A new wastewater treatment system recovering magnetically immobilized microorganisms under strong magnetic field

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Abstract A new generation of high gradient magnetic separation (HGMS) has recently received attention again, especially for its applications in the field of water and wastewater treatment. The reason for this attention is that a newly developed superconducting magnet can be used to easily generate a high magnetic field, under which even weakly paramagnetic materials can be separated at high efficiency. We have developed a new wastewater treatment process using magnetic gel particles containing immobilized microorganisms and magnetic particles. The magnetic gel particles are separated and recovered from the effluent in water and wastewater treatment processes, and are then recycled to a bioreactor directly or reused after storing. In this research, a novel type of magnetic separator without a filter matrix was designed for the separation and recovery of magnetic gel particles with different magnetic characteristics. No backwashing is required for this new type of separator. By using the separator, polyethylene glycol (PEG) gel particles with 2% magnetite were continuously separated and recovered from the PEG gel particles with 0.04% magnetite at an efficiency of around 90%. The PEG gel particles containing nitrifying bacteria and magnetic particles were available for the oxidation of ammonia solution at a slightly lower nitrification rate than the PEG gel particles with nitrifying bacteria but without magnetite.

Keywords Immobilized microorganisms; magnetic gel particles; polyethylene glycol; superconducting magnetic field; magnetic separation; high gradient magnetic separation

Introduction

The purpose of this paper is to advocate a new wastewater treatment process in which immobilized microorganisms can be separated and recovered from the effluent using a novel magnetic separator without a filter matrix and under a strong magnetic field. In a previous work (Ozaki et al., 1991), it was shown that microorganisms immobilized with paramagnetic particles (gel X) and microorganisms immobilized with ferromagnetic particles (gel Y) could be separated from the mixtures system of those immobilized microorganisms (gel X and gel Y) and activated sludge by using a permanent magnet separator and a high gradient magnetic separation (HGMS) with a conventional electromagnet in series. In the experiment, phenol, triethylene glycol and easily biodegradable organics were degraded by gel X, gel Y and activated sludge, respectively, in the bioreactor prior to the separation stage. The process was developed as a new method utilizing magnetically labeled microorganisms. However, the process could not be operated continuously, because the HGMS filter had to be backwashed frequently to prevent the filter matrix from clogging.
HGMS uses the magnetic traction force to capture magnetic particles upon a magnetized fiber filtration matrix. Magnetic entities are attracted and captured in localized regions of a high magnetic field gradient created by a filter matrix such as stainless steel wool placed in a strong magnetic field (Oberteuffer, 1973). In the actual wastewater treatment processes of steel plants, ferromagnetic and paramagnetic particles were treated by HGMS at filtration speeds 50–60 times higher than those of deep bed filtration (Yano and Eguchi, 1979). Other demonstrated uses of HGMS are in clay benification, desulfurization of coal, filtration of nuclear or thermal reactor coolant and so on. With the addition of a magnetic seeding material, such as magnetite, HGMS also removes non-magnetic pollutants, such as suspended solids, organic substances, algae and viruses. In the 1970s and 1980s, many researches were carried out on HGMS theory and application. However, the level of such activity declined thereafter, because of the high cost of electromagnet use and the complicated procedures of filter backwashing and the recovery of seeded materials.

These days, a new generation of HGMS has been developed for industrial separation technology by utilizing superconducting magnets. This type of HGMS has received attention again, especially for its applications in the field of water and wastewater treatment, because the newly developed superconducting magnet generates a magnetic field so strong that even weakly paramagnetic materials can be separated. In addition, the system has a lower initial cost than the old type and consumes less energy than the conventional electromagnet. Watson et al. (1996) made a fundamental investigation of heavy metal treatment by HGMS of direct precipitation as metallic sulfides or with bacterially produced FeS magnetic adsorbents. The application of superconducting HGMS was investigated in the purification of wastewater from a paper manufactory (Nishijima et al. 2001). The on-site landfill leachate treatment has been tested by a system combining electrolysis and superconducting HGMS (Ihara et al., 2002).

We have proposed a new wastewater treatment system in which magnetic gel particles containing immobilized microorganisms and magnetic particles from a bioreactor can be continuously separated under a strong magnetic field using a novel type of separator without a filter matrix. Figure 1 shows an example of the process, in which two kinds of gel particles (gels X and Y) are used. For example, gel X includes paramagnetic (feeble magnetic) particles and a kind of microorganisms (x) degrading a recalcitrant organic substance, while gel Y includes ferromagnetic particles and other kinds of microorganisms (y). The wastewater containing recalcitrant organic substances is treated by both gels in the bioreactor. The Y gel particles in the effluent from the reactor can be separated and recovered from the main flow by a conventional magnetic separator (eternal magnet use), whereas the X gel particles can be separated by a superconducting magnet after flowing out from the separator.
A novel type of separator without a filter matrix was designed for the separation and recovery of magnetic gel particles with different magnetic characteristics. The separator is the most important part for operating the proposed new wastewater treatment system. No back washing is required for this new type of separator. The conditions and efficiency of the separator for separating the magnetic gel particles were investigated. In addition, the substrate treatment performance by magnetic gel particles was examined, using nitrifying bacteria as an example.

Materials and method

Magnetic gel particles
Polyethylene glycol (PEG) pre-polymer was used as the main chemical for the immobilized gel containing microorganisms and magnetic particles, because PEG is comparatively harmless to microorganisms and in the gel form it is more resistant to feeding by a pump or agitation than are other kinds of gels. Actually, for the past several years, several sewage treatment plants in Japan have employed the process with PEG pellets (called the Pegasus process) to enhance nitrification, without the exchange of pellets (Sumino et al., 2001). In the modified procedure, activated sludge including nitrifying bacteria and magnetic particles (magnetite) was suspended in a PEG pre-polymer solution containing a promoter and mixed with an initiator at pH 7. This mixture was immediately polymerized. The gel was then cut into cubic pellets of 3mm. The composition of pellets was 10% (w/v) PEG, 0.5% (w/v) promoter, 0.25% (w/v) initiator, 2% (w/v) activated sludge and a small amount of magnetic particles (magnetite: Fe₃O₄). The magnetite content in the pellets was 2% (w/v) (magnetic gel particles A) or 0.04% (w/v) (magnetic gel particles B). Both prepared pellets were blended with a mixer and sieved in the size of 500 to 800 µm. The density of gel particles was approximately 1.0 g/cm³.

Developed magnetic separator and experimental procedure
The novel type of HGMS separator shown in Figure 2 was developed to separate and recover gels with different magnetic characteristics. The separator, divided into two a flow zone and a separation zone by a plastic partition, was made from a transparent plastic canister to allow observation inside the separator. The separator has one inlet (inlet (a)) for the inflow of treated water along with immobilized gels from a reactor, and two outlets, outlet (b) for effluent and outlet (c) for recovering the separated gels that have flowed through the separation zone from a gap (3 mm) in the recovery outlet (d). In this experiment, the separator was placed in the strong magnetic field of a superconducting magnet or in that of an electromagnet.

Figure 2  Schematics of new type HGMS separator
Experimental setup and procedure

The experimental set up is illustrated in Figure 3. The magnetic field was applied perpendicular to the stream flow. The total amounts of 2% gel particles (the mixed gel particles of 1% magnetic gel particles (A) and 1% magnetic gel particles (B)) were dispersed in the mixing tank. Among the gel particles (A) and (B) flowing into the zone, almost all of the (A) particles might be attracted to the direction of outlet (d) by the magnetic traction force against the drag force of fluid, according to the magnetic characteristics of gel particles. As a result, the (A) particles should be concentrated in the separation zone, whereas almost all of the (B) particles should flow out from outlet (b). The (A) particles concentrated in the separation zone were sucked at a constant flow rate of 1.16 ml/sec (1.56 cm/sec) by a pump. The experiment was carried out continuously at the fluid velocity of 9.2 to 24.7 cm/s in the flow zone under the magnetic field (H) of $1.07 \times 10^6$ A/m (1.34 T) or $1.33 \times 10^6$ A/m (1.67 T). In order to calculate the recovery ratio and outflow ratio of gel particles, the numbers of (A) and (B) particles flowing out of outlets (c) and (b) were counted for the representative samples collected. The recovery ratio, RR, of gel particles (A) at outlet (c) and the outflow ratio, OR, of gel particles (B) at outlet (b) were calculated as follows:

$$RR(\%) = \frac{\text{Numbers of magnetic gel particles (A) in the effluent at outlet (c)}}{\text{Total numbers of magnetic gel particles (A) and (B) in the effluent at outlet (c)}} \times 100$$

$$OR(\%) = \frac{\text{Numbers of magnetic gel particles (B) in the effluent at outlet (b)}}{\text{Total numbers of magnetic gel particles (A) and (B) in the effluent at outlet (b)}} \times 100$$

A similar experiment was conducted at the total gel concentration of 5% (2.5% (A) particles and 2.5% (B) particles).

We also investigated the effect of magnetic particles added to PEG gel including nitrifying bacteria on nitrification. Batch experiments were carried out using PEG gel particles containing nitrifying bacteria and magnetite or PEG gel particles containing nitrifying bacteria without magnetite, under an aerated condition in ammonium chloride solution. The PEG gels were prepared according to the procedure described above. The resulting gels were crashed and sieved to the size of 50 to 500 µm, and then put into the reactor (5 L container). The weight concentration of gel particles was around 4%. The initial concentration of ammonia nitrogen was approximately 100 mg/l.
Results and discussion

Separation and recovery of magnetic gel particles

The effects of fluid velocity on the recovery ratio and the outflow ratio of gel particles are shown in Figure 4 \((H = 1.07 \times 10^6 \text{ A/m})\) and Figure 5 \((H = 1.33 \times 10^6 \text{ A/m})\). In each figure, the total gel particles content was 2 %. The recovery ratio (RR) of (A) particles at outlet (c) in both figures increased with an increase of fluid velocity in the flow zone, and then decreased after the fluid velocity peaked at around 17 cm/sec. While a small flow of (B) particles continuously entered the separation zone from gap (d), the outflow ratio of the (B) particles at outlet (b) decreased as fluid velocity increased. A possible reason is that greater amounts of (B) particles flowed up with an increase in fluid velocity, but then the suction flow rate in the separation zone was constant. The RR values decreased in higher fluid velocity regions above the maximum point, probably because a larger portion of (A) particles was carried away, without being retained in the magnetic field, to outlet (b) by the stronger drag force in the higher fluid velocity region. This performance of the (A) particles resulted in the decrease of the outflow ratio (OR) of (B) particles at outlet (b) with fluid velocity. It should be noted that the recovery ratio (RR) of (A) particles at outlet (c) was, on the whole, 81 to 93 % under the experimental condition. In addition, the results show that the optimum fluid velocity might provide a maximum recovery ratio (RR) of (A) particles, especially under a higher magnetic field.

Figures 6 and 7 show the effects of fluid velocity on the recovery ratio and outflow ratio of gel particles in the case of a 5 % total concentration of particles. The induced magnetic fields in Figures 6 and 7 were \(1.07 \times 10^6 \text{ A/m}\) and \(1.33 \times 10^6 \text{ A/m}\), respectively. In the figures, both RR and OR decreased with increasing fluid velocity, although the RR values had more than 80 % similarly to those in Figures 5 and 6. It was observed by a video-TV system that the entry to the separation zone (outlet (d)) became clogged with (A) particles.
entrapped by magnetic force. The (A) particles retained around outlet (d) seemed to prevent the (B) particles from entering the separation zone, probably resulting in the higher recovery ratio (RR) of (A) particles in the low fluid velocity region and in the higher outflow ratio (OR) of (B) particles than the respective ratios in Figure 4. In the case of the 10% total concentration of particles, (A) and (B) could not be separated from each other, due to the clogging of outlet (d). Therefore, a larger gap size than 3 mm would be needed in order to separate gel particle at high content. From the results obtained here, it can be said that the separation and recovery efficiency of magnetic gel particles depends on the content of the particles, the flow velocity, and the gap size of outlet (d).

Concentration factor of gel particles

Tables 1 and 2 indicate the concentration factors of magnetic particles (A) at outlet (c) at the total gel contents of 2.0% and 5.0% under a magnetic field of 1.07 × 10^6 A/m. Almost the

**Table 1** Concentration factor (CF) of magnetic particles (A) (magnetic field: 1.07 × 10^6 A/m, total gel particles conc. = 2.0%)

<table>
<thead>
<tr>
<th>Fluid velocity [cm/s]</th>
<th>Content of particle (A) before separation [w/v %]</th>
<th>Content of particle (A) after separation [w/v %]</th>
<th>CF [-]</th>
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<tbody>
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<td>4.4</td>
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<tr>
<td>11.9</td>
<td>1.0</td>
<td>6.4</td>
<td>6.4</td>
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<td>15.0</td>
<td>1.0</td>
<td>7.9</td>
<td>7.9</td>
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<td>18.6</td>
<td>1.0</td>
<td>9.3</td>
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<td>20.8</td>
<td>1.0</td>
<td>10.0</td>
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<tr>
<td>24.7</td>
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same results were obtained under a magnetic field of $1.33 \times 10^6$ A/m. The concentration factor (CF) here is defined as follows:

$$CF(\%) = \frac{\text{Magnetic particles (A) content of the effluent from outlet (C)}}{\text{Magnetic particles (A) content of the influent from mixing tank}} \times 100$$

The CF values increased with fluid velocity and reached a factor of about 9 at the fluid velocity of 18.6 cm/s, providing a high rate of recovery of magnetic gel particles. The concentrated gel particles would be recycled to the reactor directly or reused after storing.

**Effect of magnetic particles on nitrification**

Figure 8 shows the effect of adding magnetic particles to PEG gel on the nitrification rate. Although the nitrifying bacteria in the gel were not so active, the ammonia in the reactor disappeared in 400 minutes. The nitrification rate by PEG gel particles containing nitrifying bacteria and magnetite was a little slower than that by PEG gel particles without magnetite. The magnetite particles in the gel might influence the nitrification rate, probably because they somewhat disturb the diffusion of substrate and oxygen in gel.

**Conclusion**

We have developed a new wastewater treatment process using magnetic gel particles containing immobilized microorganisms and magnetic particles. A novel type of magnetic separator without a filter matrix was designed for the separation and recovery of magnetic gel particles with different magnetic characteristics. By using the separator, PEG gel particles containing 2% magnetite could be continuously separated and recovered from the fluid containing both the above gel particles and the PEG gel particles with 0.04% magnetite, at an efficiency of around 90%. The separation and recovery efficiency of magnetic gel particles was found to depend on the concentration of the particles, the flow velocity and the gap size of the recovery outlet.

**Table 2** Concentration factor (CF) of magnetic particles (A) (magnetic field: $1.07 \times 10^6$ A/m, total gel particles conc.= 5.0%)

<table>
<thead>
<tr>
<th>Fluid velocity [cm/s]</th>
<th>Content of particle (A) before separation [w/v %]</th>
<th>Content of particle (A) after separation [w/v %]</th>
<th>CF [-]</th>
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<tr>
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<td>2.5</td>
<td>28.0</td>
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**Figure 8** The effect of adding magnetic particles to PEG gel on nitrification rate
In addition, a separator with a 3 mm gap for recovery was available to separate particles at a concentration of up to 5% of total gel content. A larger gap would be required for the separation of gel particles of higher content. The nitrification rate by PEG gel particles containing nitrifying bacteria and magnetite was a little slower than that by PEG gel particles without magnetite, probably due to the diffusion limitation of substrate and oxygen in gel.

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**References**


