Chemical phosphate removal from municipal waste waters: current practice and recent innovations*

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Abstract

The advantages, limitations and disadvantages of conventional chemical phosphate removal processes are reviewed. The need to minimize the costly disadvantages of these processes by optimization is stressed. Process parameters which lend themselves to optimization are identified. Attention is focussed on several innovative processes which overcome some of the inherent disadvantages of the conventional processes. The following innovative processes are reviewed: calcium phosphate crystallization, magnesium ammonium phosphate precipitation, flocculation-filtration and biologically induced chemical phosphate precipitation.

Introduction

With the implementation of the effluent phosphate standard in ''sensitive'' catchments in South Africa (GG, 1984), approximately $1{,}60 \times 10^6\ m^3$ waste water will have to be stripped of its phosphate daily. Davidson and Howarth (1983) estimate that as much as $1{,}30 \times 10^6\ m^3/d$ may initially have to be treated chemically. At an assumed cost of 3 c/m^3 this would amount to a total annual chemical treatment cost of about R14 million for the whole of South Africa. These estimates indicate the importance of chemical phosphate removal technology and the need to ensure its judicious application.

Conventional technology for chemical removal of phosphates from municipal waste waters was developed in the sixties. Since then it has been very extensively applied in Europe, Scandinavia and North America (Wiechers and Van Vuuren, 1979). The major advantage of this technology is that it can be readily applied in existing works, usually without major structural modifications and hence additional capital expenditure. However, it also has a number of disadvantages. These include: high running costs for chemicals and sludge handling and treatment; the need to supply additional plant capacity for sludge thickening, digestion and drying; mineral pollution and the reduction of effluent alkalinity and pH. In order to minimize the impact of these disadvantages, it is essential that the design and operation of chemical phosphate removal processes be optimized as far as is practically feasible. Failure to do this may result in unnecessary expenditure and a poorer quality effluent.

There exists an extensive literature on chemical phosphate removal (Wiechers and Van Vuuren, 1979). Of particular value to designers and those directly involved with process management and operation are two publications detailing American design, operational and practical experiences, viz. the United States Environmental Protection Agency's (US EPA, 1976) *Design Manual for Phosphorus Removal* and the US Water Pollution Control Federation's (US WPCF, 1983) *Nutrient Control: Manual of Practice FD-7: Facilities Design*. A Water Research Commission report (WRC, 1986) incorporating the most recent overseas and local experiences, entitled *Guidelines for Chemical Phosphate Removal from Municipal Waste Waters*, will be published shortly.

This paper highlights the more important design and operational considerations dealt with in the abovementioned *WRC Guidelines* (WRC, 1986). It also focusses attention on innovative processes which overcome some of the inherent disadvantages of conventional processes currently in use.

Process selection and optimization

Chemical phosphate removal involves the addition of chemicals to waste water to precipitate soluble phosphates and coagulate and flocculate colloidal and suspended phosphates. The flocculated mass is then removed from solution by sedimentation. A wide variety of chemicals can be used. Those most readily available in South Africa are aluminium sulphate (alum), ferric chloride, ferric sulphate and lime. The choice of chemical is determined by local availability, cost and phosphate removal effectiveness for the specific waste water under consideration.

Chemical phosphate removal from municipal waste waters is normally combined with existing unit operations such as primary sedimentation, biological treatment (in the case of activated sludge) or secondary sedimentation. The optimum point(s) for chemical addition in a works is site specific. It depends on the characteristics of the raw waste water, the type of biological treatment process being used and the actual physical structure and mechanical equipment which constitute the works.

Phosphate removal using metal salts ($Al_2(SO_4)_3$, $FeCl_3$ and $Fe_2(SO_4)_3$) is a stoichiometric process in that a fixed amount of metal ion is required for each PO_4^{3-} ion removed. As such it is ideal for removing low concentrations of phosphate from large volumes of effluent and the metal salts can readily be dosed into biological filter or activated sludge plants without the need for major capital expenditure. On biological filter plants dosing normally takes place either just prior to the primary settling or the humus tanks, or both, whereas in activated sludge plants it does not appear to be important where the metal salt is added to the mixed liquor. The major disadvantage with this type of process is that, being stoichiometric the dosing rate has to be adjusted according to the concentration of the phosphate in the stream being treated and if the inflow to the plant undergoes large diurnal fluctuations in phosphate concentration, large quantities of chemical can be wasted due to overdosing or substandard effluent can result due to underdosing.

Phosphate removal with lime is a non-stoichiometric process in that the chemical dosing rate is not dependent on the concentration of phosphate removed from the stream being treated. Rather it relies on the fact that the solubility product of calcium

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hydroxy apatite is increasingly exceeded as the pH of the water increases and virtually all of the phosphate will precipitate at pH values above 10. It is therefore ideal for removing high concentrations of phosphate from small quantities of effluent and its main application is in removing phosphate from phosphate rich side streams such as waste activated sludge from nutrient removal plants prior to disposal on drying beds or the supernatant from anaerobic digesters which receive phosphate rich waste activated sludges. It is also used for removing phosphate from the stripper supernatant in the Phostrip® process.

Lime treatment in its conventional form is not favoured as a process for removing phosphate from the main stream of a sewage works for the following reasons:

- It cannot be incorporated into a secondary biological treatment process because the high pH required has a deleterious effect on the organisms and if the pH is lowered further on in the process the calcium hydroxy apatite tends to redissolve. It therefore has to be used as a tertiary treatment and this is capital intensive.

- Because of the large volumes to be treated high dosages of lime are required to raise the pH to the desired level and large quantities of chemical sludge are produced.

Detailed procedures for the selection of the most cost effective chemical and the optimum point of addition are given in the *WRC Guidelines* (WRC, 1986). Although these procedures are used in practice, optimization of the process is often neglected. Since optimization can result in substantial cost savings, those aspects which readily lend themselves to optimization are dealt with below.

Precipitation / Crystallization

The formation of a solid phase by precipitation from solution usually takes place in three steps (Stumm and Morgan, 1970). Firstly, ions or molecules react to form clusters. Once a cluster is sufficiently large and acts as a center for spontaneous growth, it is referred to as a nucleus. The nucleation process determines the size and distribution of precipitate / crystals produced. Secondly, the nuclei grow to form crystallites. Thirdly, large crystals may eventually be formed from fine crystallites by a process called ripening.

Crystal growth kinetics is frequently controlled by interfacial processes such as adsorption and can be modelled by the following empirical rate equation,

$$\frac{dC}{dt} = k.S.(C - C_o)^n \quad \text{.................................} \quad (1)$$

where dC/dt is the rate of crystallization, k a rate constant, S the surface area, C the concentration of the substance being crystallized, C_0 the equilibrium concentration of C, and n an integer (1, 2 or 3) giving the order of reaction.

From the above a number of practical points relating to the optimization of precipitation/crystallization become evident. Firstly, the rate of crystallization (dC/dt) is directly proportional to the surface area (S) available for crystallization (see Equation (1)). In order to let crystallization reactions go to completion as rapidly as possible, so that compact short residence time reactors can be used, it is advantageous to recycle chemical sludge to a point upstream of the point of chemical addition. The advantage of readily available surface area for crystallization may be further exploited by utilizing fixed or fluidized bed crystallization reactors. If no or insufficient surface area is supplied for crystallization, nucleation will take place and may result in the formation of large numbers of very small particles which are difficult to coagulate and flocculate, giving a turbid effluent. Secondly, Equation (1) also demonstrates that the rate of crystallization is a function of the degree of supersaturation ($C - C_o$). It is therefore advantageous to have plug flow crystallizers, or, two or three small completely mixed reactors in series rather than one large completely mixed reactor.

Coagulation

Coagulation is the process whereby particles are destabilized (Bratby, 1980) and is an integral part of the phosphate removal process. Precipitation of phosphates with alum and iron salts as well as lime may give rise to stable colloids or very small particle precipitates which do not settle readily from solution. These require destabilization by coagulation in order to allow flocculation and settlement to take place. In practice the factors which determine optimum coagulation are: the characteristics of the waste water, the type of chemical and dosage used, and the degree of rapid mixing. Conditions for optimum coagulation can be established using jar-tests and by in-plant adjustment of the latter two parameters.

Flocculation

Flocculation is the process whereby destabilized particles, or particles formed as a result of destabilization, are induced to come together, make contact and thereby form large(r) agglomerates (Bratby, 1980). There are two stages in this process. The first stage, perikinetic flocculation, arises from thermal agitation (Brownian movement) and is a naturally random process. Flocculation during this stage commences immediately after destabilization (coagulation) and is complete within seconds. The second stage, orthokinetic flocculation, arises from induced velocity gradients in the liquid. Such velocity gradients may be induced by setting the liquid in motion by passage around baffles or by mechanical agitation within a flocculation reactor, or, where flocs are sufficiently formed, by sedimentation within a settling basin.

For a given flocculating system, the principal parameter governing the rate of orthokinetic flocculation is the velocity gradient applied. The degree or extent of flocculation is governed by both applied velocity gradients and time of flocculation. These two parameters influence the rate and extent of particle aggregation and the rate and extent of breakup of these aggregates. Bratby (1980) has extensively reviewed the theory and practice of flocculation and has detailed design and operational criteria for flocculation. Practical tests for establishing optimum flocculation conditions are given in the *WRC Guidelines* (WRC, 1986).

When adding chemicals in an existing waste-water treatment works it is essential to establish that adequate flocculation takes place. Open channels and stilling basins in humus tanks and secondary clarifiers are often quite sufficient for flocculation purposes. However, due care should be taken to ensure that once the chemical precipitates and biological solids have been sufficiently flocculated, these are not subsequently broken up due to shear forces acting at hydraulic jumps in channels or in constrictions or bends in pipes, prior to being discharged in the settling tank.

Rapid mixing

Rapid mixing refers to the stage in the overall precipitation/coagulation/flocculation process where precipitation chemicals are added to the waste stream to be treated. This stage is possibly the most important operation in the process since it is here that the precipitant/coagulant is thoroughly dispersed throughout the waste stream, precipitation and destabilization reactions occur and where primary floc particles are formed, the characteristics of which markedly influence subsequent flocculation kinetics. Rapid mixing is particularly important in phosphate removal processes since a lack of proper dispersion of the precipitant will predominantly result in the formation of metal hydroxide rather than metal phosphate.

The importance of the efficiency of a rapid mixing facility is evident if the time period required for destabilization is considered, i.e. of the order of 10^{-10} to 1,0 seconds (Bratby, 1980). Hence it is clear that in many instances, the traditional 30 to 60 seconds retention time allowed for rapid mixing are unnecessary and that flocculation efficiency may not improve beyond rapid mixing times of say 5 seconds (Griffith and Williams, 1972). Indeed, beyond a certain optimum rapid mixing time, a detrimental effect on flocculation efficiency may result (Letterman *et al.*, 1973). For a particular waste water and type of coagulant applied, the optimum retention time during rapid mixing is dependent on the velocity gradient and coagulant dosage applied.

Rapid mixing devices commonly used in practice are: impellers, pipe bends, sudden expansions within pipes, orifice plates within pipes and hydraulic jumps in channels. Detailed design criteria have been published by Bratby (1980) and are summarized in the *WRC Guidelines* (WRC, 1986).

Settling

As a result of the coagulation and flocculation effects of chemical addition (alum and iron salts) to raw, activated and humus sludges, mixed biological/chemical sludges commonly have higher settling rates and give clearer effluents (Heinke *et al.*, 1977). Use of poly-electrolytes can further improve the settling rates of the mixed sludges. Recommended design loading rates for clarifiers dealing with treated waste waters containing mixed sludges have been published by the US Water Pollution Control Federation (1983). These recommended loading rates suggest that sludge blanket clarifiers can be loaded about 50 per cent higher than conventional clarifiers. Furthermore, when polyelectrolytes are used conventional clarifier loading rates can be increased by 50 to 100 per cent.

The beneficial effects chemicals have on sludge settling are not necessarily due only to their coagulation and flocculation properties. Chambers and Tomlinson (1982) in reviewing the role of flocculating chemicals on activated sludge bulking, suggest a metabolic effect on the microbiological populations present in the sludge. Rensink *et al.* (1979) have demonstrated that sludge bulking in the activated sludge process can be controlled by the continuous addition of 25 to 50 mg/ℓ iron to the influent sewage.

The following operational aspects require careful consideration when chemical phosphate removal is implemented in an existing works. In activated sludge processes it should be ensured that the solids handling capacity of the secondary clarifiers is not exceeded due to the additional chemical sludge load. In biological filter plants it has been found desirable to have frequent batch or continuous sludge withdrawal from the humus tanks in order to ensure that the chemical/humus sludge blanket does not overflow (Wiechers *et al.*, 1984). This sludge tends to be dilute, but can be thickened by mixing it in with raw waste water, and settling and thickening in the primary settlers. If still too thin, the raw/chemical/humus sludge from the primaries must then be thickened prior to digestion, in order to ensure that the residence time of this sludge in the digester is sufficient for anaerobic sludge stabilization. Another reason for not keeping chemical sludge (particularly ferric phosphate/hydroxide) in the humus tanks for extended periods, is the danger of ferric iron being reduced to the more soluble ferrous form with subsequent loss of iron in the effluent.

Application in activated sludge processes

Ever since the pioneering work of Thomas (1965) in Switzerland during the mid-sixties, chemical phosphate removal in the activated sludge process has been extensively applied on a worldwide scale. Initial reservations that the addition of iron or aluminium salts would adversely affect biological purification in the activated sludge process and subsequent sludge stabilization in the anaerobic digestion process, were proved unfounded.

The amount of iron or alum used to precipitate phosphate in the activated sludge process can be significantly lower than in the biological filter process. The reason for this is probably two-fold. Firstly, because the mixed liquor is continuously recycled and seed mass is supplied for further crystallization and phosphate adsorption. Secondly, the long retention time of solids in the activated sludge process, up to 25 days, allows sufficient time for at least partial recrystallization of ferric and aluminium hydroxides to their equivalent phosphate salts or ion-exchange between the precipitated hydroxide and the phosphate ions in solution. Rabinowitz and Marais (1980) report that when chemical phosphate removal is practiced in activated sludge processes, the metal salt requirements can be as low as that required for stoichiometric reaction between the metal and the phosphate; this applies down to phosphate residuals of 1 to 2 mg/ℓ (as P). Phosphate removal by metal salt precipitation is additive to biological enhanced phosphate removal. A further limitation of the process is the pH at which it normally operates, i.e. between 7,0 and 8,0. This is not optimal for either ferric or alum phosphate precipitation, their optimum precipitation pH values being about 5,3 and 6,3, respectively (US WPCF, 1983).

Application in biological filter process

Currently the only practical means of removing phosphates from waste waters treated in biological filter processes is by chemical means. Alternatives have been proposed, for example, the combination of a high rate Phoredox process (a biological process for the removal of carbon and phosphate from waste water) with an existing biological filter process. This alternative will probably become viable only if the works has to be extended, and then at considerable capital expenditure.

Chemical phosphate removal in biological filter works is usually practiced by dosing chemicals after the filters when most of the complex (poly and organic) phosphates have been converted to the easily precipitable ortho form. If the works is overloaded split treatment prior to the primary settling tanks and the humus tanks may be a viable option. The major problem with phosphate removal in these systems is matching the chemical dosage with the phosphate load. The latter can vary by an order of magnitude or more because of large variations in both phosphate concentration, typically between 3 to 12 mg/ℓ (as P), and flow, typically peak dry weather flow may be 1,5 to 3,0 times minimum dry weather flow (Wiechers *et al.*, 1984).

A number of approaches have been developed for dealing with variable phosphate loads. For example, chemical dosage adjustment once per shift or at more frequent intervals; dosage proportional to flow with or without additional dosage adjustment matching changing phosphate concentrations. A new package system has recently been developed in South Africa (with the advantage of readily available local spares and technical assistance) which doses chemicals proportional to flow and adjusts the dose to match changes in phosphate concentrations based on predetermined daily phosphate profiles (Sugden, 1985). Gleisberg (1985) has demonstrated that about one third of the amount of precipitant required when it is added in constant amounts can be saved when the dosage of precipitant is controlled so as to be proportional to the phosphate load.

A problem which must be anticipated in nitrifying biological filter works (and to a lesser extent activated sludge works) which treat low alkalinity waste waters, is the reduction of effluent pH values to as low as 5 or even 4 when chemicals are added to precipitate phosphates. The problem arises due to the acidic nature of alum and ferric salts as well as the acid produced by the nitrification process. In such instances provision must be made for the addition of an alkali, such as lime. This will correct the pH of the humus tank effluent prior to discharge to the water environment or recirculation over biological filters. In such systems the iron or alum usage may be more optimal than at higher pH values, since the optimum pH for precipitation lies in the lower pH range. Consequently, the alkali should only be added to the waste water stream after precipitation has gone to completion. Due care must, however, be taken to ensure that plant and equipment are adequately protected against this corrosive low pH waste water.

Innovative processes

Calcium phosphate crystallization

A promising new development in the field of chemical phosphate removal from waste waters is the calcium phosphate crystallization process. Van Dijk and Braakensiek (1984) developed a fluidized bed configuration in the Netherlands, whereas Joko (1984) developed an upflow fixed bed configuration of this process in Japan. The major advantage of the process, over conventional chemical phosphate removal techniques, is that calcium phosphate pellets form and virtually no sludge is produced, and hence the handling, treatment and disposal problems associated with conventional chemical sludges are circumvented. The fixed bed process configuration also has demonstrated stability and a propensity for suspended solids removal. Potential disadvantages of the process are the use of lime with its associated operational problems as well as the increased hardness (by about 50 mg/ℓ as $CaCO_3$) and pH (to about 9,5) of the treated effluent.

The process appears to have considerable potential for phosphate removal from biological filter and activated sludge process effluent. It may also find application where phosphate precipitation with lime is currently practiced, e.g. in the Phostrip® process. Research at the University of Cape Town has established the viability of the process for precipitating phosphate from waste streams from the biological phosphate removal UCT process (WRC, 1984). The calcium phosphate crystallization process appears particularly promising for low strength sewages with a high phosphate content (Marais, 1985).

Magnesium ammonium phosphate precipitation

The Italian Water Research Institute has patented a process for phosphate and ammonia removal by ion-exchange and magnesium ammonia phosphate precipitation (Liberti *et al.*, 1981). By using clinoptilolite and a strong anionic resin, as well as a unique scheme of operation and regeneration, a secondary effluent is produced with an ammonia content of less than 10 mg/ℓ (as N) and phosphate content less than 1 mg/ℓ (as P). The polluting ions NH_4^+ and PO_4^{3-} are removed from the regenerant solutions by precipitation using magnesium chloride to form $MgNH_4PO_4$, a commonly used fertilizer. The process also removes bacteria and suspended solids. Cost evaluations show the process to be competitive with conventional chemical phosphate removal, and sale of magnesium ammonium phosphate could make it an even more attractive process. Biological fouling of the resin was not considered a problem, because macroporous resins are used. High sulphate waters may present a problem, since sulphate ions are exchanged as readily as phosphate ions. Other potential disadvantages are the complexity of the process and the uncertainty as regards the potential of $MgNH_4PO_4$ as a fertilizer in South Africa.

The magnesium ammonium phosphate precipitation process may find an application in South Africa for the treatment of anaerobic digester supernatants. These liquors are normally recycled back to the works but place high ammonia and phosphate loads on such works. Magnesium ammonium phosphate precipitation in pipelines transporting liquors from digesters already presents problems, i.e. by blocking them (Pitman, 1985). A precipitation reactor immediately after the digester may solve this troublesome operational problem, as well as reducing the phosphate and ammonia loads on the works.

Flocculation-filtration

Sulzer (Pty) Ltd of Switzerland has developed a process for removing phosphate from secondary effluents from levels of 1 to 2 mg/ℓ down to 0,1 to 0,2 mg/ℓ (as total P). This process which comprises a conventional in-line flocculation stage, followed by mixed media filtration, is known as the flocculation-filtration process. It was developed at pilot-scale over a three-year period and is currently applied at full-scale at four works in Switzerland and one in Germany. The largest installation is at Zürich's Werdhölzli works where it is expected that an average dry weather flow of 390 Mℓ/d will be treated by 1986.

Although this process is capital intensive, its running costs are low because only about 3 to 5 mg/ℓ iron salts (as Fe) are required for phosphate precipitation. Sulzer has demonstrated that under Swiss conditions the costs associated with this process constitute only about 10 per cent of the total process cost for waste water treatment. It is in fact cheaper than simultaneous precipitation chemical phosphate removal when applied to achieve a 1 mg/ℓ (as P) effluent standard and gives a much higher quality effluent, i.e. a total phosphate of less than 0,2 mg/ℓ and suspended solids of less than 5 mg/ℓ.

Biologically induced chemical phosphate precipitation

Phosphate removal in biological nutrient removal plants may be partially due to biologically induced chemical precipitation. Mino and Matsuo (1985), Arvin and Kristensen (1984) and Kerdachi and Roberts (1984) have all reported finding inorganic phosphate precipitates in sludges taken from biological nutrient removal plants. Reservations have been expressed concerning the method used to identify and quantify these precipitates, but it is currently being critically evaluated and further developed (Lötter, 1985). If chemical phosphate precipitation can be biologically induced, this may have significant practical and economic implications, e.g. no or only small quantities of chemicals need be used to remove phosphate from municipal waste waters. Arvin (1983) has proposed mechanisms for biologically induced chemical precipitation, but to date little research has been undertaken to validate the mechanisms and quantify the parameters which control them.

Conclusion

Current knowledge on chemical phosphate removal has been consolidated in the form of technical guidelines (WRC, 1986) in the Water Research Commission publication *Guidelines for Chemical Phosphate Removal from Municipal Waste Water.*

Research and development on conventional chemical phosphate removal processes are continuing in South Africa. The City Council of Boksburg and a consulting engineering firm, Scott and De Waal (Inc.), are currently investigating chemical phosphate removal at the Vlakplaats Works, Boksburg, South Africa under contract to the Water Research Commission. The emphasis is on process optimization procedures and the quantification of volumes and masses of chemical sludge formed in the process.

Innovative chemical phosphate removal processes need to be further researched, developed and tested in practice. Work on biologically induced chemical phosphate precipitation is being conducted by the staff of the Town and City Councils of Pinetown and Johannesburg, respectively. The calcium phosphate crystallization process appears to hold potential as a tertiary polishing process particularly for large waste-water treatment works. The magnesium ammonium phosphate precipitation process holds considerable potential for the insolubilization of both ammonia and phosphate from anaerobic digester liquors. The flocculation-filtration process should be considered as a cost-effective means to achieve very low effluent phosphate concentrations which may possibly be required in future from local authorities discharging effluents to rivers and streams which flow into excessively eutrophied impoundments.

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