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# System and eco-material design based on slow-release ferrate(VI) combined with ultrasound for ballast water treatment

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**Abstract**
**Background** – The problem of ballast water is caused by microorganisms. The solution to solve this problem is ballast water disinfection. Until the recent day, there have not been many practical solutions. Usually, ozone, chlorine, and UV treatments were used for disinfection. However, these methods still have many weaknesses in treating ballast water.
**Methods** – The ferrate(VI) system proposed in this paper is liquid ferrate(VI) produced on-site using a slow-released system and combined with ultrasound. This paper investigated the optimum time to produce liquid ferrate(VI), pH, and temperature.
**Results** – The optimum synthesis time has been observed to be 10 min with the produced ferrate(VI) of 42,000 ppm. The optimal pH and temperature to make ballast water harmless have been observed to be the neutral pH condition at 25°C, respectively. The design of ballast water treatment using ferrate(VI) combined with ultrasound has also been proposed in this paper.
**Conclusions** – Ferrate(VI), as the primary material in ballast water treatment, has been successfully synthesized. The process of ballast water treatment using ferrate(VI) combined with ultrasound can be performed automatically using a time sensor and a mass sensor. With the proposed design, it can be a promising solution to solve the problems related to ballast water.

**Keywords:** ferrate(VI), ultrasound, ballast water, material design

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## 1 Introduction

Ballast water is fundamental for the secure operation of ships in a marine environment. Administration of counterweight water diminishes the frame push caused by antagonistic ocean conditions or changes in the cargo weight, fuel, and water. Intrusive sea-going species and physico-chemical characteristics of balance water are among the most noteworthy dangers to the world's seas. They can cause amazingly extreme natural, financial and open health impacts. The microorganisms cause the problem of ballast water [1,2]. In order to solve this problem, ballast water disinfection needs to be conducted. In order to provide the best solution, the ballast water treatment should be cost-effective, environmentally friendly, and be able to disinfect the water to an acceptable level. However, there have not been effective disinfection methods for the ballast water. Usually, treatments using ozone, chlorine, and UV were conducted for ballast water disinfection. Ozone needs very high electricity, so the treatment is expensive. Disinfection by chlorine is a dangerous process because chlorine is a toxic compound and should not be released into the environment. Furthermore, the UV disinfection process is limited only to a small scale, and its disinfection efficiency is considered low.

Ballast water treatment needs an environmentally friendly compound as the disinfectant. It is becauseballast water will be released into the sea after the treatment. As a result, one of the best solutions to treat ballast water is the utilization of eco-friendly materials such as ferrate(VI). The ferrate(VI) reaction product is $Fe^{3+}$ which is a harmless compound. Furthermore, ferrate(VI) oxidation and disinfection ability is more excellent than ozone, and it is the strongest among all oxidants/disinfectants used in the application for water and wastewater treatment [3]. Therefore, ferrate(VI) can be an appropriate material to treat ballast water

Today, ferrate(VI) in the market is usually in the form of solid ferrate(VI). This kind of ferrate(VI) is expensive and not stable, so it cannot be stored for an extended period of time. In addition, it is not suitable for ballast water treatment because it will need a large amount of ferrate(VI), so its price should be as low as possible. One of the best solutions is the production of liquid ferrate(VI) on-site. On the other hand, ultra-sonication is one of the valuable methods for treating water treatment, including ballast water [4]. Combining ferrate(VI) with an ultrasound system will enhance ferrate(VI) disinfection efficiency [3–5]. Therefore, the design of ballast water treatment using ferrate(VI) combined with ultrasound has been proposed in this paper.

# 2 Research theory

## 2.1 Ferrate(VI)

Ferrate(VI) is a multi-functional compound that contain a hypervalent species of iron ($Fe^{6+}$). Ferrate(VI) has shown great potential as a multi-purpose water and wastewater treatment chemical for coagulation, disinfection, and oxidation [6–8]. The oxidative power of ferrate(VI) in acidic conditions is the highest among the other oxidants (Table 1), and after oxidation, its produces a non-toxic by-product [9,10].

## 2.2 Ultrasound

Power ultrasound in the 20–100 kHz range is used in chemistry. Ultrasound does not interact directly with molecules to induce the chemical change, as its typical wavelength (in the millimeter range) is too long compared to the molecules. Instead, its energy causes cavitation, which generates extremes of temperature and pressure in the liquid where the reaction happens. Ultrasound also breaks up solids and removes passivating layers of inert material to give a larger surface area for the reaction to occur. Both of these effects speed up the reaction [5,12,13].

**Table 1:** Redox potential for the oxidants/disinfectants used in water and wastewater treatment [11]

| Desinfectant/oxidant | Reaction | $E^0$ (V) |
|---|---|---|
| Chlorine | $Cl_2 + 2e \leftrightarrow Cl^-$ | 1.358 |
| | $ClO^- + H_2O + 2e \leftrightarrow Cl^- + 2OH^-$ | 0.841 |
| Hypochlorite | $HClO + H^+ + 2e \leftrightarrow Cl^- + H_2O$ | 1.482 |
| Chloride dioxide | $ClO_2 + e \leftrightarrow ClO_2^-$ | 0.954 |
| Perchlorate | $ClO_4^- + 8H^+ + 8e \leftrightarrow Cl^- + 4H_2O$ | 1.389 |
| Ozone | $O_3 + 2H^+ + 2e \leftrightarrow O_2 + H_2O$ | 2.076 |
| Hydrogen peroxide | $H_2O_2 + 2H^+ + 2e \leftrightarrow 2H_2O$ | 1.776 |
| Dissolved oxygen | $O_2 + 4H^+ + 4e \leftrightarrow 2H_2O$ | 1.229 |
| Permanganate | $MnO_4^- + 4H^+ + 3e \leftrightarrow MnO_2 + 2H_2O$ | 1.679 |
| | $MnO_4^- + 8H^+ + 5e \leftrightarrow Mn^{2+} + 4H_2O$ | 1.507 |
| Ferrate(VI) | $FeO_4^{2-} + 8H^+ + 3e \leftrightarrow Fe_3^+ + 4H_2O$ | 2.2 |

## 2.3 Ballast water

Ballast water is the water in the ballast tank of the ship. This water is usually stored in the ship for weight, balance, stability, and structural integrity. When a ship unloads cargo, it fills the ballast tanks with water. When the ship loads cargo, it discharges the water out of the ballast tanks. The ballast water causes many ecological problems [14,15]. For example, a ship that contains water from Singapore travels to Miami and empties its ballast tanks. Therefore, microorganisms normally found in Southeast Asia are released in the waters of Miami, Florida [1,2]. Associated damages and costs of controlling aquatic invaders caused by ballast water in the United States are estimated to be $9 billion annually [16,17]. Because of this, an effective method to solve the ballast water problems has to be found.

# 3 Research status

## 3.1 Ferrate(VI) research status

The research on ferrate(VI) usage for contaminant degradation has been well acknowledged. For example, ferrate(VI) effectively oxidized CN and removed heavy metals to low levels without pH adjustment [18]. However, challenges still exist for the implementation of ferrate(VI) technology due to its instability and the high production cost of solid ferrate(VI) products. As a result, research has been directed at generating and applying wet oxidation liquid ferrate(VI). Synthesis of wet oxidation liquid ferrate(VI) can reduce the production cost and overcome the lack of stability.

Recently, ferrate(VI) treatment demonstrated 100% efficiency in disinfecting coliforms, *Escherichia coli*, enterococci, and zooplanktons. An International Maritime

Organization (IMO) Treaty suggests every ship be equipped with ballast water treatment systems. Most recently, in 2012, a full-scale ferrate(VI) treatment system was installed onboard a U.S. flagged container ship. A 100% disinfection efficiency has been obtained with the ferrate doses between 1 and 4 ppm. These doses can be lower if the ferrate(VI) treatment system is combined with ultrasound [19,20].

### 3.2 Ultrasound research status

Ultrasound offers great potential in processing liquids and slurry waste by improving the mixing and chemical reactions in various applications. Ultrasound generates alternating low-pressure and high-pressure waves in liquids, leading to small vacuum bubble formation and violent collapse. This phenomenon is termed cavitation and causes high-speed impinging liquid jets and strong hydrodynamic shear forces. These effects are used for the de-agglomeration and milling of micrometer- and nanometer-sized materials and for the disintegration of cells or the mixing of reactants [13,21]. Furthermore, chemical reactions benefit from the free radicals created by cavitation and the energy input and material transfer through boundary layers. This process can enhance ferrate(VI) disinfection efficiency.

### 3.3 Ballast water treatment status

Ballast water is an essential part of the ship. The ballast water treatment system has attracted the attention of many researchers because it is dynamic and depends on the design of the vessel and the sea conditions in which it works [22–24]. The most common and widely used treatment system is filtration. Filtration is very effective against sediments and some organisms but has the disadvantage of separating tiny microorganisms [25]. Several researchers have also studied UV light and heat treatment. They have several advantages, including easy operation, but have problems of high energy consumption [26,27]. In addition, the use of some chemicals such as chlorine dioxide, as a potent disinfecting agent, has side effects with high levels of possible toxicity [28,29]. Therefore, some researchers also suggest that the ballast water treatment process must be combined [23,24].

## 4 Experimental section

### 4.1 Purpose

This research aims to develop an effective method to treat ballast water based on ferrate(VI). The ferrate(VI) system proposed in this paper is liquid ferrate(VI) produced on-site and using a slow-release system, and it is combined with ultrasound. In this paper, the optimum time to produce liquid ferrate(VI), pH, and temperature were investigated. The design was also proposed in this paper.

### 4.2 Methods

#### 4.2.1 Material design

Liquid ferrate(VI) was produced on-site. Liquid ferrate(VI) used in this experiment was synthesized by the wet oxidation method. Typically, the initial step to synthesizing liquid ferrate(VI) begins with the addition of 31 g of NaOH (Merck) to 60 mL of NaOCl (Junsei Chemical). Afterward, the solution was mixed until a homogeneous solution was formed. After the homogeneous solution was formed, 4 g of $FeCl_3{\cdot}6H_2O$ (Merck) was added to the solution. At last, the ferrate(VI) optimal synthesis time was determined.

#### 4.2.2 Determination of optimal condition

Optimal conditions were needed to obtain the best ferrate(VI) requirement for the ballast water treatment. The determined conditions in this experiment were optimal synthesis time, pH, and temperature. For determining the optimal time, time variations between 9 min and 13 min were examine. Ae UV-Vis instrument was used to determine the liquid ferrate(VI) concentration. For determining optimal pH, the acidic, neutral, and basic conditions were used.

Furthermore, the determination of optimal temperature was conducted at the temperatures of 10, 25, 35, and 45°C. The determination of pH and temperature of ferrate(VI) was observed by the application of ferrate(VI) to degrade 4-BP. This compound was used because it is a very stable and persistent compound and can be used as a parameter to determine pH and temperature conditions. GC-ECD was used for the measurement of the concentration of this compound.

#### 4.2.3 Proposed system to treat ballast water

After all of the optimal conditions have been observed, an effective system to treat ballast water can be developed. The system was designed for a ballast tank capacity of 9.650 $m^3$. The data such as optimal time, pH, and temperature were used to propose the system design. The flow system will be applied in the design. The flow reactor capacity was 0.09722 $m^3/s$ (350 $m^3/h$). Before entering the flow reactor, preliminary filtration to physically filter contaminants was conducted. The ballast water will be pumped into the flow reactor. Then, ferrate(VI) will be slowly added. Simultaneously, then ultrasound was turned on with a power of 66 W. After the process, ballast water, which is already harmless, can be directly released into the sea.

### 4.3 Results

#### 4.3.1 Material design

Ferrate(VI) in this experiment was synthesized by the wet oxidation method. The wet oxidation method involves the oxidation of a ferric-containing solution to form a ferrate(VI) solution under highly alkaline conditions. $FeCl_3{\cdot}6H_2O$ acted as the iron source and NaOCl was the oxidizing agent in this experiment. Then, 11.25 M NaOH was added to optimize the synthesis conditions [30–32]. The wet oxidation method used an oxidizing agent such as NaOCl, $O_3$, and $H_2O_2$ to transform iron (Fe) from a low oxidation state (+3) to the highest oxidation states (+6) [33]. The mixture of NaOCl and NaOH has strong oxidation power and converts yellow $FeCl_3$ into a purple solution. The purplish color indicated the formation of ferrate(VI).

The ferrate(VI) salt established using this method is $Na_2FeO_4$ (sodium ferrate). $Na_2FeO_4$ has different properties from the other forms of ferrate, and it is soluble in an aqueous solution [34–36]. Sodium ferrate has been reported as the oxidation agent for the organic contaminants and disinfecting agents [6]. The formation of ferrate(VI) formation is given as follows:

$$2FeCl_3 + 3NaOCl + 10NaOH \rightarrow 2Na_2FeO_4 + 9NaCl + 5H_2O + 4O_2. \quad (1)$$

Ferrate(VI) was obtained from the conversion of $FeCl_3$ to $FeO_4^{2-}$ with 86.1% of $FeCl_3$ converted. The high concentration of liquid ferrate(VI) was established because the excess amount of NaOCl and NaOH was used as the oxidation agent in the solution.

#### 4.3.2 Determination of optimal condition

Ferrate(VI) synthesis time was considered the critical parameter for determining the optimal condition. High production volume can be acquired with this parameter. This experiment was conducted with an optimum time ranging from 8 min to 12 min, and 10 min was observed as the best time to synthesize liquid ferrate(VI). At the optimum synthesis time (10 min), ferrate(VI) ($FeO_4^{2-}$) was successfully produced with a concentration of ±42,000 ppm. This data was based on the UV-Vis data shown in Figure 1.

The other important parameter to get the maximum degradation ability of ferrate(VI) is pH. Ferrate(VI) is a powerful oxidant in the entire pH range [30], but its oxidation power of ferrate(VI) depends on pH conditions [37]. This experiment was carried out in acidic (pH 3.2), neutral (pH 6.8), and basic (pH 10.2) conditions. Ferrate(VI) had the highest oxidation potential in acidic conditions. However, acidic conditions induced rapid ferrate(VI) decomposition into Fe(III) and oxygen. At the basic condition ($pH > 10$), ferrate(VI) was very stable due to the very slow exchange of the oxygen ligand of ferrate(VI) with water [38]. This causes a decrease in degradation efficiency in acidic and basic conditions. As a result, the highest degradation efficiency was achieved in the neutral condition. The results of the effect of pH on 4-BP degradation are shown in Figure 2

The initial concentration ($C_0$) of 4-BP used in this experiment was 1 ppm with the ferrate(VI) concentration of 0.23 mM. The best degradation process was achieved in the neutral condition with 84.1% of 4-BP degraded. The results also showed that the lowest degradation efficiency occurred in the basic condition. This phenomenon can be caused by the combined effects derived from Fe(VI) speciation

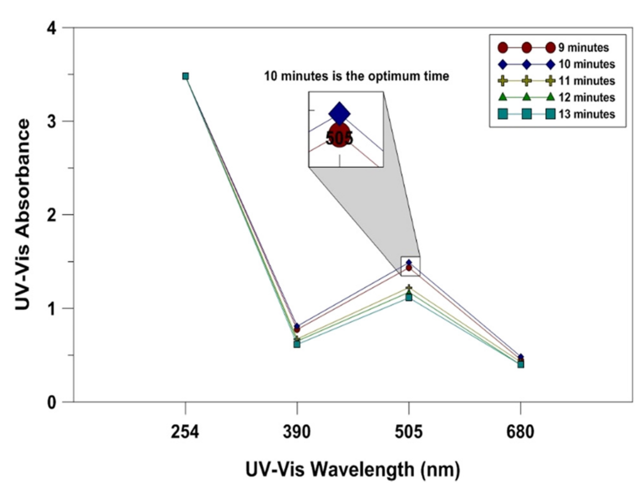


**Figure 1:** Determination of the optimal time.

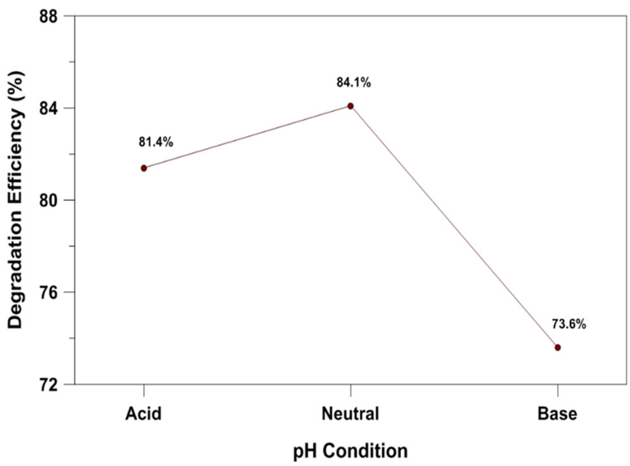


Figure 2: Determination of the optimal pH.

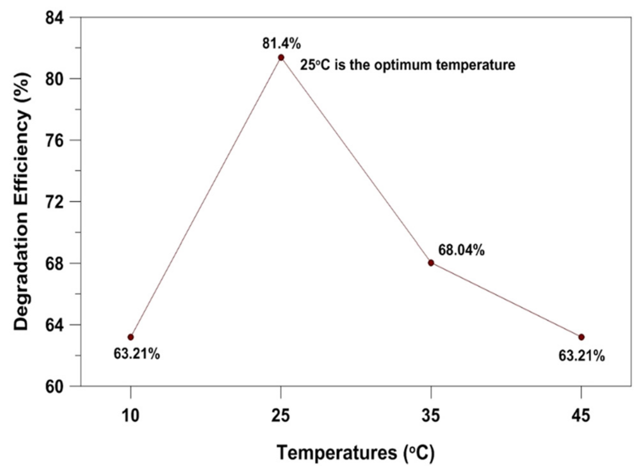


Figure 3: Determination of optimal temperatures.

and halogenated phenolic speciation. Ferrate(VI) is a stronger oxidant, and upon protonation, the reaction rate is expected to increase. Ferrate(VI) is also diprotic acid that have $H_2FeO_4$ [equation (1)], and $HFeO_4^-$ [equation (2)], [34–36] as its compounds of speciation [36]. These two specimens of ferrate(VI) has a high degradation ability and existed in acidic and neutral conditions.

$$H_2FeO_4 = HFeO_4^- + H^+, \text{pKa}, H_2FeO_4 = 3.50, \quad (2)$$

$$HFeO_4^- = FeO_4^{2-} + H^+, \text{pKa}, HFeO_4^- = 7.23. \quad (3)$$

Temperature is one of the important parameters that affects the stability of ferrate(VI). This experimental parameters affect ferrate(VI) degradation ability of 4-bromophenol (4-BP). From that observation data, the optimal temperature condition to degrade 4-BP will be established. The experiment was conducted at temperatures of 10 and 25°C. From that observation data, the optimal temperature condition for BP degradation was 25°C with 81.4% of 4-BP degraded. The results of 4-BP degradation at various temperatures are shown in Figure 3.

The degradation of 4-BP by ferrate(VI) in this experiment was established as a second-order reaction. Homogeneous decomposition kinetics of ferrate(VI) will be enhanced when the temperature increases [6,39]. The maximum degradation efficiency was obtained at 25°C; it was because at 10 ferrate(VI) decomposed rapidly since it was unstable at the high temperature.

### 4.3.3 Proposed system to treat ballast water

All the ferrate(VI) combined ultrasound systems will run automatically. First, the ferrate reactor will be filled with NaOCl. After the required NaOCl in the reactor, the NaOH sensor will be active, and a certain amount of NaOH will be put in the reactor. After that, a bubble mixer will be activated, and a homogeneous solution of NaOCl and NaOH will be made. The mixing process will be carried out for 3 min. After the mixing process, $FeCl_3$ will be added to the reactor. The ferrate(VI) sensor will be activated to detect the formation of ferrate(VI) and determine its concentration based on its mass. Ferrate(VI) will be ready to be added to the flow reactor after 10 min of reaction time. Then, ferrate(VI) produced from this system will be released slowly using the ferrate(VI) time and volume sensors.

In previous research, ferrate(VI) has 100% disinfection efficiency at doses between 1 and 5 mg/L [20,40,41]. Gombos et al. obtained impressive research results and that 5 mg/L ferrates can reduce >99.9% of indigenous bacteria and chlorine-resistant bacteria [19,20]. Since this system used ultrasound to enhance disinfection efficiency, the dose proposed for this treatment was 1 mg/L. With a flow reactor capacity of 0.09722 $m^3/s$ (350 $m^3/h$) and ferrate(VI) production of 42,000 mg/L, the volume needed to treat ballast water in the reactor was determined with the calculation as follows.

$$\begin{aligned} &M_{(\text{Fe(VI)produced})} \times V_{(\text{Fe(VI)needed})} \\ &\quad = M_{(\text{Fe(VI)required})} \times V_{(\text{reactor capacity})} \\ &42{,}000\ \text{mg/L} \times V_{(\text{Fe(VI)needed})} \\ &\quad = 1\ \text{mg/L} \times 350.000\ \text{L}/3{,}600\ \text{s} \\ &V_{(\text{Fe(VI)needed})} \\ &\quad = (350{,}000\ \text{mg}/3{,}600\ \text{s})/(42{,}000\ \text{mg/L}) \\ &V_{(\text{Fe(VI)needed})} = 0.0023\ \text{L/s}\ (8.3\ \text{L/H}). \end{aligned} \quad (4)$$

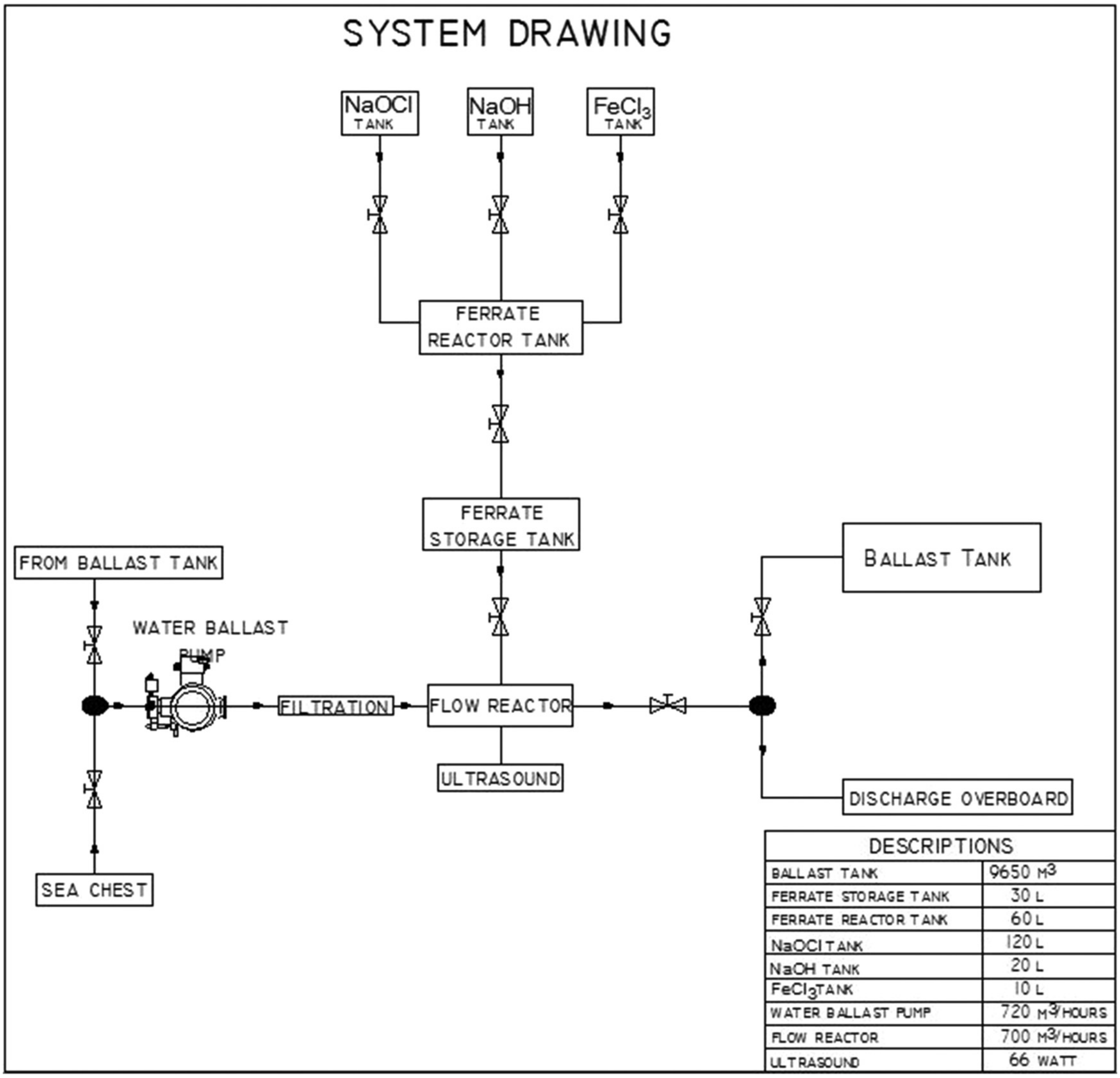


**Figure 4:** Designed system of the ballast treatment.

The ferrate(VI) volume needed to treat ballast water treatment in a 350 $m^3$ flow reactor was 8.3 L. Since liquid ferrate(VI) is not stable, ferrate (VI) will be slowly released. Ferrate(VI) will be added to the flow reactor every 3 min. With 8.3 L of ferrate(VI) needed every hour, the volume of ferrate(VI) added every 3 min was 0.4167 L. Therefore, 0.4167 L of ferrate(VI) synthesis will need 460.98 mL of NaOCl, 30.732 g of $FeCl_3$, and 238.173 g of NaOH. With a ballast tank capacity of 9,650 $m^3$ (ballast water in the ship is 50% capacity volume) and a flow reactor capacity of 0.9722 $m^3/s$ (350 $m^3/h$), the disinfection process will be finished after 49,000 s (13.75 h). The ferrate(VI) volume needed to process all ballast water was 114.42 L. In order to get optimum results, the flow reactor condition had been set to be neutral pH conditions and 25°C temperature. In order to get optimum results, see Figure 4.

## 5 Conclusions

Ferrate(VI) material design has been investigated through the experiment. The materials to synthesize ferrate(VI) have been proposed using $FeCl_3$, NaOH, and NaOCl, which are considered abundant and cheap materials. The optimum synthesis time has been observed to be 10 min, with 42,000 mg/L ferrate(VI) produced. The optimal pH and temperature to make ballast water harmless have been observed to be neutral pH and 25°C. Ferrate(VI) in combination with ultrasound has also been proposed as a treatment method in this paper. The design ran automatically using time, volume, and mass sensors.

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