is the magnetic moment, H is the magnetic field intensity, B is the magnetic induction intensity, χ is the susceptibility. Where $\chi=M/H$ indicates the difficulty of magnetization.

Due to the magnetic anisotropy of the crystal, the magnetization force can also affect the texture orientation of the crystal in the process of electrodeposition. Under the action of magnetic field, the crystal tends to take the texture orientation towards the easy magnetization axis. Feng[34] studied the electrodeposition process and coating properties of $Ni\text{-}Al_2O_3$ in magnetic field. The results showed that with the increase of magnetic field intensity, the pure nickel layer had a (200) preferred orientation, while the $Ni\text{-}Al_2O_3$ composite coating had a (110) preferred orientation. Matsushima et al.[35] have found that iron grains are arranged along the direction of magnetic field during nucleation and growth, and with the increase of magnetic field intensity, the coatings have the trend of preferred orientation along the (110) direction, which is mainly due to the material's magnetic anisotropy, and this is favorable for the preparation of magnetic materials.

2.3 magnetothermodynamic effect

The external magnetic field can not only significantly affect the physical and chemical reaction process of materials, but also adjust the micro state of materials, such as reaction heat, pH value, direction of chemical reaction, reaction rate, activation energy, entropy and so on, and then greatly improve the coating performance[36-38]. In the process of electrodeposition, the metal cations on the cathode surface are reduced to metal atoms after obtaining electrons, which can be expressed as follows:

$$M^{n+} + ne \rightarrow M \qquad (5)$$

It is easy to cause the side reaction of hydrogen evolution when the reduction potential of metal ions is relatively negative. First, H^+ becomes the adsorbed hydrogen atom(H_{ad}) after obtaining an electron, and then combine together to form hydrogen(H_2):

$$H^+ + e \rightarrow H_{ad} \qquad (6)$$

$$H_{ad} + H_{ad} \rightarrow H_2 \uparrow \qquad (7)$$

After the magnetic field is applied, additional energy is provided for the reaction. The reaction is no longer a simple temperature control, but also the contribution of magnetic Gibbs free energy to the reaction. The unit volume of magnetic Gibbs free energy(G_M) can be expressed as[39-40]:

$$G_M = -\frac{\chi_v B^2}{2\mu_0} \qquad (8)$$

Where G_M represents the magnetic Gibbs free energy(J•mol^{-1}), χ_v is the volume susceptibility of the material, B is the magnetic field intensity(T), μ_0 is the vacuum susceptibility(m•H^{-1}). In the process of material transformation, when the magnetic Gibbs free energy of the new phase and the parent phase changes, the direction of phase transformation tends to be consistent with the direction of the reduction of the magnetic Gibbs free energy in order to maintain the internal energy balance of the system, that is, the magnetothermodynamic effect. The difference of magnetic Gibbs free energy of reaction(5-7) may be more negative after the magnetic field is applied, resulting in the lowest total energy of the product of the system, and thus promoting the reaction[41-43].

3. GENERAL SITUATION OF MAGNETIC FIELD IN ELECTRODEPOSITION

In recent years, the electrodeposition technology under the magnetic field has attracted more and more attention, and the magneto-electrodeposition can be divided into single metal coating, alloy coating, composite coating and high entropy alloy coating according to the type of coating.

3.1 Preparation of single metal coating by electrodeposition in magnetic field

Because of the simple operation, low cost, excellent coating performance and mature technology, the single metal plating process has been widely developed and applied. The research of introducing magnetic field into the single metal electrodeposition process is relatively early and mature, and it can obtain a variety of metal coatings with excellent performance compared with the traditional electroplating technology. At present, the single metal electrodeposited under magnetic field mainly includes Ni, Cu, Cr, Fe, Co, Zn, Sn, Bi, Pb, etc.[44-45]. Ooi et al.[46] studied the electrodeposition process of copper film under the action of perpendicular magnetic field. It was found that the surface roughness of deposition layer became smoother due to the agitation of micro-MHD effect induced by Lorentz force.

3.2 Electrodeposition of alloy coating under magnetic field

In the process of alloy electrodeposition, two or more metals (or nonmetals) are electrodeposited. Compared with the single metal coating, the alloy coating often has better hardness, corrosion resistance, wear resistance, high temperature oxidation resistance, magnetism, aesthetics and other properties. Therefore, it is widely used in the decoration and preparation of some functional materials.

At present, the study of alloy co-deposition mainly improves the performance of the coating by adjusting the composition and microstructure of each element in the coatings. During the electrodeposition process, the characteristics of the magnetic field can be used to adjust the composition, grain size and morphology of the coatings[47-48]. Yu[49] studied the electroplating of Ni-Zn alloy coating in magnetic field. It was found that with the increase of magnetic field intensity, the Ni composition in the coating increased from 5% without magnetic field to about 21wt% under an 1T magnetic field, and the grain size of the coating significantly decreased. It was believed that the MHD effect enhanced the mass transfer of the plating solution. Aaboubi[50] studied the electrodeposition of Co-Ni-Mo alloy under a stable magnetic field. The results show that the coating becomes compact and smooth due to the influence of MHD convection, the grains are significantly refined and distributed evenly. At the same time, with the increase of magnetic field intensity, the deposition speed of metal ions increases, and the Co and Ni content of the coating increase significantly.

3.3 Preparation of composite coating by electrodeposition under magnetic fields

Composite electroplating is that the metal atoms and the inert particles dispersed in the plating solution were co-deposited when the solution ions are discharged and reduced, and the inert particles are captured into the coating matrix. It has the advantages of simple equipment, low temperature, simple operation, low cost, easy control, etc., which can not only improve the corrosion resistance, wear resistance, high temperature oxidation resistance of the metal surface, but also save materials. Therefore, composite electroplating technology can be widely used in electronics, aviation, machinery, chemical industry, metallurgy and many other fields[51-52].

The content and distribution of second-phase particles in composite coating is a key factor to determine the performances of composite coating. How to improve the content and distribution of the inert particles in composite coating has become a hot issue. When the magnetic field is introduced into the co-deposition process, the MHD effect can play a role in stirring the electroplating solution, and significantly reduce the thickness of the dispersion layer and the diffusion layer, thus facilitating the particles to enter the coating(Figure 7)[53]. At the same time, for diffusion limited processes the MHD effect causes an increase in the limiting current density($I_{\lim}$)[29]. Aogaki et al.[54] showed that the limiting current density increases proportional to $c^{*3/4}B^{1/3}$ by solving Navier-Stokes equation(Eq. 9) and this has been confirmed by other authors[55-56]:

$$I_{\lim} = 4.3\cdot 10^{3} \cdot n^{3/2} A^{3/4} D v^{-1/4} c^{*4/3} B^{1/3} \quad (9)$$

Where n denotes the number of electrons involved in the electrochemical reaction, A is the surface area of the electrode, D and c are the diffusion coefficient and bulk concentration of electroactive species in the electrolyte solution respectively, v is the viscosity of the electrolyte, B is intensity of applied magnetic field(parallel to the electrode surface). As a result, an enhanced convection in the solution will be induced. The stirring intensity induced by MHD effect can even reach the mechanical stirring intensity under the strong magnetic field, so that the nano or even submicron inert particles can be uniformly and stably suspended in solution. As the result, the mass transfer process of particles is accelerated and can prevent the nano or submicron particles from agglomeration to a certain extent. At the same time, the particle content in the coating can be increased significantly while its distribution is affected, then the comprehensive properties of the composite coatings will be improved[27, 57-58].

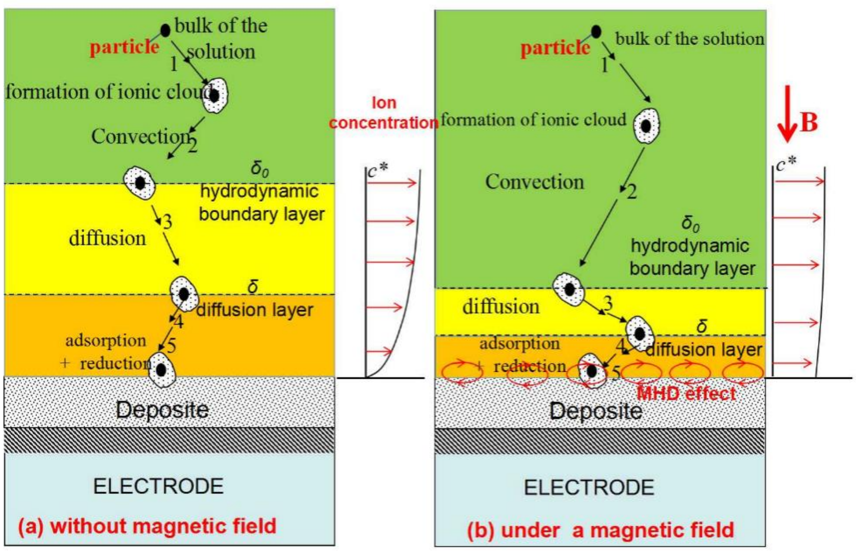

Figure 7. Schematic diagram of particle migration during electrodeposition without magnetic field and under a magnetic field[53]

Therefore, researchers worldwide have done a lot of work on composite electroplating in the magnetic field. Fu[59] has studied the composite electrodeposition process of Ni-SiC in the magnetic field. The results show that the MHD effect induced by the interaction of current and magnetic field significantly improves the mass transfer of the plating solution, the SiC particle content in the composite coating increases significantly, from 1.22wt% without magnetic field to 3.26wt% under a 0.3T magnetic field. Zhou et al.[60] found the mass fraction of Si particles in the coating can be increased from 10.4wt%

without magnetic field to 20.17wt% under a 0.2T magnetic field. At the same time, the coating surface appears many "ridge" structures with the extension direction of of MHD effect hydrodynamics.

3.4 Preparation of high entropy alloy coating by electrodeposition in magnetic field

High entropy alloy(HEA) is usually composed of Ni, Cr, Co, Ti, Cu and other elements, which not only has excellent mechanical properties, but also has good corrosion resistance, showing a great application potential in various fields[61-62]. Yao et al.[63] prepared $Fe_{13.8}Co_{28.7}Ni_{4.0}Mn_{22.1}Bi_{14.9}Tm_{16.5}$ amorphous HEA coating by electrodeposition. The surface of the coating was granular structure, but the formation and growth of the crystal nucleus on the substrate surface could not be well controlled, so the film was mostly polycrystalline or amorphous structure. Yao[64] also prepared BiFeCoNiMn high entropy alloy film by electrodeposition in N, N-dimethylformamide (DMF). The surface of the film is compact and shows soft magnetic behavior, but after annealing it shows hard magnetic properties. At present, there are few researches on the preparation of high entropy alloy coating by electrodeposition, which is mainly due to the large difference of electronegativity of the elements in HEA, which makes the composition of HEA difficult to control. At the same time, the diffusion limited processes of electroplating solution make the coating is easy to crack, thus badly affecting its comprehensive performance.

In recent years, the rapid development of magnetron electrochemistry has been achieved. It is main accepted that the MHD effect caused by the interaction between magnetic field and electric field can greatly enhance the convection of electrolyte solution[21,25,31]. Due to the high viscosity of organic solvent, MHD effect can significantly improve the mass transfer of electroactive species, especially near the cathode surface, thus affecting the performance of the coating. Therefore, we can try to use non-aqueous system electrodeposition under magnetic fields to prepare some series of HEA coatings with excellent performances.

4. PROBLEMS AND PROSPECT IN THE APPLICATION OF MAGNETIC FIELD IN ELECTRODEPOSITION

According to the current research situation and existing problems of magnetic field in electrodeposition, the research trend in this field is summarized as follows:

(1) At present, most of the experiments are about the single effect of magnetic field on the electrodeposition process, but the experimental research on the composite effect of magnetic field and other physical fields(such as ultrasonic field, electric field, microgravity or supergravity field and mechanical agitation) and high pressure intensity has not been fully carried out, and the mechanism of the interaction effect of multiple physical fields is not clear[65-67]. At the same time, the finite element analysis software such as COMSOL and ANSYS should be strengthened to analyze the interaction effects of multi-physical fields.

(2) The research on electrodeposition mainly focuses on the conventional aqueous solution electroplating, whereas the related researches on ion plating, metal electrodeposition with high

electronegativity in organic solvent and electrodeposition of high entropy alloy under magnetic fields are very few[68-69]. Therefore, we should strengthen the research in this field in order to grasp the influence mechanism of magnetic field on the electrodeposition process more comprehensively and deeply.

(3) The current research of electrochemistry in magnetic fields mainly focuses on the conventional scale electrochemical deposition, but the mechanism research in the micro scale space is relatively few, so the research in this field needs to be further expanded, such as using the technologies of synchrotron radiation, rasound Doppler velocimetry(UDV) measurement and professional high-speed camera to visualize the electrodeposition process from the micro perspective[70-71].

(4) Simultaneously, magnetic field has a significant effect on the composition distribution, grain size, texture orientation, precipitation and distribution of the second phase in the solution process of the coatings[72-75]. Therefore, the magnetic field can be used to control the microstructure of the coatings and improve the related properties. At present, the research on the solid solution treatment of electrodeposited coating under magnetic fields is very few, and the systematic theory is not formed and further research is needed[76-77].

(5) Furthermore, the current research on electrochemistry under magnetic fields is mainly concentrated below 1T magnetic field intensity, but the research on the influence of the high magnetic field, especially above 10 T, on the electrochemistry process is very few[78]. The high magnetic field can influence the paramagnetic or diamagnetic matrix materials and bath ions significantly, and the mechanism of action on the properties of deposition layer needs to be explored.

5. SUMMARY

In the new situation of high efficiency, clean, energy saving and environmental protection, the traditional technologies have high pollution and high energy consumption processes that need to be improved. Current research work shows that the external magnetic field can have a significant impact on the mass transfer, deposition speed, cathode current efficiency, morphology, structure and physicochemical properties of the deposition layer, and can obtain a series of materials with excellent performance, which will have a very broad application prospect in industry. Therefore, it is of great significance to strengthen the research of magnetoelectrochemistry.

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