Theoretical and experimental investigation of phosphate removal from seawater by multi-stage coagulation

Wei Jin, Dongliang Hao, S. Assiyeh Alizadeh Tabatabai, Maria Kennedy and Jan C. Schippers

ABSTRACT

Phosphate removal from seawater is important for biofouling control on RO membrane because phosphorous is one of nutrients for microbial growth. This paper is based on the hypothesis that multi-stage coagulation results in better phosphate removal. Therefore, comparison of phosphate removal of one-step and three-step dose coagulation from the aspect of both the theoretical calculation and experimental results is investigated in this paper. The result of theoretical calculation based on Freundlich equation shows that final phosphate concentration of three-step dose i.e. 0.43 μgP/L, is 10 times lower than that with one-step dose i.e. 4.47 μgP/L. The experimental result shows that for the three-step dose, final phosphate concentration is 1.0 μgP/L which is lower than one-step dose (i.e. 4.0 μgP/L), but not as low as the theoretical calculated value (0.43 μgP/L). This discrepancy between theoretical calculation and experimental result may be the impact of equilibrium phosphate concentration, different initial Fe:P molar and NOM competition between one-step dose and three-step dose coagulation. Although this discrepancy exists, experimental results still showed that multi-stage coagulation presented better phosphate removal in seawater to concentration levels that are lower than conventional coagulation. In other words, the problem of the high coagulant dosage in the pretreatment process while removing phosphate from seawater may be solved by application of multi-stage coagulation instead of conventional coagulation.

Key words | multi-stage coagulation, one-step dosing, phosphate removal, three-step dosing

HIGHLIGHT

- This paper is structured on one hypothesis in which multi-stage coagulation results in better phosphate removal. Therefore, comparison of phosphate removal of one-step and three-step dose coagulation from two aspects of theoretical calculation and experimental results is investigated in this paper.

INTRODUCTION

Biofouling is the formation of biofilms on membrane surface by deposition and growth of micro-organisms from raw water. Microbial growth needs nutrients such as (carbon(C), nitrogen(N), and phosphorus(P)). Therefore, limiting these nutritional compositions in the feed solution has been introduced in attempts to prevent or restrict microbial growth, instead of killing microorganisms using chemical biocide (Kooij et al. 1982; Mathieu et al. 1992; Niquette et al. 2000;
Kim et al. (2014). Among these three nutrients, phosphate is a potentially significant limiting factor as the molar ratio of carbon, nitrogen and phosphorous is 100:20:1.7 in a microbial biomass (Vrouwenvelder et al. 2010; Kim et al. 2014), an indication that a much lower concentration of phosphate than carbon is required for cell growth (Wende et al. 1989; Alphenaar et al. 1993; van der Aa et al. 2002). Therefore, even low concentrations of phosphate can more significantly affect the microbial status than other major nutrients (carbon and nitrogen) (Kim et al. 2014).

Jacobson (Jacobson et al. 2009) suggested that phosphate limitation in RO systems may be an option in controlling biofouling which was only linked to decreased cleaning frequency (Table 1). It shows that chemical cleaning of the RO units was much less frequent in the non-summer months due to the lower phosphate concentration in these months than in summer both in Plant A and Plant B. And Vrouwenvelder (Vrouwenvelder et al. 2010) experimentally showed low phosphate concentration was able to control biofouling by measuring biomass concentration, which is summarized in Table 1 that when ‘limiting P’ (no P dose) in RO feed, biomass concentration of $2.5 \times 10^2$ pgATP/cm² is much lower than that of $6 \times 10^4$ pgATP/cm² with $20 \mu$gP/L dose in RO feed water. Later, Chang-Min Kim investigated effects of phosphate limitation on performance of FO. By measuring biomass parameter of total cell number decrease, it was discovered that the phosphate limitation in FO feed water greatly reduced microbial growth (Table 1). Jacobson and Vrouwenvelder proved that phosphate limitation can control biofouling. However, the problem is that high chemical dosage in pretreatment process for the removal of phosphate causes high operational costs. Moreover, in the existing researches, only surface water and synthetic water as raw water are studied for phosphate limitation and no relevant research is currently available concerning the seawater.

Phosphorus removal started in the 1950s in response to the growing problem of eutrophication and the level of phosphorus needed to be reduced before discharging to surface water. Chemical precipitation was the initial method applied to remove phosphorus (Jiang & Graham 1998). Furthermore, biological phosphorus removal (Oehmen et al. 2007), coagulation (Wende et al. 1989) and adsorption (Zeng et al. 2004; Zhang et al. 2007; Liu et al. 2008) have

| Reference source          | Raw water             | Treatment                          | Phosphate concentration of RO or FO feed | Biofouling investigation                        |
|---------------------------|-----------------------|------------------------------------|-----------------------------------------|------------------------------------------------|
| Jacobson et al. (2009)    | Plant A: Canal water  | RSF(8 mL/LPAC) + In-line Coagulation (2–5 mg/LFeCl₃ dose) + UF(100 kDa) + RO | Summer: 3.02 µgP/L Spring: 1.56 µgP/L | Summer(RO cleaning frequency): 2 Non-summer (RO cleaning frequency): 0.5 |
|                           | Plant B: River water  | In-line Coagulation (6 mg/LFeCl₃ dose) + Sand Filter + UF(150–200 kDa) + RO | Summer: 3.95 µgP/L Spring: 2.44 µgP/L | Summer(RO cleaning frequency): 3 Non-summer (RO cleaning frequency): 1 |
| Vrouwenvelder et al. (2010)| Surface water       | MS + Coagulation + RSF + BACF + UF + RO | 20 µgP/L dose                            | Biomass concentration: $6 \times 10^6$ pgATP/cm² |
|                           |                      |                                    | ‘Limiting P’ (no P dose)                 | Biomass concentration: $2.5 \times 10^2$ pgATP/cm² |
| Kim et al. (2014)         | Synthetic secondary wastewater effluent | FO                                 | 8.0mgP/L dose                            | Biomass parameter: total cell number $9.6 \times 10^6 (\pm 3.2 \times 10^5)$ cells/cm² |
|                           |                      |                                    | ‘Limiting P’ (no P dose)                 | Biomass parameter: total cell number $3.0 \times 10^6 (\pm 5.5 \times 10^5)$ cells/cm² |
been established gradually. Chemical precipitation and biological removal are generally suitable for removing phosphate with higher concentration and usually the concentration of effluent is still high. Coagulation and adsorption is proposed as an effective removal process for the low concentration of phosphate (Tian et al. 2009). Therefore, for the low phosphate concentration in seawater, coagulation and adsorption are usually applied for phosphate removal.

Many kinds of solid materials including iron-based compounds (Bastin et al. 1999), aluminium oxide hydroxide (Tanada et al. 2003), iron oxide tailings (Zeng et al. 2004), mesoporous (Liu et al. 2008), Fe-Mn binary oxide adsorbent (Zhang et al. 2007) have been used to remove phosphate from water by adsorption. In adsorption modelling, the distribution of adsorbate between the two phases (the bulk solution and the adsorbent) is often described in terms of isotherms. The amount of solute adsorbed per unit of adsorbent (q) as a function of the equilibrium concentration of the solute in bulk solution (Ce), at a constant temperature, is called the adsorption isotherm. Five isotherm equations are used in the practice including the Freundlich, Langmuir, Temkin (two-parameter equations) and Redlich-Peterson and Langmuir-Freundlich (three-parameter equations), as described below:

Freundlich Equation $q = K C_e^{1/n}$

Langmuir Equation $q = b q_m C / (1 + b C)$

Temkin Equation $q = A + B \ln C$

Redlich-Peterson Equation $q = a C / (1 + b C^n)$

Langmuir-Freundlich Equation $q = b q_m C^{1/n} / (1 + b C^{1/n})$

Although three-parameter equations (Redlich-Peterson and Langmuir-Freundlich) often provide a better fit of the isotherm data, two-parameter equations (Freundlich, Langmuir, Temkin) are more widely used in the practice due to the convenience of evaluating two parameters than three parameters (Zeng et al. 2004).

In the case of phosphate removal by coagulation, two coagulants including ferric chloride (FeCl₃.6H₂O) and aluminum sulfate (Al₂(SO₄)₃.14H₂O) are commonly used. Two major mechanisms occur when these are added in water (Jiang & Graham 1998):

Formation of Al/Fe-hydro-phosphate complexes with general formula of $\text{Me(OH)}_x(\text{PO}_4)y$. (M = Al/Fe). These complexes either adsorb onto positively charged Al/Fe hydrolysis species or act as centres of precipitation for Al/Fe hydrolysis products.

Adsorption of phosphate ions onto the surface of Al/Fe hydrolysis species. In water treatment practice, amorphous Al(OH)₃ or Fe(OH)₃ are the predominant hydrolysis species.

Based on these mechanisms, coagulation and adsorption have similar mechanisms. Al/Fe hydrolysis species play a role of adsorbent. In order to obtain the phosphate removal capacity by coagulation, adsorption isotherm is adopted to describe amount of phosphate removed per unit of coagulant as a function of the equilibrium concentration of phosphate since mechanism of phosphate removal by coagulation is adsorption of phosphate and Fe-hydroxide-phosphate complexes onto iron hydrolysis species. Therefore, the aim of this study is: (1) to investigate phosphate removal capacity based on adsorption isotherm; (2) to investigate multi-stage coagulation for phosphate removal.

MATERIALS AND METHODS

Raw water

Raw water was collected from the intake of a seawater-UF/RO pilot desalination plant in Zeeland Province, the Netherlands. Samples were collected in clean, dark-glass bottles and stored at 4 °C. Experiments were performed in UNESCO-IHE laboratory within a week after sample collection. Prior to each experiment, bottles were shaken gently to ensure particulate matter was brought back to suspension.

Coagulant

There are two conventional chemicals used in coagulation for phosphate removal, i.e. ferric chloride and aluminum. In practice, a better phosphate removal by use of ferric chloride has been observed in desalination plants. Therefore, FeCl₃.6H₂O was used as coagulant in this research.
Chemicals

Chemicals used for phosphate measurement together with the related preparations are presented in Table 2 (Jin et al. 2014).

Experiments for investigation on phosphate removal capacity by coagulation (for isotherm equation)

The experiments for investigation on phosphate removal capacity by coagulation were performed in seawater with six different initial phosphate concentrations, i.e. 34, 49, 77, 106, 329, 1,006 μgP/L, at same coagulant dose of 1 mgFe³⁺/L. These six different initial phosphate concentrations were obtained by adding different amount of stock P solution (see section 2.3) to seawater. Coagulation was conducted in the compact laboratory mixer (Models CLM4 EC Engineering, volume: 1,000 mL) with rapid mixing intensity $RMG = 1,100 \text{s}^{-1}$, mixing time $RMt = 20 \text{s}$ and slow mixing intensity $SMG = 40 \text{s}^{-1}$, mixing time $SMt = 30 \text{min}$. Afterwards, the flocculated solution was taken and filtrated for the final phosphate concentration measurement. Figure 1 shows the main steps of the whole experimental process.

One-step dosing and three-step dosing coagulation

One-step dosing coagulation was done in seawater with initial phosphate concentration of 117 μgP/L at a coagulant dose of 1 mgFe³⁺/L. It was performed in the compact laboratory mixer (Models CLM4 EC Engineering) under condition of rapid mixing intensity $RMG = 1,100 \text{s}^{-1}$, mixing time $RMt = 20 \text{s}$, and slow mixing intensity $SMG = 40 \text{s}^{-1}$, mixing time $SMt = 30 \text{min}$.

Three-step dosing coagulation was performed in seawater with initial phosphate concentration of 117 μgP/L under condition of rapid mixing intensity $RMG = 1,100 \text{s}^{-1}$, mixing time $RMt = 20 \text{s}$, and slow mixing intensity $SMG = 40 \text{s}^{-1}$, mixing time $SMt = 10 \text{min}$, 0.35 mgFe³⁺/L dose for first step. The same procedure was applied for second and third step and only the dose for third step was changed to 0.30 mgFe³⁺/L. The total dose was thus 1 mgFe³⁺/L, which is the same dosing amount as applied in the one-step dosing.

Analytical method

Filtration of flocculated solution

Flocculated solution was filtrated through a 0.45 μm cellulose acetate filter before the phosphate measurement. The filtration procedure of flocculated solution is described below:

- The filter holder and syringe were soaked in (1 + 4) HCl or 10% H₂SO₄ for at least one hour and then rinsed with ultra pure water at least three times to take out any traces of phosphate.
- The 0.45 μm filter was set into the filter holder.
- A certain amount of flocculated solution was drawn into the syringe, screw the filter set into the syringe then push it to filtrate.
The first 5 mL of permeate was discarded and the rest of permeate collected into a clean PE bottle for phosphate measurement.

Phosphate measurement

An existing analytical method of phosphate measurement is ascorbic acid method in which is ‘orthophosphate reacts with ammonium molybdate to form molybdophosphoric acid. Through reductant (ascorbic acid), the molybdophosphoric acid is transformed into molybdenum blue whose absorbance of can be measured on a spectrophotometer at 880 nm’. However, for phosphate concentrations below 10 μg/L it is necessary to extract the molybdenum blue via hexanol and measure the extracted solution on a spectrophotometer at 680 nm, which is known as modified ascorbic acid method (Jacobson et al. 2009).

RESULTS AND DISCUSSION

Phosphate removal capacity based on adsorption isotherm

The experimental results of phosphate removal by coagulation are presented in Table 3. It can be observed that the final phosphate concentration after coagulation is 1.1, 1.8, 2.7, 3.7, 67 and 605 μgP/L when initial Fe:P is 16:1, 11:1, 7.2:1, 5:1, 1.7:1 and 0.5:1, respectively. Phosphate removal efficiency is increased with increase in initial Fe:P molar ratio when the ratio is below 5:1. Phosphate removal efficiency becomes almost constant at molar ratio (Fe:P) in excess of 5:1 molar ratio (Fe:P) which indicates that the initial Fe:P molar ratio of 5:1 is critical point for phosphate removal. Thistleton (Thistleton et al. 2002) also showed a similar result of phosphate removal as a function of initial

Table 3 | The experimental data of phosphate removal capacity by coagulation

| Sample No. | Wt. of adsorbent Fe³⁺ (g) | Initial P conc.Ci (μg/L) | Initial Fe:P | Final P conc.Ce (μg/L) | P removal efficiency | Total amount of phosphate adsorbed X₀ (μg) | Amount removal phosphate per unit wt of Fe³⁺, q (μg/g) |
|------------|----------------------------|--------------------------|--------------|-------------------------|---------------------|------------------------------------------|---------------------------------|
| 1          | 0.001                      | 34                       | 16.0         | 1.1                     | 0.967               | 32.8                                     | 32,818.5                        |
| 2          | 0.001                      | 49                       | 11.0         | 1.8                     | 0.964               | 47.0                                     | 46,975.5                        |
| 3          | 0.001                      | 77                       | 7.2          | 2.7                     | 0.965               | 74.3                                     | 74,324.3                        |
| 4          | 0.001                      | 106                      | 5.0          | 3.7                     | 0.965               | 102.0                                    | 101,994.9                       |
| 5          | 0.001                      | 185                      | 3.0          | 18.6                    | 0.899               | 166.4                                    | 166,400                         |
| 6          | 0.001                      | 329                      | 1.7          | 67.0                    | 0.796               | 262.0                                    | 262,000.0                       |
| 7          | 0.001                      | 609                      | 1.0          | 243                     | 0.601               | 366                                      | 366,000                         |
| 8          | 0.001                      | 1,006                    | 0.5          | 605.0                   | 0.399               | 401.0                                    | 401,000.0                       |
Fe:P molar ratio, for wastewater at pH 7. Therefore, there will be two isotherm equations for phosphate removal capacity based on this critical point of Fe:P molar ratio of 5:1.

Three models are mainly used in practice for adsorption isotherms as described in the equations below (Lopez-Ramirez et al. 2003):

Freundlich equation \( q = KC_e^{1/n} \)
Langmuir Equation \( q = bqmC_e/(1 + bC_e) \)
Temkin Equation \( q = A + B\ln C_e \)

Where \( q \) is the equilibrium adsorption capacity (μgP/g); \( C_e \) is the equilibrium concentration of phosphate in aqueous phase (μgP/L); and the rest of the parameters are different isotherm constants which are determined by regression of experimental data. The parameters and correlation coefficients for each model at initial Fe:P molar ration of 5:1 to 16:1 is shown in Table 4. It can be seen that correlation coefficient of Freundich and Langmuir isotherm is higher than Temkin isotherms which indicates data fitting was better with Freundich and Langmuir equation. Therefore, these two models are equally valid to fit experimental data, and Freundich equation is used for theoretical calculation in this paper.

Therefore, Freundlich equation is \( q = 28,425C_e^{0.963} \) for phosphate removal capacity for initial Fe:P molar ratio of 5:1. For initial Fe:P molar ratio of 0.5:1 to 5:1, the Freundlich equation is \( q = 74,023 \times 0.2794 \) (see Figure 2) which obtained by experimental data from Table 5.

Table 4 | Estimated isotherm parameters for phosphate removal by coagulation at initial Fe:P molar ratio of 5:1 to 16:1

| Model       | Parameter Description | Initial Fe:P molar ratio 0.5:1 to 5:1 | Initial Fe:P molar ratio 5:1 to 16:1 |
|-------------|-----------------------|-------------------------------------|-------------------------------------|
| Freundlich  | \( q = KC_e^{1/n} \) | 28,425 0.960 0.995                  | 10^6 0.033 0.993                     |
| Langmuir    | \( q = bqmC_e/(1 + bC_e) \) |                                       |                                     |
| Temkin      | \( q = A + B\ln C_e \) | 20,718 57,670 0.956                 |                                     |

Figure 2 | Coagulation isotherm for varying rang of initial Fe:P molar ratio at pH – 6.
Theoretical calculations of final phosphate concentration of one-step and three-step dose coagulation based on Freundlich equation above

Three-step dose instead of one-step dose is superior to phosphate removal. This hypothesis was based on the mechanisms involved in coagulation of phosphate: predominant mechanism for phosphate removal at pH 6 (a) formation of Fe-hydroxo-phosphate complexes (Fe OH) x (PO4) y (b) these complexes adsorb onto iron hydrolysis species. Three-step dosing is aimed to create three times formation of iron hydrolysis species, which provides more chance for formation of Fe-hydroxo-phosphate complexes (Fe(OH)x(PO4)y) and more external mass transfer for complexes to adsorb onto iron hydrolysis species than one-step dosing. Based on this consideration, this section presents the calculation of phosphate removal of one-step dosing and three-step dosing by isotherm equation above.

Calculation of final phosphate concentration for coagulation with one-step dose

Coagulation was performed at 1mgFe³⁺/L, with a volume of 1 L and m = 1,000 μgFe³⁺/L. Assuming initial phosphate concentration C₀ = 117 μgP/L, results in initial Fe:P molar ratio:

\[
\frac{1000}{56} \cdot \frac{117}{31} = 4.7:1
\]

Hence the equation \( q = 74023C_e^{0.2794} \) can be used for calculation

\[
\frac{(117 - C_e)μgP/L \times 1L}{1mgFe³⁺/L \times 1L \times 10^{-3}} = 74023C_e^{0.2794}
\]

\[ C_e = 4.47μgP/L \]

Therefore, the final theoretical phosphate concentration is 4.47 μgP/L when initial phosphate concentration is 117 μgP/L for one-step dosing.

Calculation of final phosphate concentration for coagulation with three-step dose

Calculation of final phosphate concentration of coagulation with three-step dose is the same way as that with one-step dose. The results of the calculation are shown in the Table 5.

It can be observed that the final theoretical phosphate concentration is 0.43 μgP/L when initial phosphate concentration is 117 μgP/L by three-step dose. Comparing the results, final phosphate concentration of three-step dose i.e. 0.43 μgP/L, is 10 times lower than that with one-step dose i.e. 4.47 μgP/L. Therefore, theoretically phosphate removal of three-step dose is much better than that with one-step dose, at same total iron concentration.

Experimental results of final phosphate concentration of one-step and three-step dose coagulation

The experimental results of final phosphate concentration for one-step and three-step dose are shown in Figure 3. It can be seen that experimental results of final phosphate concentration of one-step dose and three-step dose was different from calculation.

(1) For one-step dose, it can be observed that final phosphate concentration is 4.0 μgP/L which was almost the same as the results from calculation of 4.47 μgP/L.

(2) For three-step dose, final phosphate concentration is 1.0μgP/L which is better than one-step dose of 4.0 μgP/L. This experimental result is similar as calculation, but the final phosphate concentration (1.0 μgP/L) is not as low as the calculation value (0.43 μgP/L). The

| Fe:P molar ratio | First step | Second step | Third step |
|------------------|------------|-------------|------------|
| Applied isotherm | q = 74023C_e^{0.2794} | q = 74023C_e^{0.2794} | q = 28425C_e^{0.9604} |
| Final P concentration | C_{e1} = 42.93μgP/L | C_{e2} = 4.21μgP/L | C_{e3} = 0.43μgP/L |

Table 5 | Final phosphate concentration for coagulation with three-step dose
reason may be equilibrium phosphate concentration formed in first and second step impacted final phosphate concentration because aged flocs with high equilibrium phosphate concentration may release phosphate back into the solution which caused the real phosphate concentration to be higher than theoretical value. Moreover, natural organic matter in seawater may compete with phosphate to adsorb on iron hydrolysis species. And low initial molar ratio of Fe:P in first step results in limiting phosphate removal, which may be also the reason for the discrepancy between the theoretical and experimental results.

CONCLUSIONS

Both theoretical calculation and experimental results show that three-step dosing coagulation is better than one-step dosing. Theoretically, three-step dosing coagulation proved to be 10 times as efficient as one-step dosing coagulation, i.e. from 4.47 μgP/L by one-step dosing coagulation reduced to 0.43 μgP/L by three-step dosing coagulation, for a coagulant concentration of 1mgFe³⁺/L. However, experimental results show that final phosphate concentration is 1 μgP/L. The reasons for this discrepancy between theoretical calculation and experimental results may be the effect of equilibrium phosphate concentration, initial Fe:P molar ratio and NOM competition. Investigation of these reasons is recommended in follow-up study.

Although the experimental value of final phosphate concentration by three-step coagulation is higher than the theoretical value, compared with the one-step coagulation under the same dosage of 1 mg Fe³⁺/L, phosphate removal by three-step coagulation is still better. In other words, if the same phosphate removal efficiency is expected to be achieved, the dosage of coagulant by three-step dose is lower than that by one-step dose, which will play an important role in saving the actual operating cost of pretreatment of seawater RO system.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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