Segregation behavior of magnetic ions in continuous flowing solution under gradient magnetic field*

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The research of magnetic separation starts from magnetic solid particles to nanoparticles, and in the research progress, particles become smaller gradually with the development of application of magnetic separation technology. Nevertheless, little experimental study of magnetic separation of molecules and ions under continuous flowing conditions has been reported. In this work, we designed a magnetic device and a "layered" flow channel to study the magnetic separation at the ionic level in continuous flowing solution. A segregation model was built to discuss the segregation behavior as well as the factors that may affect the separation. The magnetic force was proved to be the driving force which plays an indispensable role leading to the segregation and separation. The flow velocity has an effect on the segregation behavior of magnetic ions, which determines the separation result. On the other hand, the optimum flow velocity which makes maximum separation is related to the initial concentration of solution.

Keywords: magnetic ions, magnetic separation, segregation, enrichment

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1. Introduction

Magnetic separation technology has been a hot area of research with its advantages of low cost, low energy consumption, high efficiency, ease of use, and so on.[1,2] In recent years, magnetic separation has made great progress and has a broad application prospect in many fields. It can be used to separate target samples mixed with magnetic nanoparticles, as the pretreatment of many analytes such as biological macromolecules, cells, antibodies, organic compounds, and so on.[3–6] The high gradient magnetic separation technology is proved to be useful in the removal of magnetic particles and lubricant pollution in sticky oil.[7] Magnetic separation technology can be applied to the environment when using magnetic nanoparticles as carriers to remove heavy metal ions or organic dyes in aqueous solutions.[8–10] Besides, by studying the feasibility of removing algae in fish ponds and the separation of binary mixtures of sorbitol and sucrose, it is proved that magnetic separation also has applications in ecology and food processing.[11,12]

In recent years, the research starts from the magnetic solid particles[13] to the magnetic nanoparticles,[14–18] and in the research progress, particles become smaller gradually with the improvement of research and applications. Nevertheless, few experimental studies of magnetic separation of molecules and ions under continuous flowing conditions have been reported.[19–24] Currently, the research objects of magnetic separation are mainly superparamagnetic nanoparticles represented by Fe_3O_4. These magnetic particles usually have large magnetic susceptibility and size, which makes the magnetic force large enough under the gradient magnetic field of thousands of Gausses (G). Compared with magnetic particles, the magnetic moment of molecules and ions is much smaller (for example, the magnetic susceptibility of a ferric chloride molecule is about 12 orders of magnitude smaller than that of a Fe_3O_4 particle). Thus, the magnetic force of molecules or ions is very weak under the same conditions. Besides, molecules or ions are very different from nanoparticles, and there are many obstacles such as a variety of interactions in the separation process. It is very difficult to achieve the magnetic separation of molecules and ions by common methods.

The separation of molecules or ions is needed in many potential applications with the development of science and technology. In the rectification column, for example, magnetic separation technology can be used as an assistant for the separation of liquid oxygen and liquid nitrogen. In the applications of industry, how to improve the recovery rate and reduce the energy consumption of the rectification column has been a common concern. As the temperature and pressure of the rectification column are difficult to control, complete separation is impossible. The residual liquid flowing from the bottom of the rectification column often leads to a waste of energy. However, the target liquid in the residual liquid can be further separated and enriched by using magnetic separation technology. When putting the residual liquid into the rectification column

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for cyclic utilization, the recovery rate will be improved. Correspondingly, the waste of energy is avoided. Moreover, flow behavior is always the key point in the research of magnetic separation technology. Therefore, it is a challenging and important scientific issue to research the magnetic separation of molecules and ions.

In this work, the ferric chloride ($FeCl_3$) solution was used to study the possibility of magnetic separation at the ionic level in a homogeneous mixed solution. $FeCl_3$ molecules are paramagnetic, and the magnetic susceptibility[25] is $\chi_m = 13450 \times 10^{-6}$ cm^3·mol^{-1}. $FeCl_3$ molecules mainly exist in the form of Fe^{3+}, Cl^-, and hydrated ions like $[Fe(H_2O)_6]^{3+}$ in an aqueous solution.[26] The magnetic susceptibility of molecules or ions is much smaller than that of magnetic nanoparticles, thus the magnetic force is very weak in a normal strength gradient magnetic field. Besides, many hindering factors such as diffusion exist in the separation, and the movement and diffusion of ions are greatly influenced by the flow state. Therefore, it is hard to confirm whether the magnetic separation of $FeCl_3$ solution can be achieved. In this research, a magnetic field device with large magnetic field gradient and a "layered" flow channel that can easily achieve laminar flow were designed to study the possibility of the magnetic separation of $FeCl_3$ solution. This study also built a segregation model to discuss the segregation behavior as well as the factors that may affect the magnetic separation, which could provide theoretical and experimental basis for magnetic separation at the molecular and ionic level in homogeneous mixed solution.

2. Experiments

2.1. Experimental setup

The magnetic field device is shown in Fig. 1(a), and it is composed of a permanent magnet array and a magnetic yoke. The magnetic flux density of ordinary magnets is only a few thousands of Gausses (G), thus the magnetic force is very weak when acting on molecules or ions which have tiny magnetic moment. In order to improve the magnetic force, the permanent magnet array and the magnetic yoke are stacked, which increases the magnetic flux density several times, as shown in Fig. 1(b). In the center of the maximum magnetic flux density, the magnetic field can be divided into two equal areas for placing the flow channel. The two areas are completely symmetrical, and the change of magnetic field gradient in the two areas is just the opposite. The magnetic flux density and magnetic field gradient increase gradually from right to left in the flow channel when it is located in area I, and they are larger on the side of outlet 1, with the maximum magnetic flux density about 1.7 T and the maximum magnetic field gradient about 100 T/m. On the contrary, the magnetic flux density and magnetic field gradient are larger on the side of outlet 2 when the flow channel is located in area II.

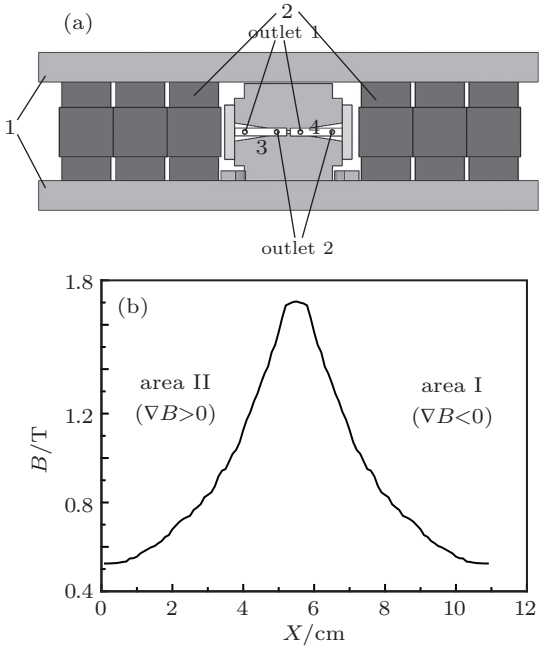


Fig. 1. Schematic diagram of (a) magnetic field device structure and (b) magnetic field distribution. 1: iron plates. 2: permanent magnet array. 3: magnetic yoke. 4: flow channel. X: width of magnetic field. B: magnetic flux density. The magnetic field can be divided into two equal areas for placing the flow channel respectively. The magnetic flux density and magnetic field gradient in the channel are opposite when the channel is placed respectively in areas I and II.

The structure of the flow channel is shown in Fig. 2. The overall space is "layered", and a separate plate is placed at the end of the channel. The "layered" flow channel structure is designed to ensure that the fluid flows in a stable laminar flow. As for the movement, the diffusion and the convection between different components are greatly influenced by the flow state. The height of the channel is about 1 mm. When the magnetic field gradient is different on the two sides of the channel, paramagnetic particles move and become enriched at the side that has larger magnetic field gradient under the effect of magnetic force. As a result, the concentration increases. In this case, the separate plate plays a diversion role, not only to prevent the fluid from being mixed at the exit position, but also to guide the fluid with different concentration flowing into different outlets, which can achieve the separation.

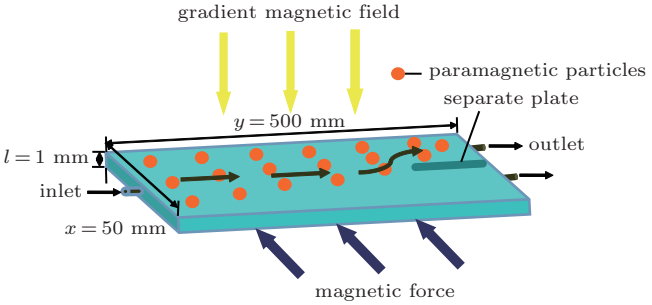


Fig. 2. (color online) Schematic diagram of flow channel structure and magnetic separation. x : channel width, y: channel length, l: channel height.

Besides the magnetic device and the flow channel, a DHL-A constant flow pump with a steady flow device is used to provide power to the flow of the fluid, and a WGD-8A grating spectrometer is used to measure the concentration of the solution.

2.2. Experimental analysis

When $FeCl_3$ solution is flowing in a gradient magnetic field, magnetic force, Lorentz force, Coulomb force, and various kinds of fluid resistance will have an effect, of which magnetic force only acts on Fe(III) ions (to facilitate the presentation, Fe(III) is used as the representative of paramagnetic ions such as Fe^{3+} and its hydrated ions). It has been shown that a large group composed of metal ions and water molecules moves in a magnetic field by magnetic force.[27,28] The magnetic force can be expressed[29,30] as $\boldsymbol{F}_{\rm mag} = \frac{\Delta\chi V_{\rm p}}{2\mu_{\rm o}}\nabla \boldsymbol{B}^2$. Paramagnetic ions move in the direction of larger magnetic field gradient under the effect of magnetic force. However, when $FeCl_3$ solution is flowing continuously in a gradient magnetic field, Lorentz force may also have an effect on the directional movement of ions. Therefore, experimental research will be used to verify whether magnetic force is the major driving force in the magnetic separation of $FeCl_3$ solution.

The flow behavior of the magnetic separation of $FeCl_3$ solution is described in the form of the convection–diffusion equation under the effect of magnetic field

$$\frac{\partial C}{\partial t} + \nabla \cdot \left[\frac{D}{RT}(-\nabla C + CF_{\rm m}) + C\boldsymbol{v}\right] + r = 0. \tag{1}$$

The first term on the left is the change of concentration versus time, the second term is the convection and the diffusion under the effect of magnetic field, and the third term is the change of the chemical reaction. D is the diffusion coefficient, R is the universal gas constant, T is the thermodynamic temperature, $\boldsymbol{F}_{\rm m}$ is the molar magnetic force, and vector $\boldsymbol{v}$ is the flow velocity. There is no chemical reaction in the magnetic separation of $FeCl_3$ solution, and if only the steady state is discussed, the first and the third terms are both zero. Then the equation can be written as

$$\nabla \cdot \left[\frac{D}{RT}(-\nabla C + CF_{\rm m}) + C\boldsymbol{v}\right] = 0. \tag{2}$$

It is not hard to see that the magnetic force, the flow velocity and the concentration are the factors that influence the magnetic separation of $FeCl_3$ solution. The influence of the magnetic force is obvious, the greater the magnetic force is, the better the separation will be. But it is difficult to read directly from the equation how the flow velocity and concentration affects the separation, therefore, experimental research is needed.

2.3. Experimental methods

It is difficult to observe and measure the movement of Fe(III) ions directly in the magnetic separation at the ionic level. Therefore, the concentration and concentration difference were measured as a breach to study the separation.

The $FeCl_3$ solutions with mass fraction about 13% were used, and the inlet flow rate was 2 ml/min. Then the concentrations in the two outlets were measured and compared with each other when the flow channel was placed in area I and area II, respectively. This was used to verify whether the magnetic force is the major driving force in the magnetic separation of $FeCl_3$ solution. When the flow channel is located in area I and area II, respectively, the directions of the magnetic force of Fe(III) ions are just the opposite, but the directions of the Lorentz force are the same. Therefore, if the magnetic force is the major driving force, the results of the concentration in the two outlets are certainly the opposite when the flow channel is located in area I and area II, respectively.

Then the flow channel was placed in area I, and $FeCl_3$ solutions of 15%, 17%, and 20% (mass fraction) were used, respectively. The concentration difference between the two outlets was measured when the flow rate was 1 ml/min, 2 ml/min, 3 ml/min, 4 ml/min, 6 ml/min, and 8 ml/min, respectively (the corresponding flow velocity is about 0.33 mm/s, 0.67 mm/s, 1.0 mm/s, 1.3 mm/s, 2.0 mm/s, and 2.7 mm/s, respectively). The influence of the flow velocity and the concentration on the magnetic separation of $FeCl_3$ solution were studied by the change of the outlet concentration difference versus the flow velocity and the concentration.

3. Results and discussion

The concentrations in the two outlets when the flow channel is located in area I and area II respectively are shown in Fig. 3. The initial concentrations of $FeCl_3$ solutions are about 13% and the flow rate is 2 ml/min (the flow velocity is about 0.67 mm/s). When the channel is located in area II where the magnetic field gradient is larger on the side of outlet 2, the concentrations in outlet 1 are less than that in outlet 2. When the channel is located in area I where the magnetic field gradient is larger on the side of outlet 1, the concentrations in outlet 1 are greater than that in outlet 2. It can be seen that the concentration is greater in the area with larger magnetic field gradient, which indicates that the direction of the movement and enrichment of Fe(III) ions is consistent with that of the magnetic force. The results of the concentrations in the two outlets are just the opposite when the flow channel is located in area I and area II, respectively. However, the concentration difference is almost the same. The opposite results of concentration and the same concentration difference indicate that the action of the Lorentz force can be ignored, the magnetic force

is the major driving force in the magnetic separation of $FeCl_3$ solution.

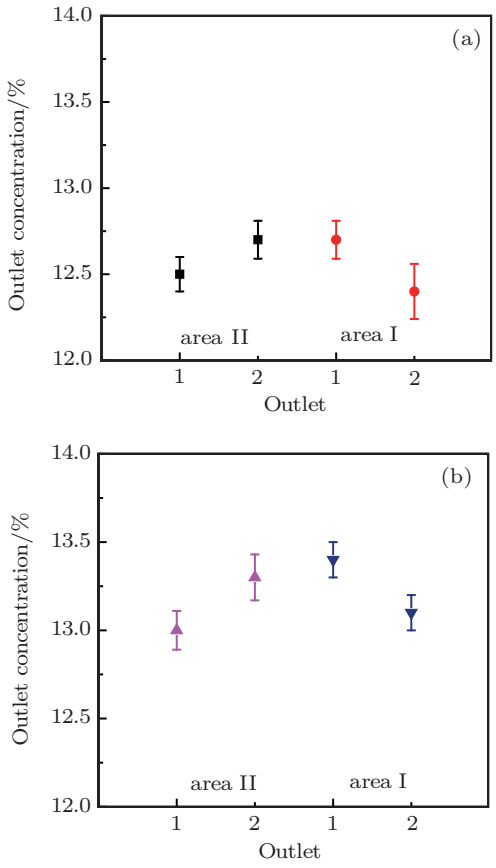


Fig. 3. (color online) The concentrations in the two outlets when the flow channel is located in different areas, with the initial concentration of the solution of (a) 12.6% and (b) 13.2%.

The results of the concentration difference between the two outlets versus different flow velocity when the flow channel is located in area I are shown in Fig. 4, where the mass fraction of $FeCl_3$ solution is respectively 15%, 17%, 20%. It can be found that the concentration difference between the two outlets increases first and then decreases with the increase of the flow velocity. The concentration difference is the result of Fe(III) ions' movement and enrichment to the area with larger magnetic field gradient under the effect of the magnetic force, as shown in Fig. 5. The flow channel can be divided into two equal parts, area 1 and area 2. Assume that the number of Fe(III) ions in each of the two equal areas at the initial state is N_0, and the number of Fe(III) ions which can be enriched in the solution is N. Then N_0+N and N_0-N is respectively the number of Fe(III) ions in area 1 and area 2 at the steady state. Assuming that t_0 is the time which is needed for achieving the maximum enrichment (when Fe(III) ions move from one side to another side of the channel, t_0 is maximum), t_0 does not change with the flow velocity because there is no transverse flow in the solution and the magnetic force of the Fe(III) ions is independent of the flow velocity. Thus, there is a critical flow velocity v_0. When $v \le v_0$, all the Fe(III) ions that can be enriched will move and accumulate in area 1, and in this case $N = N_{\max}$ is unchanged. When the flow velocity is greater than v_0, there is not enough time for some Fe(III) ions to move to area 1. Therefore, N will be decreased with the increase in the flow velocity.

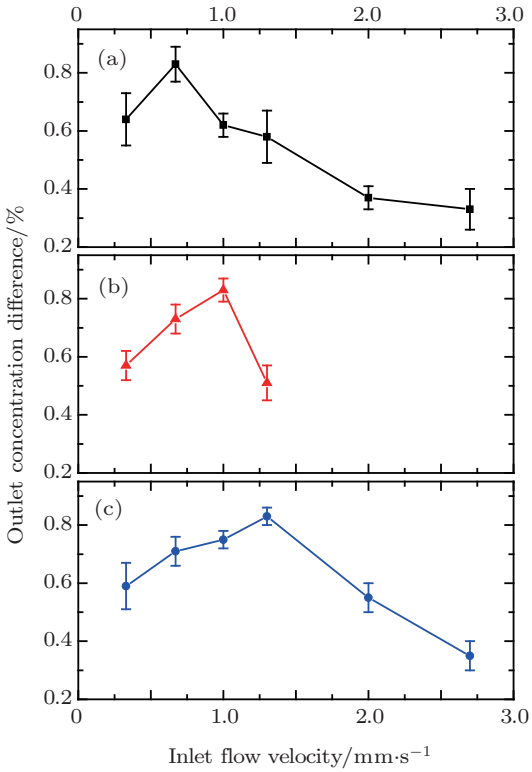


Fig. 4. (color online) The separation effect with different inlet flow velocities. (a) 15%, (b) 17%, (c) 20%.

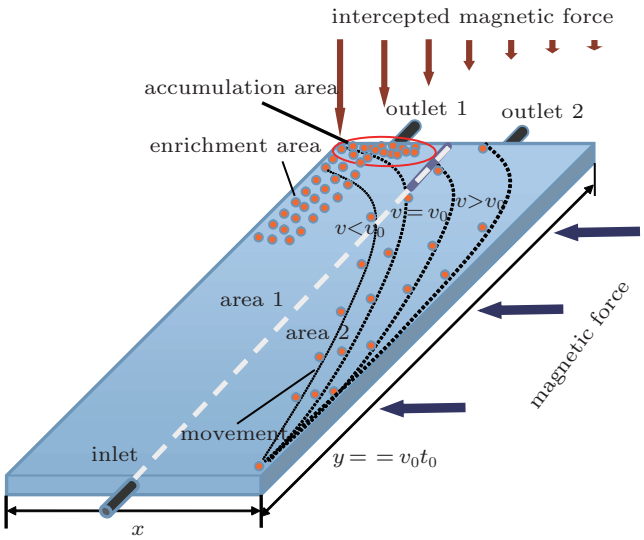


Fig. 5. (color online) Segregation of Fe(III) ions in the flow channel. Fe(III) ions move from area 2 to area 1 under the effect of magnetic force. There is a critical flow velocity v_0. When $v \le v_0$, all the Fe(III) ions that can be enriched will move and accumulate in area 1. When $v > v_0$, the number of segregated Fe(III) ions will decrease.

The enrichment of Fe(III) ions leads to the increase of the chemical potential. The chemical potential can be

expressed[31] as $u = \frac{\partial G}{\partial n}$, which is the change of Gibbs free energy when the change of the amount of components is 1 mol. Since each of the components in the solution is Fe(III) ions, the chemical potential can be written as

$$u' = \frac{u}{N_A} = \frac{\partial G}{\partial N}, \tag{3}$$

where N_A is the Avogadro constant and N is the number of the Fe(III) ions that can be enriched. In this case, the chemical potential means the average free energy of each Fe(III) ions because of the enrichment. Then the total chemical potential of the enrichment area can be expressed as

$$u^* = Nu'. \tag{4}$$

Due to the change of the chemical potential, Fe(III) ions are driven by the chemical potential driving force from the high chemical potential area to the low one. The chemical potential driving force is opposite to the magnetic force. Therefore, Fe(III) ions are bound to be balanced by the chemical potential and magnetic force at the steady state. Regardless of the susceptibility of Fe(III) ions, the magnetic force is determined by the gradient magnetic field. The gradient magnetic field is unchanged, thus the maximum magnetic force of the Fe(III) ions in the enrichment area will not change. Correspondingly, the maximum change of the chemical potential in the enrichment area will be constant. According to Eq. (4), N_{max}, the maximum value of N, is constant.

The magnetic field gradient is far greater at the exit position than that in the channel because of the sudden disappearance of the magnetic field at the exit of the channel. The Fe(III) ions are intercepted by greater magnetic force at the exit position, and only a few Fe(III) ions can flow out. Therefore, a certain accumulation of Fe(III) ions is formed (accumulation area in Fig. 5), which makes the concentration distribution very different from that in the channel.

The volume of the two equal areas in the flow channel is expressed by V_0, then the concentration of Fe(III) ions in area 1 and area 2 can be expressed respectively as

$$C_1 = \frac{N_0 + N}{N_A V_0}, \quad C_2 = \frac{N_0 - N}{N_A V_0}. \tag{5}$$

Assuming that the number of the accumulated Fe(III) ions in the accumulation area is ΔN (the intercepted magnetic force in outlet 1 is far greater than that in outlet 2, in order to discuss conveniently, the intercepted magnetic force in outlet 2 is not considered), then the concentration of Fe(III) ions in the accumulation area at the exit position can be expressed as

$$C_A = \frac{N_0 + N}{N_A V_0} + \frac{\Delta N}{N_A V_A}, \tag{6}$$

where V_A is the volume of the accumulation area, and it is also a constant. Thus, the concentration difference between the two outlets can be expressed as

$$\Delta C = \frac{2N}{N_A V_0} - \frac{\Delta N}{N_A V_A}. \tag{7}$$

According to Eq. (2), we have the following relationship between the concentration of Fe(III) ions in accumulation area and the flow velocity (detailed derivation process is shown in Appendix A):

$$C_A \propto v^{-1}. \tag{8}$$

Obviously, the concentration of Fe(III) ions in accumulation area at the exit position decreases with the increase of the flow velocity. It is shown that the number of the accumulated Fe(III) ions in the accumulation area, ΔN, decreases when increasing the flow velocity. Therefore, there must be a flow velocity v_A which makes $\Delta N = 0$.

When Fe(III) ions are accumulated in the accumulation area, the viscous resistance pointing to the outlet will have an effect. The accumulation of Fe(III) ions also leads to the change of the chemical potential, which causes Fe(III) ions to be affected by the chemical potential driving force pointing to the outlet. At the steady state, the accumulated Fe(III) ions are balanced by the intercepted magnetic force, the chemical potential driving force and the viscous resistance at the exit position

$$F = F_u + f, \tag{9}$$

where F is the intercepted magnetic force, F_u is the chemical potential driving force, and f is the viscous resistance. The intercepted magnetic force F is constant without the change of the gradient magnetic field and the susceptibility of Fe(III) ions. The chemical potential driving force F_u is dependent on the number of the accumulated Fe(III) ions ΔN. The viscous resistance can be simply expressed as $f = \lambda v$, where λ is a constant related to the viscosity coefficient, etc. In this case, v is the flow velocity of the solution.

According to Eq. (9), the chemical potential driving force can be written as

$$F_u = F - \lambda v. \tag{10}$$

It is also shown that the number of the accumulated Fe(III) ions in the accumulation area, ΔN, decreases when increasing the flow velocity, because both F and λ are positive constants. At the critical state in which all the accumulated Fe(III) ions can flow out of the channel ($\Delta N = 0$ and $v = v_A$), the chemical potential driving force $F_u = 0$. Thus there is the following relationship:

$$F = \lambda v_A. \tag{11}$$

It indicates that the flow velocity which can make all the accumulated Fe(III) ions flow out of the channel, v_A, is constant.

According to Eq. (7), the concentration difference between the two outlets is determined by N and ΔN, which are respectively the number of Fe(III) ions that can be enriched in the solution and the number of the accumulated Fe(III) ions in the accumulation area. The change of N and ΔN versus the flow velocity is shown in Fig. 6. When $v \leq v_0$, $N = N_{\max}$ is unchanged, and ΔN is decreased when increasing the flow velocity. Thus the concentration difference between the two outlets is increased with the increase of the flow velocity. When $v \geq v_A$, N is decreased when increasing the flow velocity and $\Delta N = 0$, thus the concentration difference decreases. When $v_0 < v < v_A$, there must be a flow velocity which makes the concentration difference maximum. As a consequence, the concentration difference between the two outlets increases first and then decreases with the increase of flow velocity, and there is an optimum flow velocity which makes the maximum separation.

It has been shown that the flow velocity v_A which makes $\Delta N = 0$ is constant, and the change of ΔN with the flow velocity v is determined by the constants F and λ. Therefore, the optimum flow velocity which makes the maximum separation be dependent on the critical flow velocity v_0, according to Fig. 6.

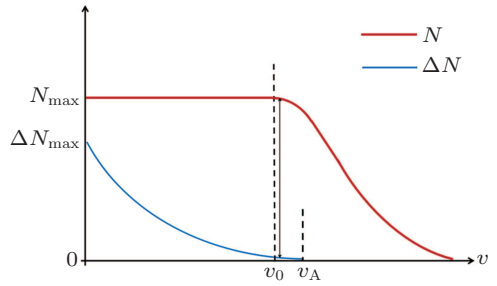


Fig. 6. (color online) The change of N and ΔN versus the flow velocity.

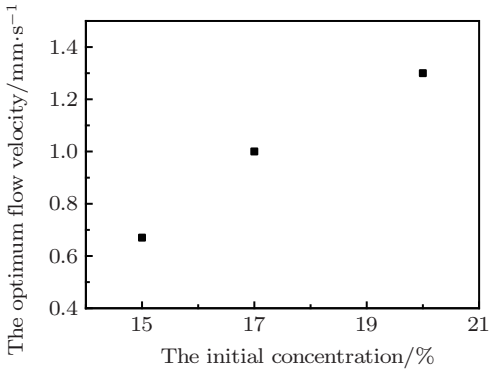


Fig. 7. The optimum flow velocity of different concentration of the solution.

The optimum flow velocity of 15%, 17%, and 20% $FeCl_3$ solution are shown in Fig. 7. It can be found that the optimum flow velocity is increased with the increase of the initial concentration of the solution. When the initial concentration of the solution is increased, the initial number of Fe(III) ions increases. Correspondingly, there are more Fe(III) ions surrounding the enrichment area. These Fe(III) ions always have greater magnetic force compared with the Fe(III) ions farther away from the enrichment area. Therefore, the Fe(III) ions surrounding the enrichment area will move and accumulate to the enrichment area faster. However, $N_{\max}$, the maximum number of Fe(III) ions that can be enriched is unchanged. As a result, the enrichment area can reach saturation more quickly. It indicates that when increasing the initial concentration of the solution, the time which is needed for achieving the maximum enrichment, t_0, is decreased. Correspondingly, the critical flow velocity v_0, which can achieve the maximum enrichment in the channel, is increased.

It has been shown that the optimum flow velocity is dependent on the critical flow velocity v_0. When increasing the initial concentration of the solution, v_0 is increased. Therefore, the optimum flow velocity is also increased.

4. Conclusion

The magnetic separation of magnetic ions has been achieved by the magnetic field device with larger magnetic field and gradient as well as the flow channel that can easily achieve laminar flow. The magnetic force is the major driving force, which plays an indispensable role, leading to the segregation of magnetic ions. The maximum number of the segregated ions is determined by the joint action of the chemical potential and magnetic force. The flow velocity also has an effect on the segregation behavior. When the flow velocity increases to a certain degree, the number of the segregated ions in the channel will start to reduce. As a consequence of the interception at the exit position of the channel, segregated ions accumulate. The number of the accumulated ions will be decreased when the flow velocity increases. As a result, the separation effect increases first and then decreases with the increase of the flow velocity. There is an optimum flow velocity which makes maximum separation. In this experiment, the optimum flow velocity is related to the initial concentration of solution, the higher the concentration, the larger the optimum flow velocity.

This study has some guiding significance for the magnetic separation of liquid oxygen and liquid nitrogen. It indicates that magnetic force plays a decisive role in the maximum separation effect when the flow velocity is optimum. Improving the performance of the magnetic device, such as using superconducting magnets, can greatly improve the magnetic force. Therefore, the separation effect will be improved to a certain extent. When using magnetic separation technology as an assistant for the separation of liquid oxygen and liquid nitrogen in the rectification column, the liquid oxygen in the residual liquid will be enriched greatly. As a consequence, the recovery

rate will be improved when putting the residual liquid into the rectification column for cyclic utilization. Correspondingly, the energy is also saved. Besides, the equipment of magnetic separation technology is reusable, and no more energy consumption exists. Therefore, magnetic separation technology has great potential application in the separation of liquid oxygen and liquid nitrogen in the rectification column.

Appendix A

The convection–diffusion equation in the steady state

$$\nabla \cdot \left[\frac{D}{RT} (-\nabla C + C\boldsymbol{F}_{\mathrm{m}}) + C\boldsymbol{v} \right] = 0 \tag{A1}$$

is studied. It can be written in the form of components, and the vertical direction is not considered, then

$$\begin{aligned} &\frac{D}{RT}F_x\frac{\partial C}{\partial x} + \frac{D}{RT}F_y\frac{\partial C}{\partial y} + \frac{D}{RT}C\frac{\partial F_x}{\partial x} + \frac{D}{RT}C\frac{\partial F_y}{\partial y} \\ &+ v_x\frac{\partial C}{\partial x} + v_y\frac{\partial C}{\partial y} + C\frac{\partial v_x}{\partial x} + C\frac{\partial v_y}{\partial y} \\ &= \frac{D}{RT}\left(\frac{\partial^2 C}{\partial x^2} + \frac{\partial^2 C}{\partial y^2}\right). \end{aligned} \tag{A2}$$

At the exit position, the concentration distribution in the accumulation area is very different from that in the flow channel because of the accumulation of the magnetic ions. In this case, the intercepted magnetic force and the flow velocity in y direction are the main effects. The influence of diffusion is very weak, thus the diffusion term can be ignored. Assuming that the concentration is unchanged in x direction in the accumulation area, the equation can be simplified as

$$\begin{aligned} &\frac{D}{RT}F_y\frac{\partial C}{\partial y} + \frac{D}{RT}C\frac{\partial F_x}{\partial x} + \frac{D}{RT}C\frac{\partial F_y}{\partial y} \\ &+ v_y\frac{\partial C}{\partial y} + C\frac{\partial v_x}{\partial x} + C\frac{\partial v_y}{\partial y} = 0. \end{aligned} \tag{A3}$$

The magnetic forces for driving in x direction and for intercepting in y direction are both unchanged when the area for study is certain. Then equation (A3) can be written as

$$\begin{aligned} &\frac{D}{RT}F_y\frac{\partial C}{\partial y} + \frac{D}{RT}CK_1 + \frac{D}{RT}CK_2 \\ &+ C\frac{\partial v_x}{\partial x} + v_y\frac{\partial C}{\partial y} + C\frac{\partial v_y}{\partial y} = 0, \end{aligned} \tag{A4}$$

where $\partial F_x/\partial x = K_1$, $\partial F_y/\partial y = K_2$, K_1, and K_2 are both constants.

The flow velocity will be increased suddenly when the solution flows into the outlet, because the size of the outlet is much smaller than that of the flow channel. Both the sizes of the channel and outlet are constant, therefore, the ratio of the change of flow velocity at the exit position is also constant. Then the gradient of flow velocity in y direction can be written as $\partial V_y/\partial y = \Delta v_y/\Delta y = Kv_y$, where K is a constant related to the size of the channel and the outlet. Compared with the sudden increase of the flow velocity in the y direction, the influence of convection in the x direction can be almost ignored. Then equation (A4) can be written as

$$\frac{D}{RT}F_y\frac{\partial C}{\partial y} + \frac{D}{RT}CK_1 + \frac{D}{RT}CK_2 + v_y\frac{\partial C}{\partial y} + Kv_yC = 0. \tag{A5}$$

The relationship between the concentration and the position can be converted into that between the concentration and the flow velocity,

$$\frac{\partial C}{\partial y} = \frac{\partial C}{\partial v_y}\cdot\frac{\partial v_y}{\partial y} = Kv_y\frac{\partial C}{\partial v_y},$$

then equation (A5) can be written as

$$Kv_y\left(\frac{D}{RT}F_y + v_y\right)\frac{\partial C}{\partial v_y} + \left[\frac{D}{RT}(K_1+K_2) + Kv_y\right]C = 0. \tag{A6}$$

The order of magnitude of diffusion coefficient D is about 10^{-9}, while the order of magnitude of the flow velocity is about 10^{-4}. Compared with the flow velocity, D is very small and can be ignored. Thus equation (A6) can be written as

$$v_y\frac{\partial C}{\partial v_y} + C = 0. \tag{A7}$$

The solution of equation (A7) is

$$C = v_y^{-1}. \tag{A8}$$

Considering the actual situation, equation (A8) can be expressed as

$$C_{\mathrm{A}} \propto v^{-1}, \tag{A9}$$

where C_{A} is the concentration of the accumulated Fe(III) ions in the accumulation area, and v is the flow velocity in y direction.

Obviously, the concentration is decreased with the increase of the flow velocity in the accumulation area at the exit position where the Fe(III) ions are accumulated.

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