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Prospectives on Application of Magnetic Powders from Coal Fly Ash for Wastewater Treatment

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Magnetic separation is a method for separating particles based on their magnetic properties. Application of magnetic powders (MP) from coal fly ash (CFA) for wastewater treatment has been considered to be a promising approach to improving the separation efficiency. This paper aims to review the perspectives on application of magnetic powders from coal fly ash (MP-CFA) for wastewater treatment based on three major technological functions. MP-CFA can enhance the treatment ability of phosphorate removal through magnetic flocculation-sedimentation process. Among three commonly used phosphorus removal coagulants, i.e., ferric chloride, aluminum sulfate and poly aluminum chloride (PAC), the addition of MP results in the best performance for the floc settling of PAC coagulation. The alkali modified MP can further improve the coagulation precipitation for chemical phosphorus removal. MP-CFA also can enhance the removal of heavy metals by adsorption and chemical precipitation. The MP effectively adsorbs copper ions at low concentration by removing copper ions through chemical precipitation, which can be speed up by adding the magnetic powder, especially using the alkali modified MP. MP-CFA can be delivered to activated sludge process to improve the solid-liquid separation of sludge from treated wastewater. Activated sludge supplemented with MP exhibits excellent settleability and concentration of mixed liquid volatile suspended solids (MLVSS) can be greatly increased to enhance volumetric biotreatment capacity.

1. Introduction

Magnetic separation is a method for separating particles based on their magnetic properties (Karapinar, 2003). Initially, it was applied in mining and ceramics to separate and collect metals. Since the 1960s, magnetic separation has been widely used in water treatment industry due to its high separation efficiency, small footprint, and easy operation (Anden et al., 1976). It can be divided into two sub-methods, i.e., adding and non-adding magnetic seeding materials. The latter is suitable for the wastewater which contains magnetic substances, such as electroplating wastewater (Krumm, 1992). For non-magnetic wastewater, it is necessary to add magnetic seeding materials in order to fulfil magnetic adsorption or magnetic flocculation. Magnetic adsorption process removes particles via chemical or physical adsorption of fine magnetic seeds. The development of magnetic adsorption is limited by many factors such as high cost of adsorbent, and difficulty of desorption (Nithya et al., 2021). Magnetic flocculation can work effectively by combining conventional coagulation technology with magnetic separation technology together (Zhou et al., 2019). The coagulation process inevitably produces many fine particles, which could greatly affect the quality of effluent (Ye et al., 2017). But magnetic flocculation can enhance the sedimentation of flocs by adding magnetic seeds. The high specific surface area and surface chemical bond of magnetic seeds help magnetic seeds to combine with pollutants flocs to form magnetic flocs, and then the pollutants can be rapidly captured by magnetic disks. Magnetic flocculation not only makes the flocs denser, but also greatly accelerates settling of the particles. It is easy to operate and requires a small footprint. At present, magnetic flocculation technology has been gradually applied to the removal of organic

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matter, phosphorus, heavy metals and other pollutants in wastewaters. The research about magnetic flocculation mostly focuses on dosage of coagulant, dosage of magnetic seeds, dosing sequence of coagulant and magnetic seeds, magnetic field strength, and hydraulic retention time, etc. The nature of magnetic floc, mechanism of binding magnetic seed and flocculation, and reclaiming method of magnetic seeds still need to further investigation.

A challenge for magnetic flocculation is how to strengthen the bonding force between MP-CFA with the flocs. To solve this problem, three methods can be applied i.e. a) use of coagulant aid as a bonding agent between flocs and MP-CFA. Du et al. (2019) added polymeric ferric sulphate (PFS) as the coagulant aid to magnetic flocculation. The result showed that 30 mg/L PFS doubled sedimentation efficiency; b) modification of MP-CFA to improve adsorption performance between flocs and magnetic seeds; c) MP-CFA can be coated with coagulant, leading to the flocculation reaction occurred on the surface of the magnetic seed, which achieves directly contact between the flocs and MP-CFA.

The concentration of mixed liquid volatile suspended solid (MLVSS) in aeration tank is one of factors limiting the treatment capacity of conventional activated sludge (CAS) due to sludge settling performance in secondary sedimentation tank. Magnetic activated sludge (MAS), which is activated sludge mixed with magnetic seeds, can greatly increase the concentration of MLVSS and enhance sludge settling performance. The magnetic activated sludge can be rapidly separated from effluent water by gravity or magnetic force (Sakai et al., 1997). The magnetic seeds work as a media to combine with sludge, and result in rapid solid-liquid separation. In addition, MAS process could maintain high sludge concentration and minimize the amount of waste sludge (Ying et al., 2010). The magnetic seeds in the waste sludge can also be reclaimed, avoiding the negative effect on the sludge treatment devices and minimizing the addition amount of magnetic seeds.

The physical- chemical property of magnetic seeds is critical for magnetic separation. The particle size of the magnetic seeds normally ranges between 48 to 75 μm, which has favourable dispersibility and adsorption performance. Magnetite (Fe_3O_4) has been considered as a magnetic material commonly used for magnetic separation. As it is not cost effective, research has been conducted for cost-effective and easy-to-obtain magnetic seeds for years.

Coal fly ash (CFA) is an industrial waste generated from the thermal power plant, which is also an excellent magnetic material (Lee et al., 2014). It has special micro-sphere structures of the Si-O-Si and Al-O-Al bonds on the surface of CFA, which enhances the adsorption performance (Yao et al., 2015). CFA has high content of iron oxide, which has magnetic property. At present, the research has performed to use CFA for sewage treatment mainly on its adsorption performance (Kankrej et al., 2018). To improve adsorption of CFA, the surface of CFA is modified by acid or base treatment to increase its specific surface area (Pengthamkeerati et al., 2008). Some researchers synthesized zeolite using fly ash by changing surface crystal form to improve the removal of phosphorus, heavy metal and other pollutants (Lankapati et al., 2020).

Application of MP-CFA for wastewater treatment has been considered to be a promising approach to improving treatment efficiency via. the following three functions: a) MP-CFA is used to enhance the treatment capacity of phosphorus removal through magnetic flocculation-sedimentation process; b) MP-CFA is used to enhance the removal capacity of heavy metals by adsorption and chemical precipitation; and c) MP-CFA is delivered to activated sludge to improve solid-liquid separation performance of the sludge. In the previous study, MP-CFA produced by Jiuquan Steel (Group) Co. was used to enhance the precipitation performance for the removal of phosphorus and copper ions, as well as to improve the solid-liquid separation efficiency of activated sludge. The physicochemical parameters including particle size distribution (20-100 μm), pore size distribution (14-30 nm), X-Ray Diffraction (XRD) and magnetic induction intensity (49.8 emu/g) were monitored and analysed. The properties of the MP-CFA used met the application requirements for wastewater treatment. This article aims to review the perspectives on the application of magnetic powder from coal fly ash for wastewater treatment based on three technological functions, i.e., phosphorus removal, heavy metals removal and magnetic activated sludge technology (Figure 1) and described as below.

2. Phosphorus removal by magnetic flocculation

Phosphorus pollution is one of the key factors causing eutrophication of aquatic environments. Coagulation is commonly used method to remove phosphorus from wastewater due to relatively high removal effect and simple operation. Phosphorus removal by coagulation method is ultimately achieved through solid-water separation by sedimentation of flocculent particles which contain P solids. The fine particles inevitably produced during coagulation process have negative impact on sedimentation separation and may lead to low P removal efficiency. Magnetic flocculation allows the magnetic seeds to bound with the fine flocs to speed up settling velocity. The adsorption performance and large specific gravity of magnetic seeds could strengthen the combination with floc particles and enhance the efficiency of sedimentation.

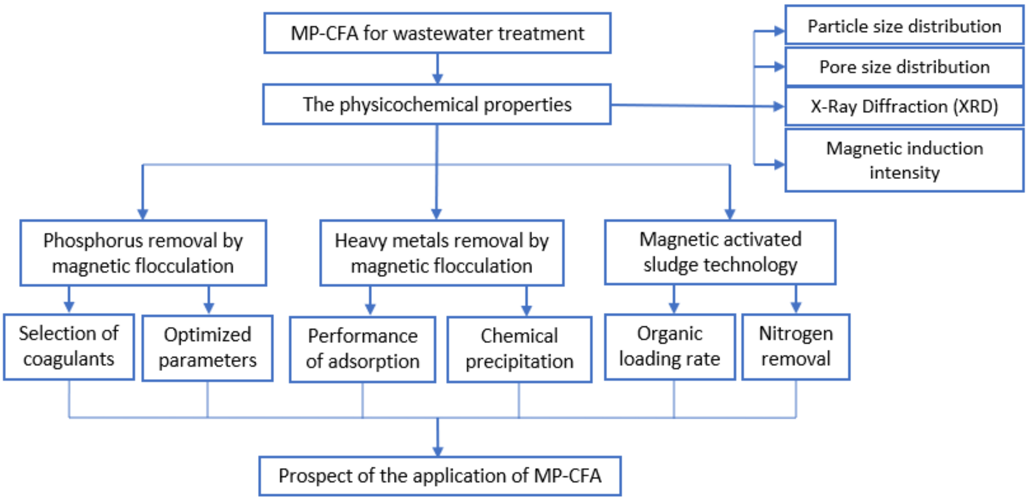

Figure 1: Outline of the procedures for the application of MP-CFA to wastewater treatment.

Studies have shown that magnetic flocculation has high performance for phosphorus removal. Zhao (2012) investigated the effect of the operational parameters on phosphorus removal, including coagulation time, magnetic dosage, coagulant dosage, hydraulic retention time, magnetic field strength and flow rate. The results indicated that, except for mixing time, all other parameters had significant impact on phosphorus removal. Under optimal conditions, phosphorus concentration dropped from 0.5 mg/L to 0.01 mg/L. The research proposed a new magnetic separation system, i.e., open gradient superconducting magnetic separation (OGMS) was introduced before high gradient superconducting magnetic separation (HGMS). As a pre-treatment device, OGMS improved effluent quality and made full use of available magnetic space. Franzreb et al. (1998) determined the optimal dosage of magnetic seeds and the dosage of coagulant. They concluded that without adding magnetite, a higher dosage of coagulant (1.16 times) should be maintained to achieve a 90 % phosphorus removal efficiency. Increasing amount of magnetite had positive effect on the precipitation, which not only reduced the amount of coagulant, but also made the precipitation efficiently. Huang (2004) used aluminium sulphate as coagulant and Fe_3O_4 as magnetic seeds. They found that pH had great influence on phosphate removal. Within pH range of 4.5 to 7.0, more than 95 % of phosphate was removed. They also found that with low magnetic field strength (200 kA/m) and high water flow rate (0.5 cm/s - 6.1 cm/s), the phosphorus removal efficiency of more than 95 % was achieved.

MP-CFA can enhance the treatment ability of phosphorate removal and settling performance of the floc through magnetic flocculation-sedimentation process. In the previous research, the MP-CFA produced by Jiuquan Steel (Group) Co. was used to enhance the precipitation performance for coagulation phosphorus removal. At a concentration of phosphorus of 10 mg/L, the dosage of coagulant, pH, stirring method, the dosage and order of MP-CFA were studied. The reults showed that MP-CFA accelerated flocs precipitation. Among three kinds of commonly used phosphorus removal coagulant, i.e., ferric chloride ($FeCl_3 \cdot 6H_2O$), aluminium sulphate ($Al_2(SO_4)_3 \cdot 18H_2O$) and poly aluminium chloride (PAC), the addition of magnetic powder had the best effect on floc settling property during PAC coagulation. When 6 g/L MP-CFA was added, the turbidity of 0.98 NTU was reached within 1 min and the tubidity decreased 56.1 % compared with non-adding magnetic powder. The floc settling stabilized within 1 min after adding MP-CFA, while coagulation without adding MP-CFA need over 15 min to finish the floc settling. The alkali modified magnetic powder further improved the coagulation precipitation for chemical phosphorus removal. When the alkali modified magnetic powder dosage is 14 g/L, the turbidity decreased to 0.5 NTU.

There will be a large market requirement for the phosphorus removal due to the strict effluent standard for municipal wastewater (lower than 0.5 mg/L). On this basis, the magnetic flocculation can play an important role in phosphorus removal.

3. Heavy metals removal by magnetic flocculation

Water pollution caused by heavy metal has become serious environmental concern, which poses a health risk to human and all living organisms on the earth. The major treatment methods to remove heavy metals in wastewater include chemical, physical and biological methods. Chemical precipitation is the most popular treatment method. Heavy metal can directly react with hydroxide or sulphide to form precipitates, and could also combine with coagulant to form flocs. The solid-liquid separation efficiency impacts the effluent quality greatly. In conventional treatment process, gravity clarifiers and sand filters are designed for the separation of particles containing heavy metals and require larger footprint. In addition, the conversional process requires adjustment of pH within an appropriate range and probably also requires further filtration treatment in order to maintain effluent quality at environmental standards. Magnetic flocculation is expected to simplify this process and reduce the cost. Magnetic seeds work as nuclei during flocculation to form flocculating precipitates incorporated with heavy metal, which settle rapidly due to heavy density of magnetic powers by gravity.

Magnetic flocculation exhibits excellent removal performance for the treatment of heavy metals due to faster flocculation, settling and solid-water separation. Wang et al. (2013) studied the effects of magnetic powder dosage, flocculant dosage, pH and coexistent ion on the removal of copper and zinc ions from simulated tin smelting wastewater. Poly aluminium chloride (PAC) and polyacrylamide (PAM) were selected as flocculants. This research found that the removal efficiency of the copper and zinc ions were more than 90 % and 70 %, when magnetic powder was combined with PAC or PAM. pH had significant effect on the removal efficiency of copper and zinc ions. Magnetic flocculation had excellent performance for the removal of high concentration copper ions. Chen et al. (2016) used poly aluminium chloride (PAC) and anionic polyacrylamide (PAM) as coagulant and flocculant to treat 200 mg/L copper ions with magnetic powder. The dosages of coagulant and flocculant, pH, and dosage and size of magnetic seeds were investigated. The results indicated that high removal efficiencies of 94.72 % was achieved; fast settling velocity, larger floc size, and small volume of floc were obtained simultaneously when magnetic seeds with particle size of 300-400 mesh were used with dosage of 2.0 g/L. Qiu et al. (2017) studied complex nickel removal by magnetic flocculation, and N,N-bis-(dithiocarboxy) ethanediamine (EDTC) as flocculant as well as heavy metal chelating agent. The result showed that the residual Ni concentration was reduced to from 50 mg/L to less than 0.1 mg/L under optimal conditions. Magnetic seeds were reclaimed by more than 76 %. They found that the effective binding sites between flocculation and magnetic seeds increased as the dosage of magnetic seeds increased.

MP-CFA can enhance the removal of heavy metals by adsorption and settling of floc solid during chemical precipitation. Hydroxides have effective removal capacity for heavy metals, which can be quantified. In the previously study, it is investigated that the adsorption of copper ions with three different concentrations (20, 50, 100 mg/L) by magnetic powders and found that the magnetic powder had effective adsorption capacity for copper ions even at low concentration of 20 mg/L. The removal of copper ions through chemical precipitation was enhanced by adding the magnetic powder. The alkali modified MP-CFA had positive effect on the copper removal efficiency, being with the average removal efficiency of 98.7 %, 94.2 % and 86.0 % for the three concentrations of copper ions. The immobilization of metals on alkali-modified MP-CFA was a spontaneous process (Sočo and Kalembkiewicz , 2015). Along with the dispersion of alkali-modified MP-CFA in water, copper ions spontaneously reacted with the hydroxide on the surface of alkali-modified MP-CFA. After the reaction, most of copper hydroxide kept on the surface of the MP-CFA, which was rapidly combined with MP-CFA. In addition, the coagulant aid, such as polyacrylamide (PAM), could strengthen the combination between them through the bridging effect of macromolecules.

The heavy metals face the stringent emission requirement at present. In the future, the magnetic flocculation for the removal of heavy metals will be a cost-effective method with a promising prospectives.

4. Magnetic activated sludge technology

Activated sludge (AC) process is widely used for the wastewater treatment. In order to improve the treatment performance, maintenance of high concentration of mixed liquid volatile suspended solids (MLVSS) could be favourable. The concentration of MLVSS in aeration tank of conventional activated sludge process cannot be maintained at higher than 3-4 g/L due to slow sludge settling performance in secondary sedimentation tank if MLVSS is too high. Membrane Bio-Reactor (MBR), which relies on membrane module to intercept sludge (without secondary sedimentation tank), is one of achievable approaches to separate sludge effectively, can maintain a high MLVSS concentration in aeration tank, reducing facility footprint compared with the conventional activated sludge process. The drawbacks of MBR such as membrane fouling, high cost of membrane modules, high energy consumption and high operational costs have limited MBR application. Delivery of MP-CFA to activated sludge process is an alternative to enhancement of process performance via improving MLVSS concentration and solid-liquid separation. Activated sludge with magnetite powders exhibits excellent settleability and high MLVSS concentration, leading to a higher treatment performance.

Magnetic activated sludge (MAS) technology is considered to be more cost-effective than MBR. As shown in Figure 2, magnetic seed is added to activated sludge to form MAS, resulting in high MLVSS concentration, which can be quickly settled by magnetic field or gravity, as well as has less waste sludge discharge. Moreover, the magnetic seeds in waste sludge can be reclaimed, avoiding the negative effect on the sludge treatment and minimizing the cost due to supplementation of magnetic seeds. MAS is expected to become a promising technological approach of sewage treatment process in the future. Liu (2018) studied the effect of adding magnetic powder to sequencing batch reactor (SBR) on the performance of pollutants removal and microbial community. The results showed that the removal efficiency of ammonia nitrogen (NH_4^+-N) and chemical oxygen demand (COD) in the magnetic activated sludge sequencing batch reactor (MAS-SBR) could be higher by respective 7.76 % and 4.76 % than those of the conventional SBR. The waste sludge of MAS-SBR was reduced by 6.86 % compared with the conventional activated sludge process. The addition of magnetic powder significantly affected the abundance and diversity of microorganisms in the sludge. Ying et al. (2010) used MAS to treat wastewater from actual dairy farms, and compared the effects of continuous aeration and intermittent aeration on the removal of organic matter and nitrogen. The results indicated that regardless with continuous aeration or intermittent aeration, the MAS process had a high removal efficiency of COD (91 %). The intermittent aeration cycle leaded to simultaneous nitrification and denitrification in a single tank in the MAS process with almost zero of ammonia nitrogen in effluent. Sakai et al. (1997) indicated that magnetic separation apparatus separated treated water from sludge suspension of 13-22 g VSS/L within 5 min, and the removal efficiencies of TKN and COD reached 89 % and 92 %.

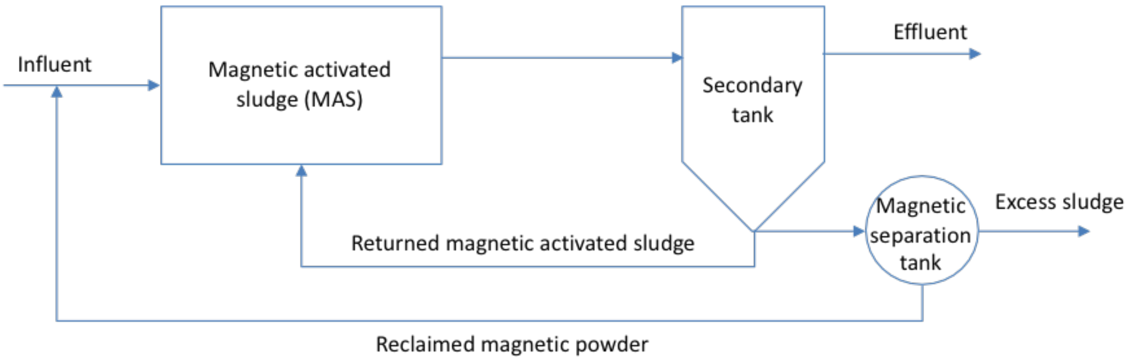

Figure 2: Diagram of magnetic activated sludge (MAS) process

To date, a little research has been conducted using MP-CFA in MAS. In the previous research (unpublished paper), MP-CFA was tested in a laboratory scale of MAS-SBR to investigate the effects of magnetic activated sludge on the removal of COD and ammonia nitrogen through improving activated sludge settling performance. In this experiment, the dosage of MP-CFA was 2.5 g/L. The organic loading rate was increased from 0.037 to 0.67 kg COD/ (kg MLSS·d). The results showed that the sludge volume (SV_{30}) in MAS-SBR was obviously lower than that in control SBR, indicating better settling performance of the sludge. When the organic load rate was increased to 0.5 kg COD/ (kg MLSS·d), the SV_{30} in MAS-SBR was maintained below 30 %, while that in control SBR was increased by more than 30 %. With the increase of the dosage of magnetic powder, better settling performance of sludge was observed. It can be concluded that MP-CFA can improve the volumetric biotreatment capacity through enhancing the solid-liquid separation of activated sludge. In the future, more research work will be carried out on nitrogen removal by using MAS in the anoxic process, or simultaneous nitrification-denitrification process.

5. Conclusion

In this article, the prospective on the application of magnetic powder from coal fly ash for wastewater treatment was reviewed based on three major functions, i.e., phosphorus removal by enhancing magnetic flocculation-sedimentation process, heavy metal removal by enhancing the adsorption and chemical precipitation, and magnetic activated sludge process with high MLVSS concentration and enhanced solid-water separation efficiency. The alkali modified magnetic powder can enhance either phosphorus removal using coagulant of PAC, or heavy metals removal through chemical precipitation. Magnetic activated sludge (MAS) technology is expected to a promising technological approach for future wastewater treatment due to maintenance of high MLVSS concentration and enhanced sludge settleability.

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