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Review

Magnetic separation: an alternative method to the treatment of wastewater

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ABSTRACT

Increasingly strict discharge limits for wastewater have accelerated the research for effective treatment technologies. It is now well known that mineral processing techniques can provide solutions to the environmental problems. Magnetic separation method has been seen in this regard as an alternative method for wastewater treatment. As matter of fact, it has been well known that magnetic separation has been used for the removal of magnetic solid particles from wastewater. Recent progress in this field is the removal of non-magnetic water pollutants such as virus, algae and dissolved pollutants by magnetic separation. Results of research done in the past ten years have shown that the magnetic separation method can also be used for non-magnetic pollutants by seeding the wastewater with magnetic seeds (e.g., magnetite) which preferentially adsorb or co precipitate the material that is to be removed. In such applications, flocculants also may be added to facilitate the agglomeration and removal of the pollutants. With the advantages of high performance, cheapness, simplicity, compact process and workability at high flow rates, the magnetic separation could be an alternative method for wastewater treatment. This article describes the removal of non-magnetic water pollutants from wastewater by the magnetic separation method with a combination of magnetic seeding technique by giving examples of industrial applications in this field.

Keywords: Wastewater treatment; Magnetic seeding; Magnetic separation

1. INTRODUCTION

Magnetic separation is a method for the separation of particles on the basis of their magnetic properties and has been used in the mineral processing and the recycle industries for many years for concentration and purification requirements. Efficient use of magnetic separation methods in mineral processing has encouraged its use in wastewater treatment. The introduction of high gradient magnetic separators (HGMS) in the 1970s has made the magnetic separation method applicable to the handling of weakly submicron magnetic particles. Where the suspended particles to be removed are magnetic, the HGMS is directly applicable without additional unit operation, providing working at flow rates several hundred times faster than conventional filtration. As a matter of fact, the usage of magnetic separators for this purpose is not a new subject. A number of studies have been reported on the utilization of HGMS method for the removal of magnetic pollutants from wastewater (Oberteuffer *et al.*, 1975; Harland *et al.*, 1976; Svobada, 1987). Earlier publications include the application of magnetic separation for the treatment of steel plant wastewater to remove magnetic suspended particles and first full scale HGMS treatment was installed in 1977 at the Chiba Plant of the Kawasaki Steel Corporation to treat the gas scrubber wastewater from the degassing process (Svobada, 1987).

Advantages of magnetic separation such as low cost, simplicity and ability to work at high flow rates have encouraged the studies, which provide the application of magnetic separation to the removal of other water pollutants having no magnetic properties. In recent years, a number of researchers examined the removal of non-magnetic water pollutants such as suspended solid particles, organic compounds, algae, virus and heavy metal ions by using magnetic separation methods (Anderson *et al.*, 1983, Anand *et al.*, 1985; Treshima *et al.*, 1986; Van Valsen *et al.*, 1991; De Reuver, 1994; Yang *et al.*, 1994; Nunez *et al.*, 1995; Franzreb *et al.*, 1998; Gillet and Diot, 1999b).

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As a result, it was proved that the application of magnetic separation method could be used for the removal of non-magnetic water pollutants.

The objective of this study was to undertake a review of the applicability of magnetic separation method to the wastewater treatment for the removal of non-magnetic pollutants. Although there is much laboratory work related with this subject, the review was to be seeking to describe the projects performed in industrial scale. This review has been brought out in the hope that it will encourage an increased interest in the possibility of the application of magnetic separation method to the wastewater treatment.

2. APPLICATION OF HGMS TO WASTEWATER TREATMENT

Separation is an essential unit in wastewater treatment facilities. Many separation systems that are used are sedimentation, filtration, centrifugation and flotation. Amongst them, the magnetic separation method is rather a new and not well-known process, although a number of reports have been published on the subject (Moffat, 1994; Prenger *et al.*, 1994). The main reason for the late introduction of magnetic separation in wastewater treatment has been stated as the poor magnetic susceptibility of the pollutants to be removed from the water and consequent need for specific magnetic separation systems (Van Valsen *et al.*, 1991; Gillet and Diot, 1999a).

Magnetic separation method usually offers a solution to the problem of purifying urban and industrial wastewater. Two types of application are available; the method can be used directly for the removal of paramagnetic solid particles in wastewater streams or for the removal of non-magnetic pollutants with the combination of magnetic seeding and coagulation/flocculation techniques (Moffat *et al.*, 1994). The latter case will be discussed in the following sections.

2.1. Removal of non-magnetic pollutants from wastewater by using magnetic seeding technique

Although most pollutants in wastewater systems are non-magnetic, it is still possible to use magnetic separation for the removal of non-magnetic pollutants such as suspended solids, organic substances, algae, viruses and dissolved pollutants such as heavy metal ions by using magnetic seeding technique which requires the adding of magnetic seeding material to the wastewater. It can be said that the availability of high-gradient magnetic separators in the seventies made this concept feasible for large-scale applications (Svoboda, 1987).

Application of the magnetic separation method to wastewater treatment relies on a strongly magnetic seeding agent like magnetite, which can be prepared chemically or mined from iron ore deposits. Other oxides like Fe_2O_3, Cr_2O_3, or MnO_2 could also be used as a seeding agent, but the magnetite is accepted as the most convenient seeding material for HGMS because of its strong magnetic character and availability. Removal of non-magnetic pollutants by magnetic separation technology requires an additional treatment such as precipitation/flocculation/coagulation. The process has three main stages; attachment of the pollutants to a magnetic seeding material, the subsequent magnetic separation, and finally recovery of the magnetic seed from the sludge. When the pollutants are agglomerated onto the surfaces of magnetic seeding material, they can be swept out from the process water at very high flow rates by a magnetic filter such as HGMS.

The seed, finely divided magnetite prepared chemically or mined, provides a very effective surface for the adsorption of bacteria, viruses, algae and many other colour components (Anderson *et al.*, 1983; Anderson and Priestley, 1983; Kolarik, 1983), for fine phosphate particles (Van Valsen *et al.*, 1991; Shaikh and Dixit, 1992; Franzreb *et al.*, 1998), for radio nuclides (Nunez *et al.*, 1995) and for metal ions (Anand *et al.*,1985; Treshima *et al.*, 1986; Anderson *et al.*, 1991; Chen *et al.*, 1991; Gillet and Diot, 1999b; Feng *et al.*, 2000; White and Athanasiou, 2000; Karapinar, 2000) as well as providing nucleation sites for agglomeration of coagulates.

The preliminary studies on this subject were conducted by De Latour (1973). In his tests on water from the Charles River it was reported that the colour as well as the quality of the filtered water could be improved.

Recent applications of magnetic seeding technique for different wastewater treatment aids are described in the following sections.

2.1.1. SIROFLOC® process for potable water and sewage treatment

The Sirofloc process has been developed by The Commonwealth Scientific and Industrial Research Organization (CSIRO) in Australia. The process was originally developed for potable water treatment and has been further developed for the treatment of sewage and industrial effluents. The process is now marketed as the SIROFLOC® (1) Process For Potable Water Treatment and SIROFLOC® (2) Process For Sewage Treatment (Anderson and Priestley, 1983; Anderson *et al.*, 1983; Bolto, 1996; Boker *et al.*, 1994; CSIRO, 1999).

The mark (1) SIROFLOC® Process was designed to remove colour, turbidity, algae and other impurities from water. After the process development that is technically and economically feasible (Anderson and Priestley, 1983), a fully continuous pilot plant capable of treating 60l/min of highly coloured anaerobic ground water containing significant levels of both soluble iron and H_2S was constructed in Mirrabooka (Anderson *et al.*, 1983). The pilot plant design for this process is shown in Figure 1. At the time of the report (Bolto, 1996), total capacity of constructed or being construction large-scale plants supplying drinking water was given in Table 1.

Table 1
Total capacity of plants (Bolto, 1996)

Mirrabooka, Western Australia	20Ml/d
Bell Bay, Tasmania	20Ml/d
Redmires, UK	20Ml/d
Littlehempston, UK	45Ml/d
Kaohsiung, Taiwan	2Ml/d
Rivelin, UK (under construction)	75Ml/d
Killyhevlin, Northern Ireland (being designed)	36Ml/d

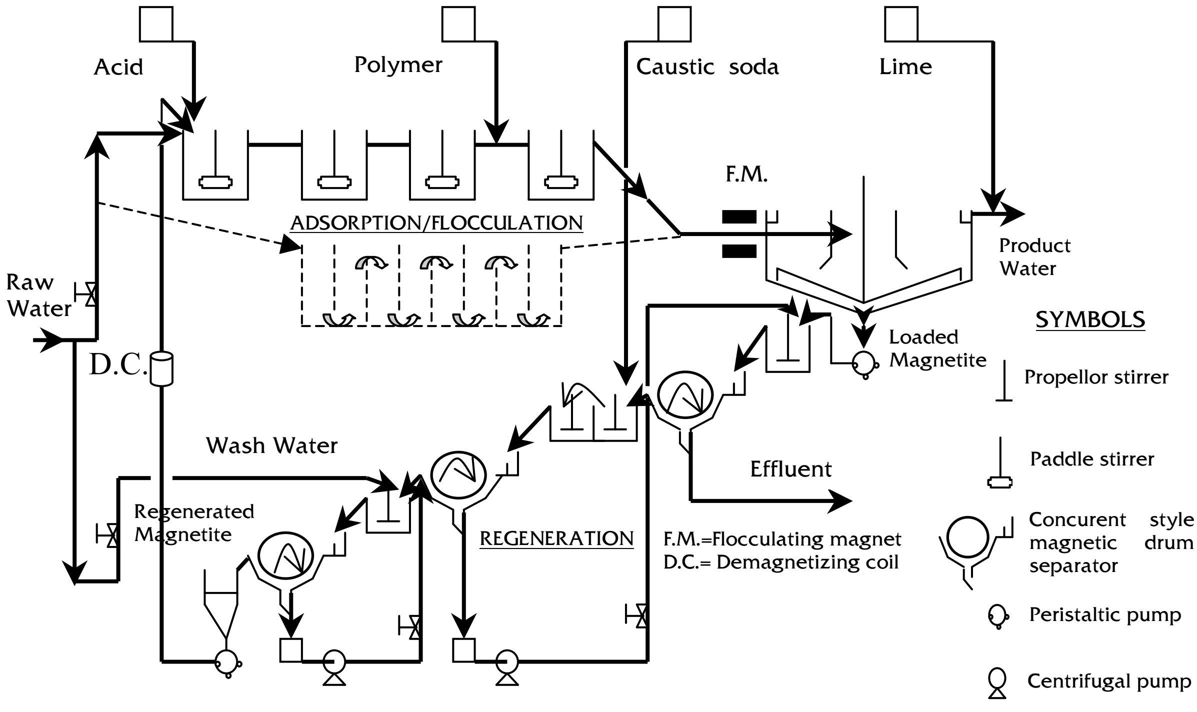

Figure 1. Pilot plant design for the SIROFLOC potable water treatment process (Anderson and Priestley, 1983)

In the mark (1) SIROFLOC® Process, magnetite is used in conjunction with a suitable polyelectrolyte or alum for removing colour, turbidity, algae and other impurities from water. The process based on the adsorption of the impurities onto magnetite and the removal of them together. The oxide rapidly adsorbs the soluble coloured anions of fulvic and humic acid. If turbidity is present, a coagulant aid in the form of a cationic polymer is added to bind the negatively charged turbidity particles to the negative surface of the oxide, as depicted in Figure 2. When magnetically flocculated, the particles are separated rapidly from the treated water. After the separation, the magnetite particles loaded with impurities are treated with alkali or acid to reuse. When magnetite is used with alum, it can also be recycled.

It is reported that the total process time is about 17 min. Apart from the advantages of speed and reuse of primary coagulant, simplicity to start and shut down, reaching specifications for the product water in a few minutes and not being sensitive to low temperatures are among the advantages of this process.

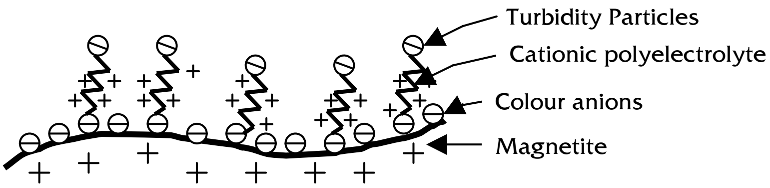

Figure 2. Diagrammatic representation of attachment of turbidity and colour bodies to finely divided magnetite particles, which possess a positive surface charge (Bolto, 1996)

To make a direct comparison, the Sirofloc pilot plant was operated in parallel with a full-scale conventional process. From the results obtained it is claimed that there is no significant difference in operating costs between the two processes and total capital cost is less than for conventional process because of the relative speed and simplicity of the magnetite process. But for hard alkaline waters, magnetite process should show a significant reduction in operating costs. When the sludge handling is considered, conventional process, which includes simple drying of the sludge and then transportation of it to suitable disposal site, are not suitable in many locations and more costly secondary treatment of the sludge becomes necessary. In this concept, sirofloc process has advantages over the conventional process.

The mark (1) SIROFLOC® Process was commercialized and reports claimed that the plants are operating successfully in Australia and UK. The process has been patented by CSIRO, which owns the technology, and licensed to Austep Pty Ltd. and though them to Davy John Brown Pty Ltd. in Australia and Davy Energy and Environment Ltd. in the UK.

The SIROFLOC® (2) sewage treatment process (STP) was developed after the successful application of the SIROFLOC® (1) Water Treatment Process. It is reported that pilot scale trials have been successfully completed at Melbourne and Sydney in Australia, and at Sheffield and Seamer in the UK. The results of these trials were used to design a 5Ml/day prototype of the process at Malabor, Sydney. At the time of the 1994 report, it had been successfully operated since October 1992. The process is owned by the Sydney Water Corporation (50%) and CSIRO (50%) and is licensed to engineering contractors Davy John Brown Pty. Ltd.

The intense public pressure to reduce the environmental impact of the discharge of sewage into the near coastal region made the Sydney Water Board find a solution to improve the treatment level of the their sewage treatment plants. What they needed was a compact process having a similar performance with conventional, biological and secondary treatment to reduce the suspended solids and grease. The SIROFLOC® STP was chosen amongst the processes including dissolved air flotation (DAF) and chemically assisted sedimentation (CAS). In this concept, conventional biological treatment process was not considered because of its high cost.

The process utilizes finely divided particles of magnetite combined with an inorganic coagulant to aid the rapid separation of colloidal and suspended solids from sewage. The magnetite and inorganic coagulant are recovered and reused within the process, thus minimizing the operating cost. A schematic flow sheet of the process is shown in Figure 3.

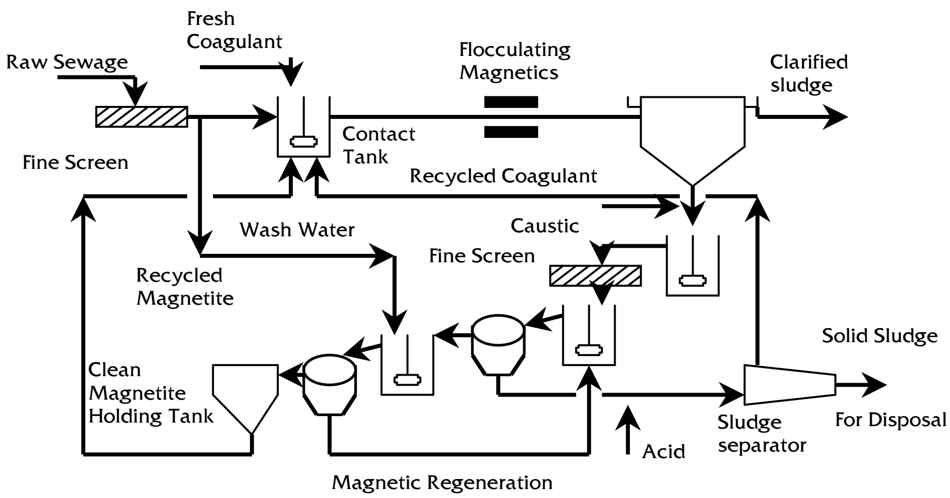

Figure 3. SIROFLOC® sewage treatment plant (STP) schematic layout (Booker *et al.*, 1994)

It is reported that, the process can achieve reductions of about 87% in total suspended solids, 90% oil and grease and 60% COD. The biological oxygen demand (BOD) is about in half. Bacteria, phosphate and heavy metals removals are, 99%, 89% and 73-89%, respectively. Since the soluble material is not removed, there is no reduction in ammonia. The performance is not affected by variations in raw sewage quality and effluent quality. As the use of magnetite allows rapid coagulation and clarification of sewage; raw sewage can be treated to a high quality within 15 minutes, so the process is extremely compact and required very little land space.

Because of the high rate and rapid start-up that can be achieved, the SIROFLOC STP has been proposed as an effective process when there may be rapid fluctuations in sewage flow. Apart from this application, it has been reported that as a high-rate process, SIROFLOC STP has being studied for sewer overflow treatment (Booker *et al.*, 1996).

2.2.2. Magnetic separation of phosphate from sewage

Magnetic phosphate removal is a process to remove and recover phosphates from domestic and industrial wastewater: There are two examples of magnetic phosphate removal on the industrial scale, undertaken in the Netherlands and Germany. In both applications, a new magnet has been designed for wastewater treatment applications and the motivation for the work is to meet the phosphorus discharge limits of Urban Wastewater Directive (91/271) and consequently, the reduction of damage caused by phosphorus discharge to environment.

The process developed in the Netherlands can be summarized as follows: Smit Nymegen Magnetic Water Treatment Systems (MWS), a subsidiary of Dutch company Smit Transformation BV, developed the process. It requires the usage of a magnetic carrier material (magnetite) and it is based on the binding of calcium phosphate/calcium carbonate to its surface.

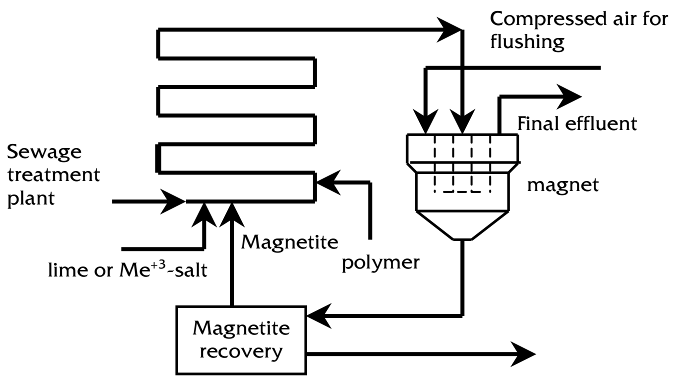

Figure 4. Schematic diagram of magnetic phosphate removal plant (Van Valsen *et al.*, 1991)

In the first stage, phosphates are precipitated with lime (or iron and aluminium salts) and the precipitated phosphates are then attached to the magnetite. In the second stage, removal of phosphates from wastewater is achieved by using a magnetic separation unit, where the applied magnetic field is 0.2 Tesla. After magnetic separation, magnetite is recovered for reuse.

The operating period of the magnetic separator is determined by the loading amount of the matrix. When the matrix is filled up to 70-80% of the separation chamber volume (it takes 30-40 minutes), the flow and the power are cut and the magnetic particles are washed out of the matrix. In the magnetic recovery unit, magnetite is uncoupled from the phosphate particles by shear forces. Thereafter, using a drum magnetic separator for reuse selectively separates magnetite. The remaining product, phosphate in the water, is then processed for final product use.

As mentioned before, a novel feature of this process is the use of a special magnet, which meets the specific used in wastewater treatment processes designed by Smit Nymegen. The new magnet called “Aquamag” has advantages over the conventional magnet that is used in mineral processing facilities. It has no clogging and backwash problems. Moreover, this new magnet has a lower power consumption of 0.02kwh/m^3. Two types of magnet were developed; an electromagnet that is for flows larger than c. 30m^3/h and a disc separator with permanent magnets of the rare earth type for flows smaller than c. 30m^3/h.

At various sewage works, two pilot scale plants (50m^3/h prototype plant and 20m^3/h transportable plant) were tested and the results showed that the magnetic separation reduces the total phosphate concentration (1.3-5.3mg/l) to values as low as 0.1-0.5mg/l with the efficiencies ranging from 80-85%.

Lime was used in the range of 140-300mg/l dosages. The amount of magnetite used was 1000mg/l and the system was able to recover 98% of it. In addition to phosphate removal, BOD and COD levels were also reduced and the system showed a softening effect.

It is reported that the cost of the magnetic phosphate removal with lime is in the range of 15-17Dfl/pe/yr and the process is competitive with other techniques under comparable conditions and requirements. When the lime is used for precipitation, costs are independent of phosphorus influent concentrations. Unlike lime, for iron chloride, costs of the process are increased with increasing phosphorus concentration. The reasons given were an increase in the iron chloride consumption and in the production of iron phosphate sludge that has to be handled and dumped. However, at the conditions where the phosphorus concentration is lower than 5-6mg/l, the use of iron chloride is economically attractive over the use of lime.

No mention is made in the literature of patents or license agreements, however, there are at least two reasons to assume the process is patented; that are full-scale plant applications undertaken in the Netherlands and importation of the Envimag System to US (section 3).

It was reported that this magnetic phosphate removal system could be used as a tertiary treatment at sewage treatment works, as a P-stripping unit in a "side stream" of biological P-removal plants and a P-removal unit for industrial wastewater.

Taking into consideration the requirements to be complied with magnetic separator in water treatment, with the improvement of permanent magnet technology within the last decade, different types of magnetic separators based on permanent magnet were developed at the Institute for Technical Chemistry of the Forschungzentrum Karlsruhe (Germany) (Franzreb, 2001a). The first prototype carousel magnetic separator employed a solenoid and was tested for elimination of magnetite-containing iron phosphate and iron hydroxide flocs (Franzreb *et al.*, 1998). The results show that phosphate concentration up to 14mg/l PO_4^{3-}-P in the inlet were reduced to less than 1mg/l PO_4^{3-}-P in the outlet at the filtration rates about 40m/h. To reduce the energy consumption, the second prototype based on rare-earth magnets was developed and tested for "third-stage" phosphate elimination (Franzreb and Höll, 2000). As compared to the first prototype, the filter throughput has been increased by more than 500% to about 3-4m^3/h with the dimensions and the magnet system used remaining the same. At the inlet concentration of 2mg/l P, separation efficiencies of approximately 80% can be achieved by continuous operation at filtration rates of 100m/h. It was claimed that the system represents a real alternative to the well-established solid-liquid separation methods for phosphate elimination, e.g. sand filters or micro sieves.

In co-operation with the company Steinert Electromagnetbau GmbH an industrial prototype of the carousel magnetic separator was built. This separator will be a part of a mobile pilot plant including in addition a production unit for synthetic magnetite and a flocculation stage. For the application of phosphate elimination, this pilot plant is expected to have a throughput of about 10m^3/h.

Taking into consideration the cost of the sealing of large rotating parts and the risk of wear when the suspension contains abrasive particles, cyclically operating magnetic filters was finally developed. The main feature of this kind of separator is that although it is based on permanent magnets it can be switched "on" and "off". During the operation, the suspension is pumped through the filter until a certain pressure drop or effluent turbidity is exceeded. Then the flow is stopped and the magnet system is switched "off", whereupon a short intensive rinsing, preferably in the counter flow direction cleans the matrix. Finally, the magnet is switched "on" again and a new filtration cycle is started. Thus, it is claimed that the operation of this type of magnetic filter is comparable to one of sand filtration.

The advantage of the system are given as follows; very small energy consumption and cheap. During the year 2001 the system was tested in several pilot applications in steel industry, where the filtration of magnetic micro particles was conducted at filtration rates of up to 1000m/h (Franzreb, 2001b).

2.2.3. Envimag system installed in USA for removing heavy metal ions from industrial waters

This is an example of magnetic separation plant recently installed at a chemical company in the United States (De Reuver, 1994). The Chemical Company where Envimag's wastewater treatment system is in operation produces semi-manufactured plastic products. The production process requires copper as a catalyst; during polymerization, small quantities of the copper catalyst dissolve in the process water and emerge in a 180m^3/h wastewater flow at concentrations between 1000 and 3000 parts per billion.

To meet the discharge limits (40ppb for copper discharge) imposed by the Environmental Protection Agency (EPA), the American Chemical Industry came face with the problem of finding a suitable treatment solution and found a suitable candidate in the Envimag process (previously name the Smit Nymegen Magnetic Water treatment System (Van Valsen, 1991). The process was shown to meet the imposed requirements, workability at high wastewater flow rates and the ability to provide low discharge limits. Both chemical and economical evaluations were done to make a careful selection. At the end, it was found that magnetic separation proved to the most effective amongst the others. There is no detailed information about the other systems that were evaluated in the selection process. Hyper filtration was only given as a one of the other systems that was considered.

As in other applications, this system is also based on the magnetic carrier principle. Pollutants are attached to the magnetic carrier material, magnetite and then removed from the wastewater streams by a magnet. The magnetite is recovered and recycled within the process. In the pre-treatment stage, control of acidity and redox potential are required for the conditioning of the wastewater. pH is controlled by carbon dioxide gas, and sulphide containing chemical is used for the precipitation of the dissolved copper fraction. A polymer is also added to the system to improve the efficiency of the pollutant adherence onto the magnetite. By applying this procedure, it was reported that copper concentration was reduced to the level below of the 20 part per billion.

As in other applications, high separation efficiency, low space requirements in comparison with competitive systems, no blockage and clogging problems because of the open structure inside the magnet and low energy requirement are amongst the advantages of the process. A schematic diagram of the Envimag system is shown in Figure 5.

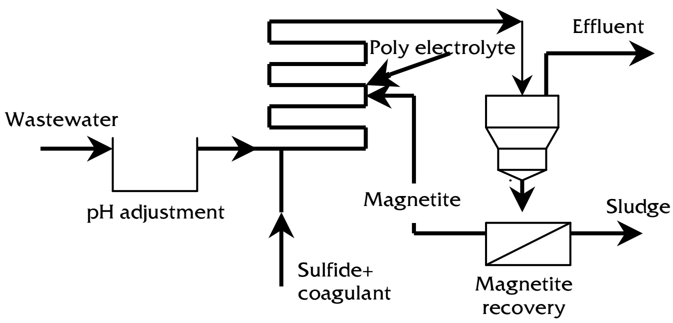

Figure 5. Schematic diagram of the Envimag magnetic wastewater treatment system (De Reuver, 1994)

In this article, the author particularly called attention to a common misunderstanding, that electromagnets consume considerable amount of energy. He dismissed this claim by giving an example from the results of this application. An envimag electromagnets, specially designed for application to wastewater and suitable for a flow rate of $300m^3$/hr consumed only 3-5kW.

The total costs of the process were given as follows; the cost of the skid-mounted installation was approximately US$450 000 and the cost of chemical and energy demand for per m^3 of wastewater are US$ 0.08 and 0.1kWh, respectively.

2.2.4. Magnetic removal of metal ions, depending on superconducting magnets technology

This development was undertaken at the Laboratoire Environment et Mineralurgie in France, and includes the development of a new superconducting separator producing high magnetic fields with low energy consumption (Gillet and Diot, 1999a, b).

Taking in to consideration the high magnetic field requirement for the separation of materials having low magnetic susceptibility and/or very small particle size (generally present in wastewater treatment); they developed a separation system that uses superconducting magnets that are completely automated.

The separator consists of three main elements, the magnet, the cryogenic system and the programmable control unit. A Nb-Ti monolithic multistrand conductor was used to build a super conducting solenoid, which provides a magnetic field of 5 Tesla on the solenoid axis.

Unlike most of the similar systems, this cryomagnetic unit does not require external liquid helium supply. The main features of the system include the operating in a continuous mode and no power requirements once the magnet is energized.

It was reported that this separator could be used in both mineral processing and wastewater treatment aids. Nevertheless, it has been seen that much effort was made to clean up industrial effluents containing dissolved metals. Both the synthetic and the industrial effluent (mostly coming from surface-treatment plants such as rinsing water) were treated by the Superconducting magnetic separator (Gillet and Diot, 2000b).

Magnetite was obtained by the precipitation of a mixed solution of ferrous and ferric salts and the removal of metal ions achieved by adsorption on and/or co-precipitation with the hydrated magnetite. When the matrix is saturated, it is cleaned by either dissolving of precipitate with a circulating acid solution in the field of magnet or by water without a magnetic filed.

The results for industrial rinsing fluids showed that purification performance obtained is more than 99%. It is claimed that the process treats important volumes of industrial effluents charged with metals in solution without adding solid or coagulant salt. When the effluents include small amount of dissolved metals, which are difficult to treat with conventional precipitation and thickening, it is claimed that this method is promising for the treatment. In addition to common advantages of magnetic separation in wastewater treatment, generating high magnetic filed is an important feature of this separator for application of magnetic separation to the wastewater treatment. The process described was patented (French patent 930800-30-06-93(BRGM_INPL).

3. SUMMARY

The examples presented in this paper are industrial –scale applications. There is also much laboratory work undertaken in this subject, which was not reviewed here. The results of laboratory studies indicate that application of magnetic separation to the wastewater treatment is technically feasible. The examples given in this paper are showed that the process is also economically feasible.

Main reasons for the late introduction of magnetic separation in wastewater treatment have been accepted as the poor magnetic susceptibility of pollutants and the need for specific magnet that meets the wastewater treatment requirements. In this regard, magnetic carrier technology can provide the solution for poor magnetic susceptibility of pollutants. It can be said that, in principle, all substances that can be effectively attached to a magnetic seeding material can be separated out by using a magnetic separation method. As for the second reason, innovation showed that a new magnet that is suitable for wastewater treatment application has already been developed. Therefore, there is now no reason not to use this method in the area of wastewater treatment, on the contrary there are many reason to use of it.

Laboratory and pilot plant tests indicate that this technique is capable of excellently purifying wastewater at high flow rates. The magnetic separation technique for wastewater treatment is characterized by high elimination performance, compact process, low cost, workability at flow rates several hundred times faster than conventional filtration and incomparable in many wastewater treatment systems.

It is possible that magnetic separators will be a central component of large water-treatment facilities. Applications show that this innovation has already begun all over the world.

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