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Effects and Mechanization of a High Gradient Magnetic Separation Process for Particulate and Microbe Removal from Ballast Water

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Abstract**—**As a pretreatment process of ballast water treatment, the performance of high gradient magnetic separation (HGMS) technology for the removal of particulates and microorganisms was studied. The results showed that HGMS process could effectively remove suspended particles larger than 5 μm and had ability to resist impact load. Microorganism could also be effectively removed by HGMS process, and the removal effect increased with increasing magnetic field strength. The maximum removal rates for *Escherichia coli* (*E. coli*) and *Staphylococcus aureus* (*S. aureus*) were 4016.1% and 9675.3% higher, respectively, than without the magnetic field. In addition, the superoxide dismutase (SOD) activity of the microbes decreased by 32.2% when the magnetic field strength was 15.4 mT for 72 min. The microstructure of the stainless steel wool was investigated, and the results showed that particle removal by HGMS has common function by the magnetic force of the high-strength, high-gradient magnetic field on weakly magnetic particles in the water, and on the stainless steel wool.

Keywords**—**HGMS, particulates, superoxide dismutase activity, steel wool magnetic medium.

I. INTRODUCTION

WITH the rapid development of industry and shipping, the invasion of marine organisms into the ballasts of ships has brought serious threats to the marine environment, the health of marine ecosystems, and sustainable development [1], [2]. A combined technology using a HGMS process and UV radiation (Patent number: ZL200910073103.X) was studied for ballast water treatment [3]. In this process, the HGMS equipment is placed at the output port of the ballast pump and is used to remove plankton and suspended particles larger than 50 μm. Subsequently, UV radiation is used to inactivate the remaining microorganisms. Compared with the other ballast water treatment methods, such as the PureBallast system developed by Alfa-Laval and Wallenius Water AB [4] and the Greenship Sedinox system developed by the Greenship Company [5], this system could meet numerical standards, enable high-speed treatment, and eliminate concerns regarding marine pollution from residual chemicals. Moreover, this process can greatly reduce the amount of sludge and dead organisms that accumulate on ships [6], [7].

HGMS is a type of physical method; using a high-background magnetic field, a magnetic gradient produces a magnetic force with a magnitude sufficient to weakly attract magnetic particles of almost colloidal dimensions to the surface of a matrix element and thus separate them from the nonmagnetic particles [8], [9]. HGMS techniques, both direct and indirect (also called magnetic seeding), have been applied to: purify kaolin clay, desulfurize coal, process rare metals ores, treat water polluted with heavy metal ions [10], organic substances and microorganisms [11], treat water from conventional and nuclear power plants [12], treat urban waste water; and purify industrial gas.

Previous studies have focused on investigating or separating ferrous contaminants from oil [10]-[13] or evaluating the removal of magnetic solids from fluid flows [12]-[14]. The performance of HGMS for microbe removal in ballast water treatment, which would have a significant impact on the subsequent UV irradiation, has not yet been investigated and still needs to be verified. When the HGMS technique is used to treat ballast water, it can facilitate physical entrapment and magnetic separation and also can have a magnetic biological effect. Some studies have shown that the magnetic field has a magnetic biological effect, and the magnetic field can affect the enzyme activity, genetic material, and cell membrane of microbes, which can eradicate the microorganism if the magnetic field has sufficient strength [15], [16]. In this paper, a continuous-flow experiment using HGMS for ballast water treatment was conducted, in which the efficiency of the removal of particulates and microorganisms with a high-gradient magnetic filter was studied, and the mechanism of the high-gradient magnetic filtration process was also discussed by testing the changing of the microstructure of the stainless steel wool.

II. MATERIALS AND METHODS

A. Experimental Water Sample

The artificial seawater used in this experiment was prepared according to the International Convention for the Control and Management of Ships' Ballast Water and Sediments. The turbidity of natural water was simulated using kaolin, due to the high buoyancy in seawater turbidity, and was based on average water quality; the turbidity was adjusted to 20~250 NTU.

E. coli and *S. aureus* were used as reference bacteria because

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they are commonly used as indicators in disinfection studies. Moreover, both are frequently detected in surface water, wastewater, hospitals, and even in soil and they are easily cultivable. The inactivation of the bacteria was assumed to proceed by the same mechanism as all other less resistant microorganisms. The cells were grown at 37 °C under aeration in LB broth. The physical and chemical indicators of the water were based on the average quality of Chinese offshore water. Specifically, the pH was 7.5~8.0, the average salinity of the artificial seawater was 35 PSU (the ratio of sodium chloride, magnesium chloride and potassium chloride was 3:2:1), and the turbidity, which was adjusted with diatomaceous earth and artificial seawater, was approximately 70 mg/L.

B. The Design of the Experimental Device

The experimental apparatus shown in Fig. 1 indicates the inductance coil, magnetic yoke, magnetic medium, and reaction zone, which are the primary components of the experimental device. The DC inductance coil produced a single-direction magnetic field due to the relative magneto-conductivity of the magnetic medium. The magnetic field was filled in the reaction zone and was far greater than that of water. The magnetic flux lines appeared on the corresponding bend in space and were more concentrated near the steel wool than near the space without the steel wool. As a result, the magnetic flux density was very high near the steel wool, and the reaction zone had a high-gradient magnetic field.

The main parameters of the process included are: a flow rate set from 50 to 100 L/h and a DC threshold-stabilized source (220 V AC to DC) of 7 A and 15 V. The water was up-welled and ran parallel to the direction of the magnetic field. Fe_3O_4 and stainless steel wire were chosen as the magnetic seed and magnetic medium, respectively. The HGMS equipment used enamel wire, which is pliable. The diameter, length, turns, and operating current of the enamel wire were 1.50 mm, 25 cm, 1000, and 3 A, respectively. There was no need to cool the equipment because the magnetic field intensity was only 0.0432 T.

The coil was approximated as a long, straight solenoid with a well-distributed magnetic field within, and the magnetic field intensity measured at the center line was the same as that of the device. Changing the voltage and current of the DC power can control the magnetic field of the inductance coil; when the current is 0~8 A, the magnetic induction intensity is 0~20 mT.

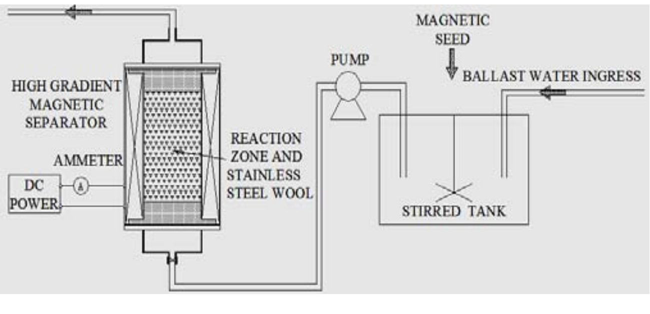


Fig. 1 The experimental device diagram

The measurement of turbidity in this experiment was completed by using an LP2000 desktop microcomputer nephelometer produced by the Hanna technology service center (Beijing). SOD can remove free radicals, protect damaged cells, and balance the oxidation and antioxidant substances in the body. The experiment used the xanthine oxidase method to measure the activity of SOD, and the experimental reagents were from Nanjing Jiancheng Biological Engineering Institute.

C. Nomenclature

- N_0 concentration of *S. aureus* prior to treatment (CFU/100 mL).
- N_t concentration of microorganisms after treating for time t (CFU/100 mL).
- log N_0/N_t concentration of microorganisms on a log 10 scale.

III. RESULTS AND DISCUSSION

A. Magnetic Field Intensity and Particulate Removal

Particles, especially of larger size, are one of the important factors in UV radiation and can reduce the UV radiation dose on the microorganism by shielding, inclusion, and scattering [17], [18]. As a pretreatment process, the performance of HGMS for particle removal was studied under magnetic intensities of 0 mT, 5.1 mT, 10.3 mT, 15.4 mT, and 21.0 mT when the flow rate was 20 L/h, and the steel wool filling rate was 6%. The residual turbidity is shown in Fig. 2.

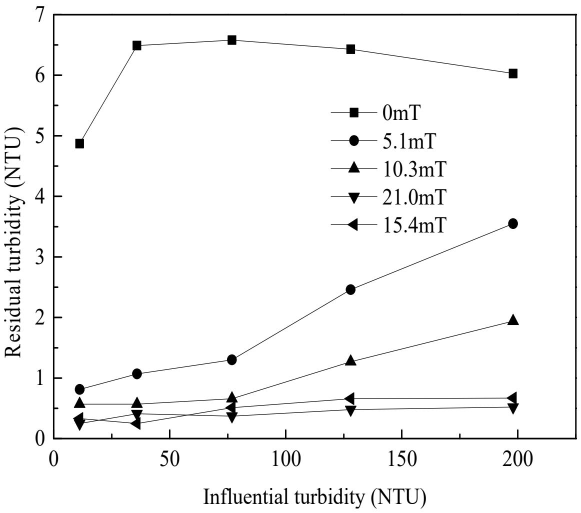


Fig. 2 Effect of magnetic field strength on HGMS for turbidity removal

We can see from Fig. 2 that the HGMS process showed good performance for particle removal, even when the magnetic field strength was 0 mT. The turbidity of the effluent was maintained at between 5 NTU and 7 NTU in the HGMS process with a magnetic field strength of zero when the in-fluent water turbidity changed from 11.2 NTU to 198 NTU; under these conditions, the particles were removed in the HGMS process through the physical interception, filtration, and adsorption to the steel. Tang et al. [19] used steel wool medium for direct filtration and showed that the steel wool had a high treatment capacity under high speed and continuous operation. However, this result was not sufficient for the subsequent UV irritation [20].

The magnetic induction intensity was changed from 0 to 21 mT by varying the voltage and current of the DC power, and the effluent water characteristics showed that with the same water quality, the turbidity of the effluent decreased by increasing magnetic field strength in the HGMS device, especially at high influent turbidity. For example, when the influent turbidity was 198 NTU, the residual turbidity was 6.03 NTU，3.55 NTU，1.94 NTU，0.52 NTU， and 0.67 NTU when the magnetic field was 0 mT, 5.1 mT, 10.3 mT, 15.4 mT, and 21.0 mT, respectively. On the other hand, the residual turbidity was always below 1 NTU and had a limited relationship with the magnetic field strength when the magnetic intensity was greater than 15.4 mT. Lower effluent turbidity is compatible with the requirements of the subsequent ultraviolet treatment and also showed that the HGMS device has the ability to resist the impact load for turbidity removal. Since increasing the magnetic field intensity required an increase in the current and thus higher energy consumption, a magnetic field strength of 15.4 mT was selected for the subsequent tests because the effluent turbidity was similar when the magnetic field strength was 21.0 mT. Research at the ChangZhou Drinking Water Treatment Plant showed that when the quantity of magnetic particles was 200-300 mg/L, the dosage of the coagulant was 5-12 mg/L, and the magnetic field strength was 0.2-0.4 T, and the turbidity of river water with turbidity of 100-150 NTU could be purified to within 5 NTU, with a removal rate of 98.5%, which was within the national standards for the quality of drinking water after purification [21].

Large-sized particles can protect microbes from UV irradiation by shadowing, parcels, and the scattering effect [22]. As a result, a particle size of greater than 5 μm was selected as the standard for the HGMS process in accordance with the previous works [23], [24]. The effect of the HGMS process with different magnetic field strengths on the removal of particles with diameters greater than 5 μm is shown in Fig. 3.

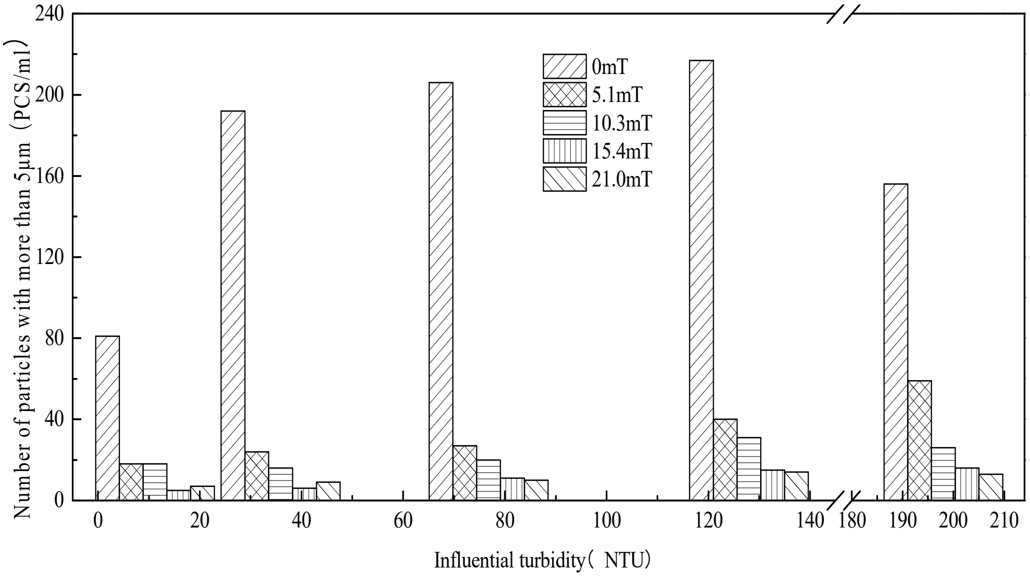


Fig. 3 Effect of HGMS with different magnetic field strengths on the removal of particles with diameters greater than 5 μm

When the magnetic field strength was 0 mT, there was no magnetic field in the reaction zone. The device completely depended on the physical retention, filtration, and adsorption of the steel wool to remove the particles, and the number of particles with diameters greater than 5 μm was more than 80 PCS/mL. After the reaction zone was magnified, the number of 5μm particles decreased with increasing magnetic field intensity in the HGMS device. When the magnetic field intensity reached more than 15.4 mT, under different influent turbidity levels, the number of particles with diameters greater than 5 μm remained less than 18 PCS/mL, and the effect of particle protection on the UV sterilization was negligible [25]. At the initial turbidity of 128 NTU, the number of particles with diameters greater than 5 μm in the effluent of the 15.4 mT magnetic field decreased by 91.7%, 55%, and 41.9%, relative to the numbers at 0 mT, 5.1 mT, and 10.3 mT, respectively. The experimental results showed that the HGMS device can effectively remove the suspended particles larger than 5 μm from water with and has the ability to resist the impact load.

B. Effect of HGMS on Microorganism Removal

Two types of microorganism, *E. coli* and *S. aureus*, were used as reference bacteria, and the performance of the HGMS for bacteria removal at different magnetic field strengths was conducted with a flow speed of 20 L/h, an initial turbidity of 71 NTU, and a filling rate of 6%. The experimental results are shown in Fig. 4.

The HGMS process could effectively remove *E. coli* and *S. aureus*, with the maximum removal rates of 0.928 and 0.832, respectively, when the magnetic field was 21 mT, which were 4016.1% and 9675.3% higher than those without the magnetic field. On the whole, the microorganism removal rate increased with increasing in the magnetic field intensity, and the removal rate of the high-gradient magnetic field on *E. coli* was slightly higher than that on *S. aureus*. This result may be related to the flocculation characteristics of the cell surface [26].

The magnetic field has the effect of sterilization. When the magnetic field strength is 4000 Gs, the sterilization rate can reach more than 20% [27], [28]. Many microorganisms in water, such as *E. coli*, which is the most common, can be adsorbed onto the iron oxide or other magnetic particles and are effectively removed by the HGMS device [29]. On the other hand, the magnetic force directly affected the cell cytosol, enzymes and other aspects of the bacteria, causing intracellular enzymes to be inactivated [30], [31].

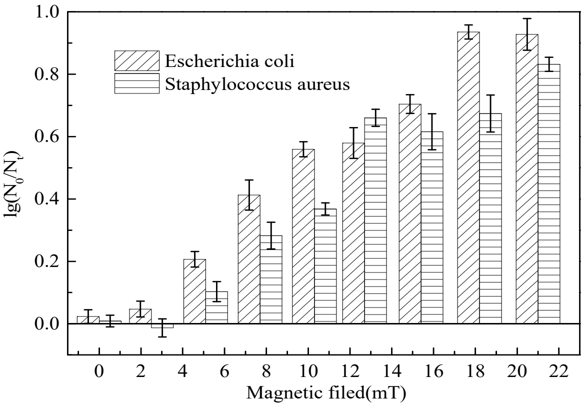


Fig. 4 Effect of HGMS with different magnetic field strengths on microbe removal

C. HGMS Process on the Intracellular Bacterial Enzyme Activity

Under different flow rates, the magnetic field strength was maintained at 15.4 mT, the magnetic seed dosage was 1 g/L, and the dosage of the coagulant was 100 mg/L, and the SOD activity of the bacteria influenced by the magnetic field was conducted, and the experimental results are shown in Fig. 5. In Fig. 5, the SOD of microorganisms treated with the high-gradient magnetic field decreased with the residence time increasing. Taking the 72-min effluent as an example, the SOD activity of the bacteria decreased by 32.2%.

Some research has shown that magnetic fields could induce the activity of α-amylase, dehydrogenase, and proteases [32]. Iwasaka et al. suggested that magnetic fields had an effect on the active center of the enzymatic reaction of plasmin with a substrate, especially on the hydrogen bonds of the charge-relay system [33]. Morelli et al. studied the effects of extremely low frequency (ELF) electromagnetic fields (EMFs) on several (seven in total) membrane-associated enzymes [34] and claimed that the activities of three of these (alkaline phosphatase, acetylchlinesterase, and phosphoglycerate kinase) exhibited a decrease in activity (approximately 50%) when exposed to ELF EMFs. The observed effect disappeared when the enzymes were dissolved in a buffer containing the detergent Triton X-100, suggesting that the fields may modify the membrane organization and structure by acting directly on the anisotropy of the diamagnetic susceptibility of the membrane phospholipids. In the HGMS process, the very high magnetic flux density near the steel wool (up to 1000 Gauss/micron), will produce forces, such as the Lorentz force, on nearby microorganisms. The magnetic field can influence some metalloenzymes, the active centers, coenzymes or prosthetic groups of which contain traces of metal ions. The arrangement of these components will be altered by the Lorentz force in a high-gradient magnetic field, so the enzyme activity could also be affected [35], [36].

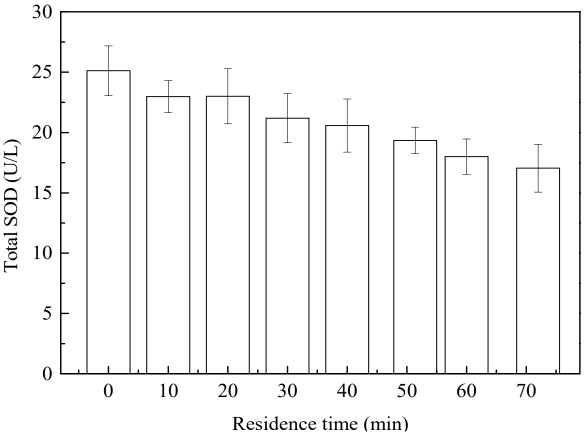
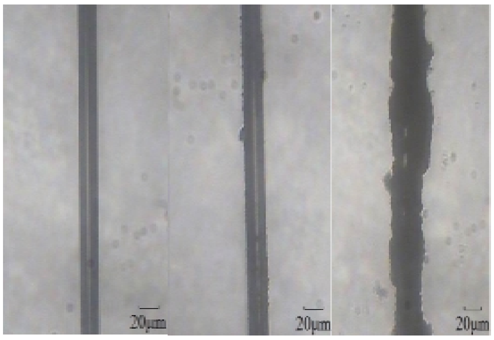


Fig. 5 Total SOD of *S. aureus* after treatment with different residence times

D. Change of Steel Wool Magnetic Medium

The HGMS used a large amount of stainless steel wool as the filter matrix. The magnetic field from the electric current caused the stainless-steel wool to generate a magnetic field. The magnetic force of the steel wool had an effect on the suspended impurities in the liquid that was greater than that of the flow force and of the gravity of the particles themselves. As a result, the suspended impurities were trapped in the steel wool matrix and were subsequently removed, thus achieving purification. To directly analyze the mechanism of particle removal by the magnetic field, the microstructure of the stainless steel wool was photographed under different conditions, and the results are shown in Fig. 6.



(a) (b) (c)

Fig. 6 Changes in the steel wool under different running conditions: (a) steel wool (b) steel wool running at 0 mT (c) steel wool running at 15.4 mT

As shown in Fig. 6, the original steel wool had a smooth surface with no attached particles, and the diameter was 23 μm. Without a current, a small amount of particulates were on the surface of the steel wool, but the surface was mostly smooth, and the particles were completely removed through interception by and adsorption to the steel wool. Because the diameter of stainless steel wool is larger than that of the suspended particles (Φ = 23 μm), and the surface was mostly smooth and had no pores, the adsorption of the particles was weak. When the magnetic field strength of the reaction zone 15.4 mT, the diameter of the steel wool significantly increased after a long residence time, and the surface was bound by many particles, which completely covered the steel wool and made the surface rough.

The above results suggest that the separation effect of HGMS on bacteria and suspended particles in wastewater was not the result of natural adsorption; instead, it primarily depended upon the magnetization and magnetic force of the high-strength, high-gradient magnetic field on weakly magnetic particles in water and the stainless steel wool. Because the steel wool was of high magnetic permeability, the reaction area between the internal and external magnetic fields produced bending magnetic field lines in the space near the steel wool, changing the magnetic force and forming a magnetic field gradient. When the magnetic particles passed through the reaction zone, the trajectory was shifted by the magnetic force in the magnetic field gradient and they were trapped and adsorbed onto the magnetic medium [37].

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