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Using Ultrasound as a Pretreatment Method for Ultraviolet Disinfection of Wastewaters

(MSc Thesis in Resource Recovery - Sustainable Engineering)

By

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Using Ultrasound as a Pretreatment Method for Ultraviolet Disinfection of Wastewaters

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Preface

This final 30 credit points degree project, is the conclusive part of the Master programme in Resource Recovery- Sustainable Engineering (120 credits) at the University of Borås.

The project was carried out at Professor Farnood's laboratory, department of Chemical Engineering and Applied chemistry, University of Toronto.

This research work has been quite challenging and gave me the opportunity to think independently and to be critically minded.

ABSTRACT

In this study, the effects of neutral particles addition on the breakage of wastewater flocs to improve the efficiency of sonication pretreatment for UV disinfection process have been studied. Kaolin particles as a potentially useful material that is neutral, natural and cheap were added to wastewater samples prior to sonication. Results obtained in this study indicated that hard and small kaolin particles do not have any significant effect on the particle breakage efficiency by ultrasound. The addition of kaolin particles did not significantly increase the cavitation activity (as characterized by potassium iodide actinometry) either. These findings contradict earlier reports that neutral particles can act as nucleation sites and hence enhance cavitation intensity. In this work, sonication of wastewater samples for 60s in the absence of kaolin particles resulted in an approximately one log decrease in the number of surviving bacteria colonies at the tailing level and 1.4 log units increase at the initial slope of coliform removal in UV dose response curve, however addition of kaolin particles prior sonication did not significantly affect the UV dose response curve .The results presented in this study should be treated as preliminary and further detailed investigations are needed to better evaluate this issue.

Keywords: Wastewater treatment; UV disinfection; Ultrasound; Ultraviolet; Sonication; Kaolin; breakage of wastewater flocs

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To my husband, Navid, whom without his immense patience, support, encouragement and love I would never have been capable to accomplish my work.

To my beloved parents, for their constant love and support all the time

To my kind little sister, Arghavan

Thank you all….

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Chapter 1

1. Introduction

Disinfection is the final stage in wastewater treatment plants with the main purpose of killing, inactivating or preventing growth of pathogenic microorganisms that exist in the water and decreasing the spread of probable waterborne diseases caused by municipal drinking water. Lack of proper water disinfection and distribution methods can lead to a wide range of waterborne diseases. Therefore, the disinfection of water should be able to influence a wide range of pathogens while not producing toxic by-products by itself. The severity of disinfection normally depends on the water resource. The public drinking water, which is supplied from ground water resources, are rather clean that through a clean and safe distribution it would not need to be treated in harsh disinfection conditions. However the domestic drinking water which is supplied from wastewater or surface water resources must be disinfected and purified thoroughly. Even the water used for irrigation may have to be disinfected before being used in agricultural lands to avoid accumulation of some contaminates in soil and consequently, in ground water [1].

Wastewater treatment is generally implemented in four stages: preliminary, primary, secondary and tertiary treatment. In preliminary treatment, larger particles that can be problematic for further stages are removed by the means of screening, sedimentation, flocculation, and flotation [2]. In primary treatment, suspended and insoluble materials are usually removed by means of screening, or settling tanks. The effluent from primary stage contains soluble organic materials and fine particles [2]. Secondary treatment includes the biological treatment of wastewater which is the most efficient method for removing organic materials existing in wastewater. In this stage, the organic matters of wastewater are degraded aerobically by certain microorganisms. Microorganisms degrade the organic matters of wastewater in two possible ways, either in form of suspended particles or by growing on another media as a biofilm. In the suspended growth system, both organics and microorganism are presented in suspension. In this process, the organic materials are consumed by the microorganisms that further results in formation and growth of the flocs. Then, solids are settled and separated in the clarifier. The effluent of clarifier has low organic content but still needs to be disinfected to remove microorganisms. In contrary, in the fixed film system, the microorganisms grow on a media and produce biofilm, in this case as the effluent containing organic matter is passing through the media, the microorganisms

consume the organics to grow and the biofilm is formed on the media [2]. As mentioned before, the effluent of secondary treatment step still contains pathogenic microorganisms and has to be disinfected in tertiary treatment with different disinfection method that targets these microorganisms.

Summary of a typical wastewater treatment process is shown in Figure1. 1.

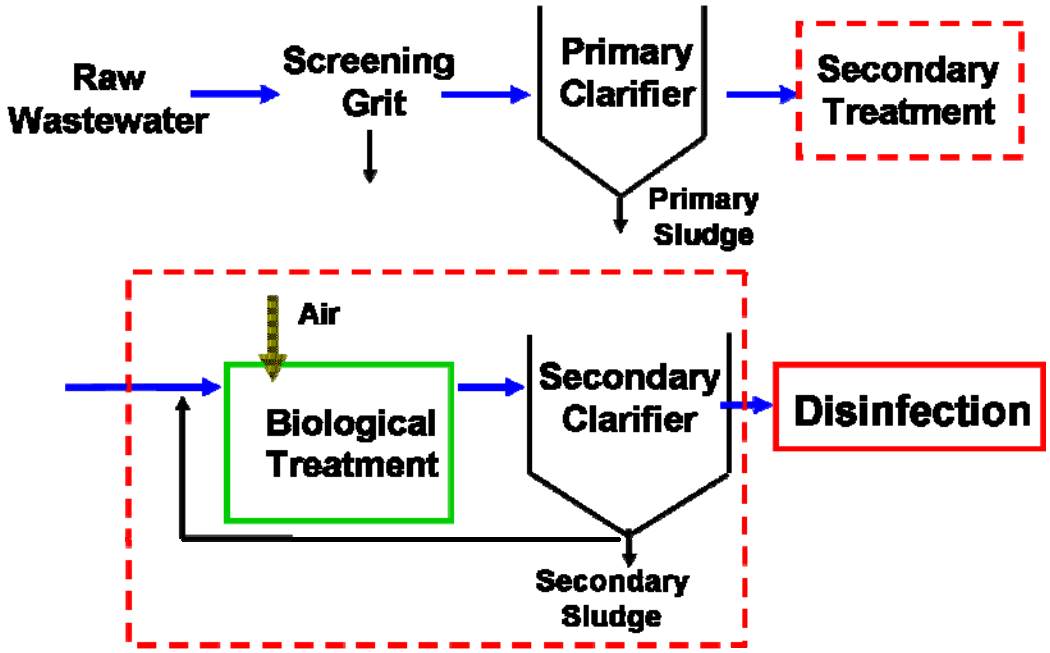

Figure1. 1General schematic of wastewater treatment process using the activated sludge process for secondary treatment .Image modified from Metcalf &Eddy [2] ***internal communication with Yaldah Azimi (2009), Professor Ramin Farnood's research lab, department of Chemical Engineering and applied Chemistry, University of Toronto***

Since this study is focused on disinfection processes and the effects of particles on efficiency of sonication pretreatment. The objectives of this study are given in the following section.

1.1. Activated Sludge process

The activated sludge is a biological treatment process which is first applied in England around one century ago [2]; it is a secondary treatment process in which microorganisms consume the organic materials through wastewater. In this method ,the organics are oxidized aerobically through wastewater in the aeration tank by means of microorganisms and CO_2, H_2O, NH_4 and a newly formed biomass consists of new featured microorganisms are produced [2]. The method

consists of two main reasons: oxidation of biodegradable organics through wastewater, easier separation due to the flocculation of new biomass particles through the effluent. Microbial flocs are defined as a result of particles aggregation during the organics consuming process through the effluent [2].

There are different methods for disinfection of wastewater and purification of drinking water which can be classified into three groups: chemical, physical and photochemical.

1.2. Chemical disinfection of wastewater

Some chemicals have the potential to oxidize and destroy the microorganisms' cell walls. Common chemicals which are used in chemical disinfection of wastewater and purification of municipal drinking water are chlorine (Cl_2), hypochlorite (ClO^-), chloramines (RNHCL), chlorine dioxide (ClO_2), bromine (Br_2) and ozone (O_3). Regarding to their oxidation potential, their effectiveness can be considered respectively: $O_3 \geq Cl_2 > Br_2 > ClO_2 > ClO^- >$ RNHCL [1].

Generally, the above chemicals are quite effective disinfectants, however there are concerns in using these chemical as the disinfectant agents which have to be considered. These chemicals are inherently harmful to human health and to the environment and they result in the production of hazardous by-products. In addition, storage, odor, production process and transportation of these chemicals may pose a threat to the environment and to the wastewater plant operators [1-4, 6]. In particular, chlorination by-products such as trihalomethanes (THM) and haloacetic acid (HAA) are extremely toxic and may cause cancer in human [8, 10, 16]. Furthermore, to address the release of unreacted chlorine, dechlorination is often necessary that is an expensive process [1-4, 6]. Similarly, ozone can also oxidize bromide ions and produces a toxic and hazardous by products [1].

1.3. Physical disinfection of wastewater

Physical disinfection includes some mechanical parts like sedimentation of large materials, screening and filtration. This method cannot be used single handedly for disinfection of wastewater and purification of drinking water; it has to be in the combination with other methods to improve the water disinfection. It can be used as a pretreatment method before other methods [1].

1.4. Photochemical disinfection of wastewater

The most environmentally friendly method introduced for disinfection of wastewater is photo chemical disinfection which includes UV disinfection. It does not produce hazardous by-products as chlorination disinfection does and does not have the risk of escaping ozone through the atmosphere which is happening during the ozone disinfection [1-7, 17, 15]. In the figure 1.2 the different light rays respectively has been shown:

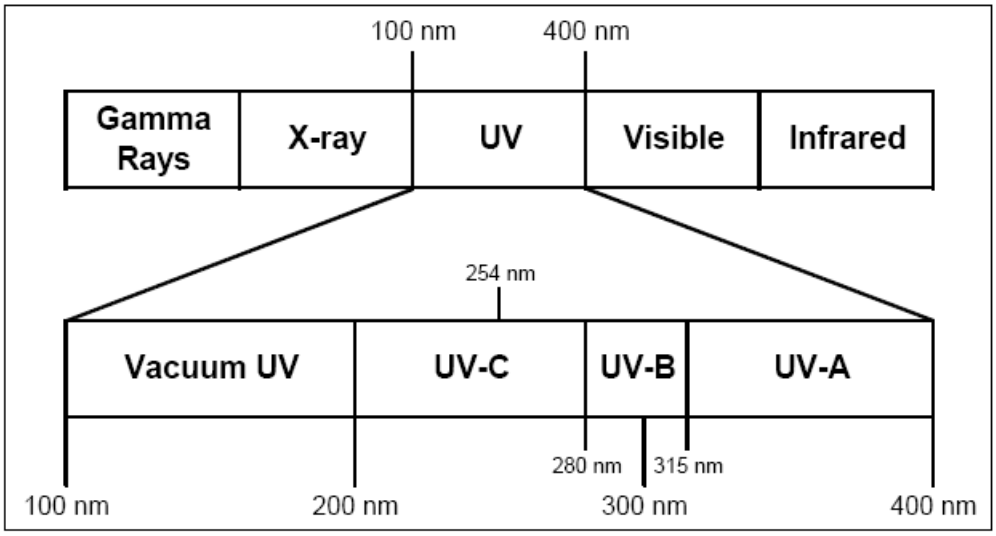

Figure 1.2 Range of electromagnetic waves [13]

To achieve the greater efficiency for wastewater disinfection, a combination of different disinfection methods is usually necessary.

UV light is absorbed by the microorganism's nucleic acid (DNA and RNA) and alters their DNA structure [7, 12, 18]. In this way UV irradiation can break microorganisms' cell wall and stop their growing and reproducibility inside the water; it also works for viruses and spores [6]. UV light also has the ability to produce hydroxyl radicals that can oxidize the cell wall and inactivate microbial microorganisms inside the water [9].

The pathway of UV light through the water is affected by the presence of dissolved organics and suspended particles, in this way the UV light photons may not reach the targeted microorganisms through the water and cannot further do the disinfection. Generally, the presence of suspended particles affect the UV light transmittance through water by scattering and absorbing of UV light, shading the targeted microorganisms and shield other existing pathogenic microorganisms [19-21]. As a result, particle-associated microorganisms may remain active in wastewater even after high UV doses. This phenomenon that is commonly called tailing effect and usually occurs at high UV dosages increases the UV dose demand of the effluent [19, 20].

Previous studies [21-24] have shown that the tailing effect principally occurs due to the presence of large particles in wastewater. Thus, to address the tailing phenomenon and improve UV efficiency, the amount of suspended large particles has to be reduced as much as possible. Ultrasound has been shown to be an effective pretreatment method to break large particles and hence enhance UV disinfection of wastewater [25]. Nevertheless, ultrasound assisted UV disinfection may not be always cost-effective [26, 27].

The efficiency of UV disinfection extremely depends on concentration of microorganisms inside the water, particulate size, UV dose absorbed by the microorganisms and UV transmission through water [11].

Tuziuti *et al.* [26] stated that the addition of particles can increase the yield of sonochemical reactions. Accordingly, in this study the addition of kaolin has been considered to enhance the sonication effect. Chemical actinometry is employed to quantify cavitation activity in order to investigate ultrasonic efficiency under different experimental conditions.

Objectives

The hypothesis of this study is that addition of kaolin particles can be beneficial for the sonication process due to the following possible phenomena:

1. Increasing the cavitation
2. Enhancing particle breakage with ultrasound
3. Increasing the microbial elimination rate and/or decrease the tailing level

The rationale for this research is that if kaolin particles enhance breakage of flocs and microbial elimination rate, decrease the tailing level and hence, the energy requirement of the ultrasound-assisted UV disinfection process may be significantly lowered. Therefore, specific objectives of this thesis are:

1. Evaluating the effectiveness of ultrasound as a pretreatment method for decreasing the level of tailing in UV disinfection
2. Investigating the effect of kaolin on efficiency of ultrasound treatment

Thesis outline

This document is prepared in 6 chapters as follows:

- ***Chapter 1*** provides a brief introduction to wastewater treatment
- ***Chapter 2*** provides background and literature review on ultraviolet light and ultrasound and UV disinfection of wastewater
- ***Chapter 3*** explains the experimental methods
- ***Chapter 4*** represents the results and discussion on the results
- ***Chapter 5*** summarizes briefly the significant findings of the study

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Chapter 2

2. Background

2.1. UV disinfection of wastewater

Approximately 10% of the total sunlight reaches to the earth consists of UV light. The use of UV light for the disinfection of wastewater has been started at the beginning of the 20th century [1]. The first performance of UV disinfection for drinking water was started in Marseille, France in 1906_1909 in large scale and it was used for disinfection of ground water in another city in France, Rouen [1]. During the World War I the improvement of UV disinfection of wastewater has been stopped for a while. In the United States, the implementation of UV disinfection started in 1916 in Henderson, Kentucky [1]. All the UV disinfection implementations for wastewater were stopped during 1930s and the chlorine disinfection became the preferable method again due to its lower costs and easier way to implement. In 1950s UV disinfection of wastewater improved again. Nowadays, in Europe, there are more than 3000 UV disinfection instruments which are being used in different types of water disinfection like supplying municipal potable water and ultra pure water for pharmaceutics and medical industries. In the United States and Canada, the wide implementation of UV disinfection of water is driven by increase in need for wastewater treatment and environmental concerns over disinfection by-products [1, 2].

In 1986 and 1996, there were new discussion about the conjunction of UV disinfection and ozone disinfection together. Nowadays, there are new methods for conjunction of UV with ozone, H_2O_2 and catalysts [1].

In practice, UV light can be generated by an electrical discharge through the mercury vapor lamps. UV light can be absorbed by the microorganisms' nucleic acid (DNA and RNA) [1] and subsequently by destroying their molecular structures and prevent their reproducibility [2]. UV can inactivate bacteria, viruses and spores [5, 6, 7]. UV light has also the ability to produce hydroxyl radicals. Hydroxyl radicals are strong oxidants that can inactivate microorganisms [8]. The efficiency of UV disinfection depends on the concentration of microorganisms, particulate size, UV dose absorbed by the microorganisms and UV transmission through wastewater [9].

However, the destructive effects of UV light may be reversed through the repair mechanism [10, 11].

Although the disinfection of wastewater and purification of drinking water is applied for the decontamination of water, chemical disinfection processes can produce harmful genotoxins. Genotoxins are suspected carcinogen, hence their investigation is extremely vital for protecting the public health. Earlier studies by Haidera et al. [3] found that chlorination and subsequently dechlorination processes produce such hazardous by-products. Similar investigations on UV disinfection of water by the standard low pressure UV lamps (254 nm) shows that the UV disinfection of water is one the best available methods to minimize the production of genotoxins [3, 4].

2.1.1. UV light classification

Regarding to the spectrum of electromagnetic radiation ultraviolet appears with the wavelengths ranging from 100-400 nm (figure 1.2). However, the region between 200-300 nm has the best ability to stop the reproducibility of microbial particles [12, 13]. UV light, specifically around the wavelength of 254 nm can penetrate through the cell wall and get absorbed by cellular material and can prevent the replication of the cells or kill the cells [12, 13].

UV light is divided into three subgroups regarding to their wavelengths, the table below has shown these three subgroups [1, 5]:

Type	Range	Comment
UV-A	From 400 to 315 nm	Between 400 and 300 nm, called near UV
UV-B	From 315 to 280 nm	Called medium UV
UV-C	From 280 to 200 nm	Range to be considered in water disinfection

Table 2.1 UV light subgroups [1]

2.1.2. UV dosage

The UV irradiation energy reaches to surface water with the unit of mJ/cm^2 is called UV dose. It is essential in UV disinfection of wastewater to measure the amount of UV energy that is delivered to the disinfection medium [2].

The microbial inactivation degree depends on the UV dosage received by the microorganism defined by:

UV Dose (mJ/cm^2) = $I \times t$

Where, I is the average UV light irradiation intensity and t is the UV light irradiation exposure time [48].

The UV light intensity is reduced when it passes through the media like water and has to be corrected for UV transmittance of wastewater. UV transmittance indicates the ease of passing UV light through water and water absorbing tendency.

2.1.3. UV Dose Response Curve (UV-DRC)

UV dose response curve is the plot of surviving colony forming units (CFUs) versus UV dose. UV dose response curve usually is presented in a semi-logarithmic form and consists of two parts: a linear initial slope at low UV doses (approximately smaller than 10 mJ/cm^2) corresponding to an exponential decay in CFUs, followed by a near-plateau region at high UV doses (approximately greater than 30 mJ/cm^2) known as the tailing region [14].

2.2. Factors Influencing UV Disinfection

As mentioned before, microorganisms' concentration, particulate size, absorbed UV dose by the microorganisms and UV transmission in the water affect the efficiency of UV disinfection [15]. Table 2.2 indicates the major parameters affecting UV disinfection.

Parameters	Typical values
Percent transmittance (T) or absorbance	35-65
Total suspended solids (TSS) (mg/l)	5-10
Particle size (µm)	10-40
Iron (mg/l)	Less than 0.3
Hardness (mg/l)	Less than 300
Flow rate or hydraulics	-

Table 2.2 Key parameters affecting UV disinfection and their typical values [17]

The key wastewater parameter in UV disinfection is the UV transmittance or UVT. UVT indicates the ease of passing UV light through the solution and furthermore the UV demands for the different effluents [16]. Since 254 nm is the most effective wavelength for microbial inactivating, UV transmittance is usually measured by an UV spectrometer operating at the wavelength of 254 nm [5, 6, 8]. In this wavelength the UV transmittance percentage relating to the distilled water is set at 100%. A low UV transmittance shows that a lesser amount of the UV light can reach the targeted microorganisms, and hence lower disinfection efficiency is obtained.

Dissolved particles through water can affect the UV transmittance adversely due to their UV absorption characteristics. The existence of suspended particles and dissolved chemical compounds which can absorb UV light such as iron can affect the UV light transmittance. The particles can decrease the efficiency of UV disinfection by absorbing or scattering the UV light, or protecting the microorganisms from exposure to UV light.

Figure 2.1 indicates the effect of particles larger than 8 microns on the UV dose response curve for filtered and unfiltered effluent [17].

Qualls *et al.* [18] have obtained similar results which indicate removing the larger particles can increase the level of microbial inactivation. From their work, it can be concluded that the adverse effects of UV disinfection on larger particles may occur due to the presence of more resistant coliforms in bigger size particles.

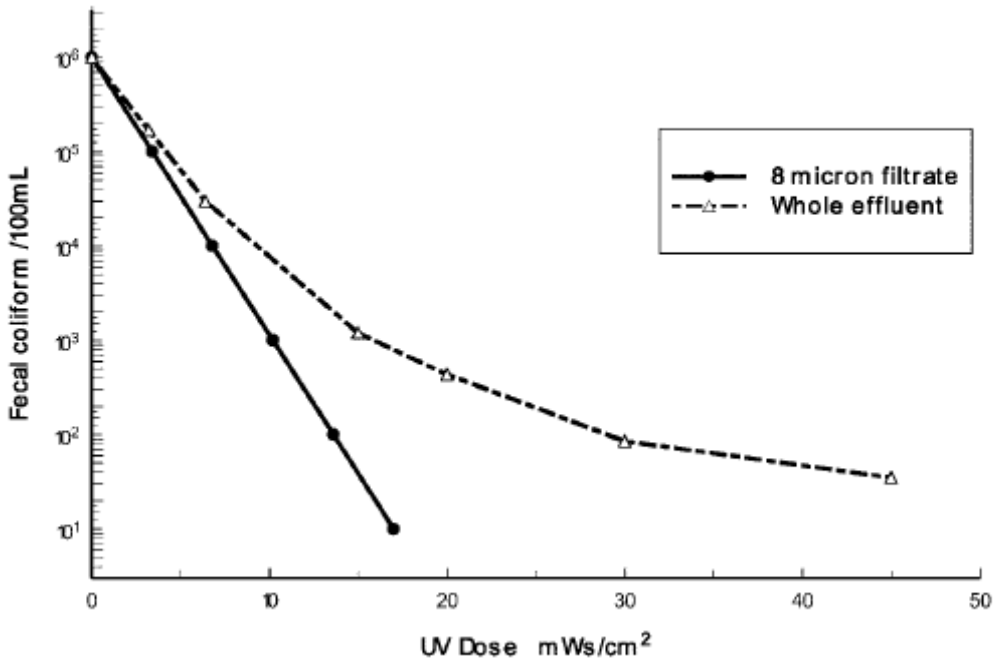


Figure 2.1Typical UV dose response for filtered and unfiltered wastewater [17] similar results established by Quals et al. [18] and Tan [19]

2.3. UV Absorbance and Scattering of Microbial Flocs

As mentioned before while UV light irradiates to the solution containing solid particles, it may be absorbed, scattered, or passed through the solid materials. Figure 2.2 represents the possible incomplete penetration of UV light into wastewater particles.

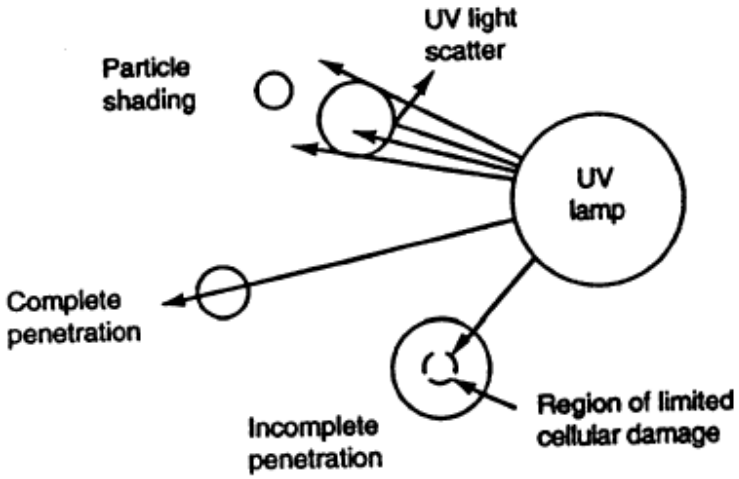


Figure2. 2 Schematic showing possible interactions between UV light and wastewater particles [20]

2.4. UV light penetration into wastewater particles

Loge *et al.* (1999) [20, 21] has reported that ultraviolet light can be highly absorbed by wastewater particles; but it can still inactivate the microorganisms by penetrating to some extent through their materials. Since wastewater particles such as activated sludge particles are highly porous [22] it was suggested that as microbial flocs highly absorb UV light it can only penetrate through particles porosity not through the solid material.

2.5. Tailing Phenomenon

Tailing phenomenon usually occurs at high UV dosages due to the presence of microbial flocs, which may absorb or scatter UV light photons during their pathway through water or provide shielding for the other microorganisms and prevent UV light reaching them[17, 23]. In this phenomenon a quantity of the microorganisms are still active through water even after high UV light exposure time. However, tailing also occurs in chemical disinfection of wastewater where an amount of bacteria can survive due to the incomplete penetration of chemical agent into the suspended particles [24, 25].

Tailing phenomenon is illustrated in Figure 2.3; the figure indicates how the rate of microbial inactivation decreases at higher dosages in the tailing phenomenon.

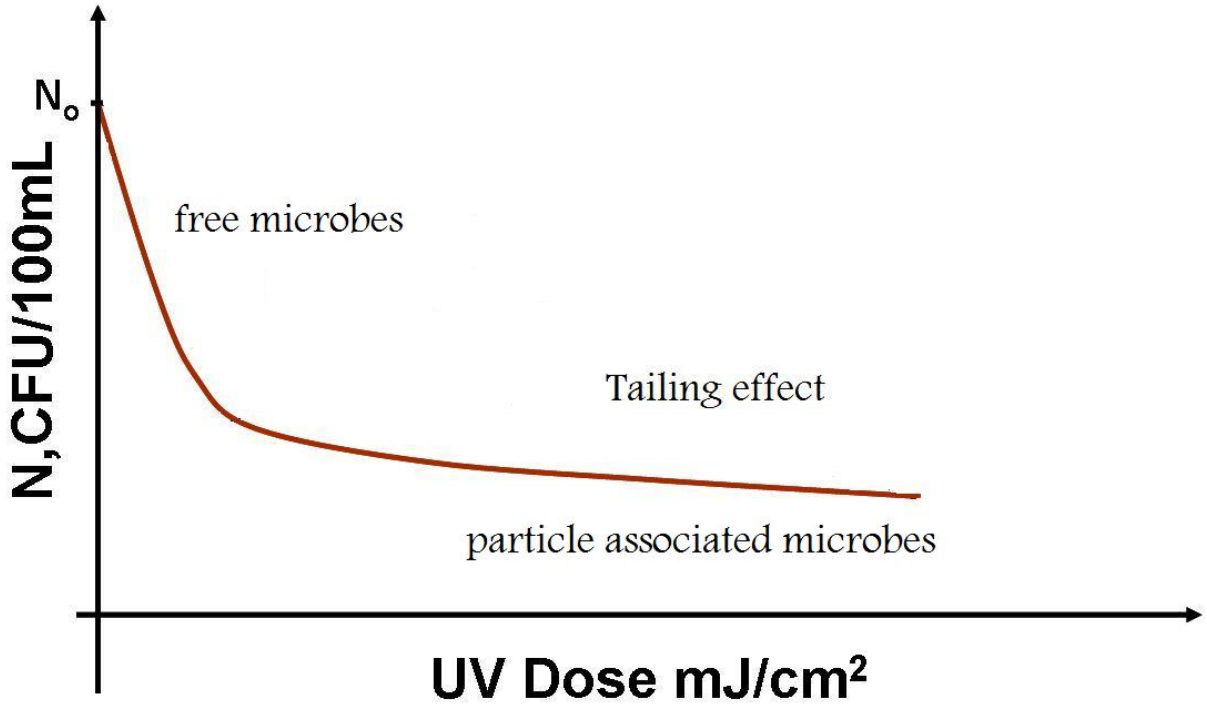


Figure 2.3 Illustration of a typical UV dose response curve, tailing at higher dosages can be seen

There have been a number of methods suggested for decreasing the degree of tailing. Qualls *et al.* (1985) [26] and Das (2001) declared that by filtration of effluent approximately through 8-10 microns filters as an upstream process before UV disinfection tailing effect will be reduced [17,26]. Blume (2004) [27] implied the use of ultrasound as an upstream process to reduce the size of suspended particles and hence improve the efficiency of UV disinfection.
It has been mentioned in many studies that particle size affect on tailing degree and subsequently on efficiency of UV disinfection [26, 28-30].

Madge *et al.* [30] implied that particles size can obstruct UV disinfection and reduce the UV disinfection efficiency, they concluded that the effluents containing small particles can be

disinfected by UV light faster than the ones including large particles. However, in their study the particle size did not exceed 20 μm. Tan [19] studied the effect of particle size on UV disinfection of microbial flocs through activated sludge process. In his study, to obtain various particle size fractions sieving method was done; it is concluded that particles greater than size ranges of 45-53 microns are mostly responsible for tailing effect in UV dose response curve, and since large particles are UV resistant particles ,effluent containing large particles indicates more resistance against UV light [19].

2.6. Modeling of UV Disinfection Performance

Microbial response varies for different microorganisms in various effluents; it represents the probability of microbial survival in the presence of UV light irradiation and indicates the pathogenic microorganisms' concentration before and after decontamination. A number of models have been developed for describing and predicting UV disinfection performance through effluents. Table 2.3 shows a summary of several theoretical models that have been published in the literatures.

Model	Equation	Reference
One-hit	$\frac{N}{N_0} = e^{-kD}$	**[31]**
Multi-target	$\frac{N}{N_0} = 1-(1-e^{-kD})^m$	**[31]**
Multi-hit	$\frac{N}{N_0} = e^{-kD}\sum_{i=0}^{m-1}\frac{(kD)^i}{i!}$	**[31]**
Double -exponential	$\frac{N}{N_0} = (1-\beta)e^{-k_1 D} + \beta e^{-k_2 D}$	**[33]**
Modified two population	$\frac{N}{N_0} = (1-\beta)\left(1-(1-e^{-k_1 D})^m\right) + \beta e^{-k_2 D}$	**[33]**

Cairns et al.	$\frac{N}{N_0} = (1-\beta)e^{-k_1 D} + \sum \beta_r e^{-kT_\mu^{\ r} D}$	**[34]**
Emerick et al.	$\frac{N}{N_0} = (1-\beta)e^{-kD} + \frac{\beta}{kD}(1-e^{-kD})$	**[35]**

Table2. 3 Theoretical models for describing UV disinfection performance

The simplest and the most common model describing UV light performance is single exponential model which assumes that a single hit can cause microorganism inactivation [31]. In this case the probability of survival will correspond to a first order kinetics [2, 32]:

$$\frac{N}{N_0} = e^{-k_0 D}$$

Where N is the number of survived bacteria at a determinate dosage and N_0 is the number of bacteria at dosage zero when there is no decontamination by UV light yet, k_0 is the inactivation constant. In this model, it has to be considered that when the microorganisms are associated with the solid particles subsequently the received dosage is less in comparison with the condition they are particulate-free. However this model cannot explain the effect of particulate matter associated to the microorganisms. Figure 2.4 indicates the single exponential model (one hit model) at the presence of particulate-free microbes.

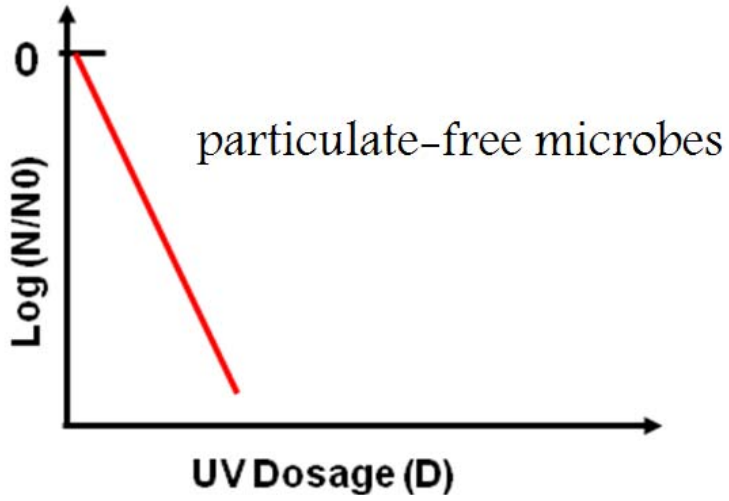


Figure 2.4 Illustration of a typical single exponential model in presence of particulate-free microbes

Another simple mathematical equation introduced for the UV-DRC of wastewater is the double exponential model [14, 29, and 33]. In this model, two coliform subgroups are respectively considered as "UV susceptible coliforms" and "UV resistant coliforms" through wastewater. The first one is a group of coliforms which are not associated with the particles (free microbes) or just associated with small suspended particles that are readily disinfected. These coliforms are simply disinfected at low UV doses, and the second group contains coliforms which are associated with large particles suspended through wastewater. As the suspended particles can work like a shield for the coliforms and protect them against UV light they usually need higher UV dose to be disinfected. This model is represented by:

$$N/N_o = (1-\beta)e^{-k_1 D} + \beta\, e^{-k_2 D}$$

Where,

N = the number of surviving coliforms after UV light irradiation at a specific UV dosage which is considered as number of CFU (Colony Formation Unit),

N_o = the initial coliforms number or CFU before UV irradiation typically called Dose 0,

k_1 = constant of UV-susceptible coliforms inactivation rate,

k_2 = constant of UV-resistant coliforms inactivation rate,

β = the division of UV resistant coliforms to the total initial coliforms [14, 29, 33].

Figure 2.5 illustrates typically the double exponential model (double hit model) at the presence of particulate-free microbes and particle associated coliforms.

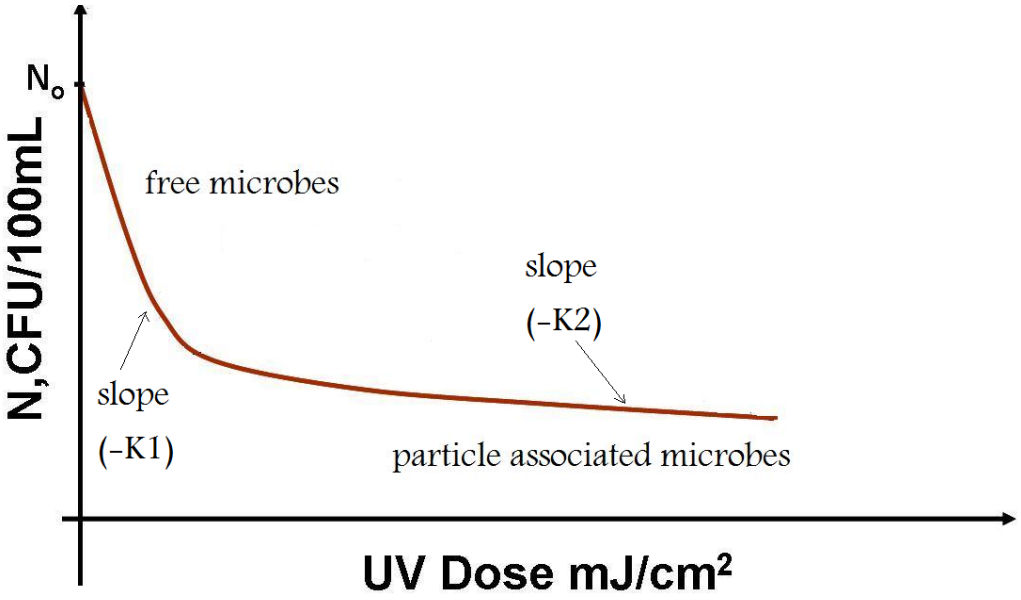


Figure 2.5 diagram of a typical double exponential model

2.7. Different types of UV lamps

Several commercially available sources of UV light are listed below [2]:

1. Mercury vapor lamps (low, medium and high pressure)
2. Low-pressure high-output mercury vapor lamps (LPHO)
3. Electrode-less mercury vapor lamps
4. Metal halide lamps
5. Xenon lamps (pulsed UV)
6. Eximer lamps
7. UV lasers
8. Light emitting diodes (LED)

Among the above sources of UV light, mercury vapor lamps are the most common for UV disinfection [5, 6, 8]

2.7.1. Conventional UV lamps (mercury vapor lamps)

The first UV lamps (mercury vapor lamps) were manufactured by Hewitt in 1901[1]. These lamps work in different pressure of mercury vapor [1, 5]:

1. Low pressure mercury lamps: they work at pressure ranges of 100-1000 Pa
2. Medium pressure mercury lamps: they work at pressure ranges of 10-30 kPa
3. High pressure mercury lamps: they work at pressure ranges up to 10 atm

Normally, for UV disinfection of wastewater the low and medium pressure lamps are used [1, 5].

2.7.2. Light emitting diodes (LED)

UV mercury vapor lamps have a short life (approximately one year). As mercury is a hazardous material, it is preferable to replace this kind of mercury UV lamps by new ones which do not have hazardous characteristics; mercury vapor lamps energy consumption is high and produces hazardous wastes. UV solid-state light emitting diode (LED) is a new type of UV disinfection instruments. UV-LED is a semiconductor device and emits light in a narrow spectrum, UV-LED lamps have a longer life and their electricity consumption is lower than mercury vapor lamps, their efficiency is higher than mercury vapor lamps [6]. They are usually manufactured in wavelength range of 370-400 nm (UV-A) [5]. However, they have found limited applications in wastewater applications were a large UV dose (tens of mW/cm^2) has to be delivered in a short period of time (in the order of several seconds) to flowing wastewater (typically millions of gallons per day).

2.8. Disadvantages of UV disinfection

Microorganisms which are damaged during UV irradiation might be repaired by cell repair mechanisms. For instance, during transportation or distribution of treated water, damaged microorganisms get enough time to be regenerated and repaired**.** Microbial repair may increase the UV dose demand of effluent but it does not change the result [2, 36-39].

2.8.1. Microbial repair in UV disinfection

Microbial repair is an enzymatic reaction that leads to DNA repairing of microorganisms. Microbial repair consists of photo reactivation and dark repair. Photo reactivation needs light for repairing the cells. To avoid this phenomenon treated water can be simply kept away from light after disinfection. Dark repair phenomenon is not as significant as the photo reactivation. Dark repair is concerned to some microorganisms repairing which does not require light for repairing

but it can also happen in the presence of light. It usually occurs during water distribution through pump lines due to growth of biofilm in pump lines [2, 38, and 40].

Kashimada *et al.* [40] have studied the bacteriostatic effects of UV disinfection for effluents, they reported that survival microorganisms concentration is significantly low just after implementation of UV disinfection nevertheless the concentration of microorganism grows over after a while; the research claims that the result of UV disinfection is much better for drinking water in comparison with UV disinfection of effluents.

Although UV disinfection of wastewater is an efficient way, sometimes using the chemical disinfectant is necessary during the UV implementation. UV is not as efficient as chlorination for inactivation of viruses; chlorination is sometimes required for removing the algal sedimentation of materials, besides the oxidation of some substances should be done with the chemical disinfectant [2].

2.9. Effect of temperature and pH on UV microbial response

Effect of Temperature and pH on UV microbial response extremely depends on the microorganisms types; temperature has a minimum effect on UV microbial response, in pH=6-9, microbial response is independent to the pH [2].

2.10. Implementation of UV/O_3

In some cases, UV disinfection of water does not work separately; this happens when some resistant compounds exists through the water, UV cannot destroy these compounds, like N-Nitrosodimethylamine (NDMA), which are toxic and cause cancer in human body. These kinds of materials must be removed from drinking water because of their intensive effects on human body; UV disinfection degrades these compounds to dimethylamine (DMA). The problem is that the degraded product (DMA) produces NDMA again by the regeneration after degradation; in this case combination of UV and ozone is applicable. DMA has the tendency to react with the hydroxyl radicals (oxidation by ozone), so it produces methylamine as final product and the concentration of DMA decreases inside the water [15, 41].

2.11. Ultrasound as a pretreatment process

Using ultrasound as a pretreatment prior UV disinfection of wastewater due to improving UV light disinfection efficiency was studied first by Oliver and Cosgrove in 1975 [42].

In their study, secondary effluent was applied as the targeted wastewater sample; the effluent was sonicated via a 20 kHz, 300-watt ultrasound device for 5 minutes. By using this method, they observed a considerable enhancement in the UV disinfection of wastewater. Blume and Neis (2003 and 2004) have repeated the same experiments via 10s using ultrasound [27,43], Joyce *et.al* (2006) [44] studied effect of using ultrasound as a pretreatment for UV and also electrolysis disinfection and reported that using ultrasound prior these disinfection methods were considerably more effective than using these disinfection methods single handedly.

Yong *et al.* (2009) [14] investigated the effect of sonication as a pretreatment on UV disinfection kinetics of primary effluent and concluded that sonication improved the UV light disinfection performance. In their study the double-exponential model was considered as the representative equation to describe UV light performance. In their study, it is proved that by increasing the sonication time the initial inactivation rate increased and the tailing level in the dose-response curve decreased. They considered particles larger than 60 μm are mostly responsible for occurring tailing phenomenon; it is described in their study as sonication reduces the amount of large particles and generates a great amount of small particles through wastewater sample the UV transmittance usually decreases after sonication and this could occur due to the UV light absorption or scattering by a large amount of small particles through samples.

2.11.1. Cavitation

The main mechanism of sonication is based on the cavitation phenomenon which includes the whole procedure of creation, expansion and collapsing of microbubbles throughout liquid phase when negative pressure is applied to the medium during sonication [14, 45, and 46].

Microbubble collapsing typically produces high temperature and pressure condition locally throughout the liquid phase; however the whole liquid mass stays at ambient conditions. This collapsing of microbubbles can produce other physical and chemical changes. Some changes can be achieved through the liquid bulk caused by microbubble collapsing are creation of radicals ,

generation of shock waves and local acoustic micro streaming . These can generate a great shearing force inside the liquid bulk which can mix and break particles [14, 45, and 46]. Figures 3.1 and 3.2 typically indicate the procedures of microbubble collapsing due to cavitation.

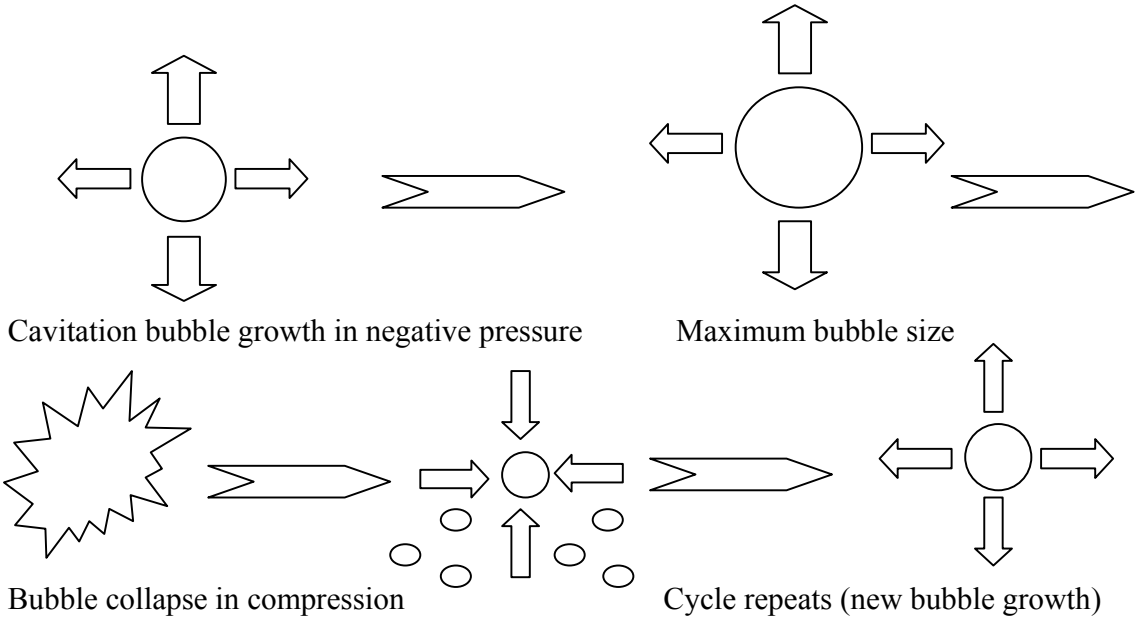


Figure 2.6Microbubbles collapsing procedures due to cavitation based on http://www.variclean.nl/Ultrasoon/theorie.php [Accessed November 5, 2010]

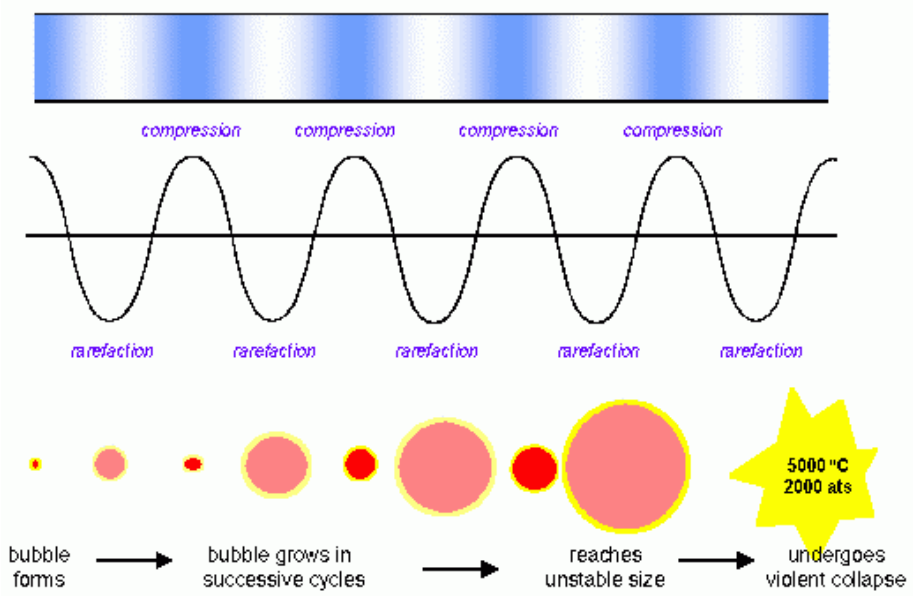


Figure2.7 Microbubbles collapsing procedures due to cavitation ***source:*** **http://www.deafwhale.com/stranded_whale/barotrauma.htm [Accessed November 5, 2010]**

2.11.2. Sono-chemical Effect

Sonochemical reactions are recognized as such chemical reactions in which the violent collapse of cavitation bubbles created by intense sonication generates oxidants such as hydroxyl radicals and hydrogen peroxide in liquid bulk [47].

There are different methods to evaluate the acoustic cavitation effects such as hydrophone, thermo electrical, iodine dosimetry, Frick dosimetry, terephthalate dosimetry, phenolphthalein dosimetry, porphyrin dosimetry, aluminium foil erosion and degradation of polymer chains [48].

2.11.3. Iodine Dosimetry

In this study the Iodine Dosimetry (Chemical Actinometry) was considered to evaluate the cavitation effects, this method is based on the fact that sonication through the water generates Hydroxyl radicals and subsequently Hydrogen Peroxide (H_2O_2) which can quickly react with the Iodine ion (I^-) to liberate I_2 [45,48-50], the amount of iodine indicates the sonochemical cavitation efficiency . The Iodine amount is measured by UV spectrometer at wavelength of 350 nm, concerning to the reactions below the concentration of I_3^- is measured by spectrometer which is equal to Hydrogen Peroxide concentration. In this case H_2O_2 concentration is calculated based

on the Beer-Lambert law, it implied that by increasing of H_2O_2 concentration the absorbance is increased. Hence, in order to increase the efficiency of this kind of chemical reactions, generating a great amount of cavitation bubbles through the liquid bulk seems necessary. Particle addition with the proper size and amount is a suitable suggested technique to increase the amount of microbubbles generated by sonication; particles due to their surface roughness characteristics and by providing a greater surface area can supply nucleation sites for cavitation microbubbles [47].

The reactions occurring during sonication through water:

$H_2O \rightarrow OH^\circ + H^\circ$ $\quad 2OH^\circ \rightarrow H_2O_2$ **During sonication through water**

$2I^- + H_2O_2 \rightarrow I_2 + 2OH^-$ **In the cuvette**

$I_2 + I^- \rightarrow I_3^-$

$A = \varepsilon LC$ **Beer-Lambert Law**

A: absorbance
L: length of solution the light passes through
C: concentration of solution
ε: Molar absorption coefficient

2.11.4. Effect of particle addition on sonication efficiency

Tuziuti *et al.* [47] studied the effect of size and amount of alumina(Al_2O_3) addition on sonication efficiency during 60 s by two different methods: measurement of I_3^- absorbance and measurement of acoustic noise; they have reported that sonication yield increases by alumina particles addition just under the amount of 20 mg of alumina. It has been concluded that the sound transmission decreases through the solution due to higher amount of alumina addition, subsequently they set the particles amount on the highest suggested amount (20 mg) and it has been reported that just the particles with the mean diameter larger than 10 μm affect the

sonication yield. The possible reason that the smaller particles does not affect the sonication yield may concern to their light weight that they can easily travel with the liquid bulk altogether and cannot provide the condition for bubbles collapsing. Advantages of neutral particles addition on sonication has been observed by the use of ultrasound combined with TiO_2 by Torres *et al*. [51] and silica particles by Suri *et al*. [52] for the degradation of organic pollutants.

Chapter 3

3. Experimental methods

3.1. Sample Collection

In this study, wastewater samples were collected from Ash Bridges' Bay municipal wastewater treatment plant that is located at the eastern region of Toronto, Canada. The plant is capable of treating 818000 m^3 of water per day, and includes an activated sludge biological treatment unit in its secondary treatment. Treated effluent is disinfected with chlorine before discharging into the Lake Ontario. Mixed liquor samples were collected from the aeration tank before discharging into the secondary clarifier.

Secondary effluents were also collected at the end of the secondary clarifier, right before the point that effluent is channelled to be disinfected. In order to ensure that the storage does not change sample characteristics, the samples were taken and processed freshly.

3.2. Sieving

In order to deal with samples with a consistent particle sizes, the collected mixed liquor samples were passed through the sieve trays (U.S.A. Standard Testing Sieve) and collected between two sieves with opening sizes of 32 and 150 µm. After this, obtained fraction sizes were collected on the sieve with the size of 32 µm. These particle fractions were gently washed with distilled water for at least 15 minutes to make sure all particles smaller than 32 µm were washed away. The remaining larger particles were then collected off the sieve. The sample was then suspended in deionized water and used for particle size distribution analysis and sonication test.

3.3. Particle Size Distribution Analysis

Particle size distribution analysis was carried out using a Multisizer 3.0 particle size analyzer set with a 280µm aperture tube (Beckman Coulter Canada, Mississauga, Ontario, Canada). Samples were diluted with a solution of NaCl with a concentration of 9.7 g/L in order to get a proper concentration and then analyzed to evaluate the particles size distribution. It has to be mentioned that the Multisizer operates based on Coulter principal, which means the multisizer only indicates the size of solid fraction in a porous particle (solid volume). In this case, the realistic

particle sizes are greater than the reported ones by the equipment [53]. In this study, the various particle sizes have been mentioned refer to their apparent sizes calculated from sieve openings.

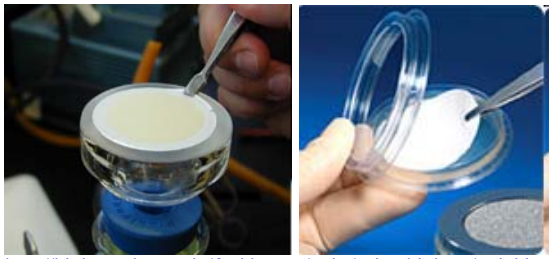
Figure 3.1Multisizer 3.0 particle analyzer

Yuan (2007) [54] reported the relationship between the realistic particle size and their solid volume size (Coulter) for the same equipment.

$$D = 0.82\, d^{1.24}$$

Where D is the actual wastewater particle sizes according to the sieve opening and d is the Coulter particle size measurement which is determined by the multisizer.

3.4. UV Bioassay



http://biology.clc.uc.edu/fankhauser/Labs/Microbiology/Drinking_Water/14_remove_membrane_fr_platform_P8141458.jpg&imgrefurl

[Accessed November 5, 2010]

In this study a low-pressure mercury vapor UV lamp (Trojan Technologies, London, Ontario, Canada) has been used which approximately 85% of its UV light irradiation is at a wavelength of 253.7 nm [55]. This UV light system consists of a horizontal stainless steel case where two UV lamps have been located inside, subsequently a black vertically downwards collimated tube with the size of 22cm in length and 9cm in diameter has been located which provides a uniform UV irradiation.

UV incident intensity (I) is measured at the center of the solution surface in mW/cm^2 by means of a calibrated IL radiometer with a SED240 sensor and a NS254 filter (International Light, Newburyport, MA, USA) [48]. The UV exposure time for each UV dosage is specified by a spreadsheet which is developed by Bolton *et al.* [56]. The spreadsheet calculates the UV exposure time based on intensity and UV absorption at 254nm. However there are some correction factors which can also interfere the UV exposure time for each UV dosage, such as the Reflection Factor, Petri factor, Water Factor, and Divergence Factor [56]. The UV absorptions are measured by Lambda 35 UV/Vis spectrometer (Perkin Elmer, Wellesley, MA, USA, Wellesley, MA, USA) at the wavelength of 254nm.

In this study, sample was poured in a 20 mL volume Petri dish with diameter size of 4.8 cm, during the UV irradiation time sample was constantly stirred with a magnetic stirrer within the Petri dish .Samples were received different UV dosage ranges between 0 and 60 mJ/cm^2. Then the disinfection degree was evaluated through the number of surviving fecal coliform units after UV irradiation at a definite dose. In order to count the number of surviving fecal coliforms the membrane filtration method was used by means of sterile filters (Millipore sterile 0.45µm) and for rinsing the particles on the filter, a buffer solution contains of KH_2PO_4 (13.6 g/L) at pH 7.2 was used [57].

After filtration a number of surviving fecal coliforms remained on the sterile filter were cultured on the m-FC agar plate (VWR, Mississauga, Ontario), then the cultured media was incubated at a temperature of 45ºC for approximately 24± 2 hours, after incubation time the colony formation units (CFUs) were counted.

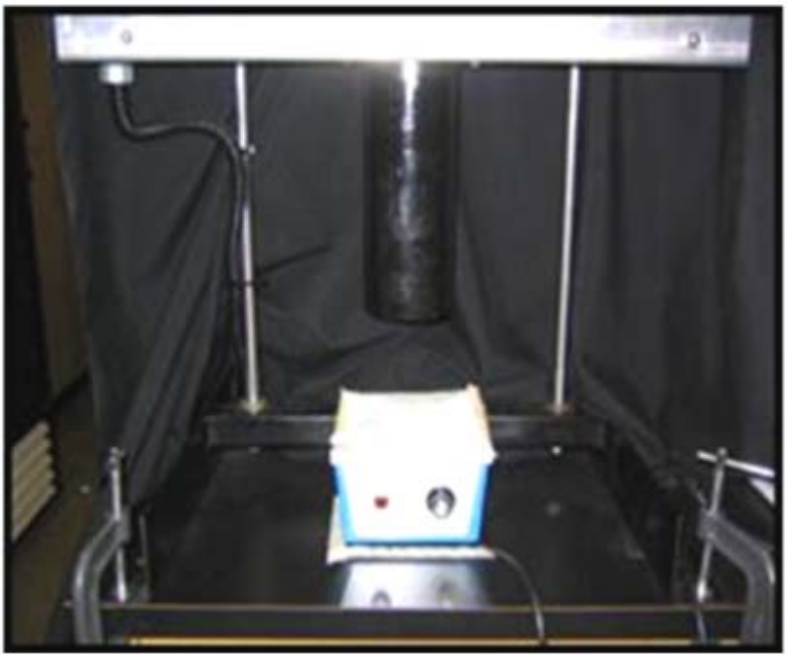


Figure 3.2Low-pressure mercury vapor UV lamp

3.5. Sonication

The utilized ultrasound instrument (Advanced Sonics Processing Systems, Oxford, USA) is a conventional reactor consists of an acrylic cylinder reaction chamber in 10.8cm diameter and 25cm height, water-cooled, magneto restrictive which receives the maximum electrical power of 600 W. For each experiment, 1 L of wastewater was sonicated in the reactor at 300Wand 20 kHz frequency initially at room temperature (22±1°C).

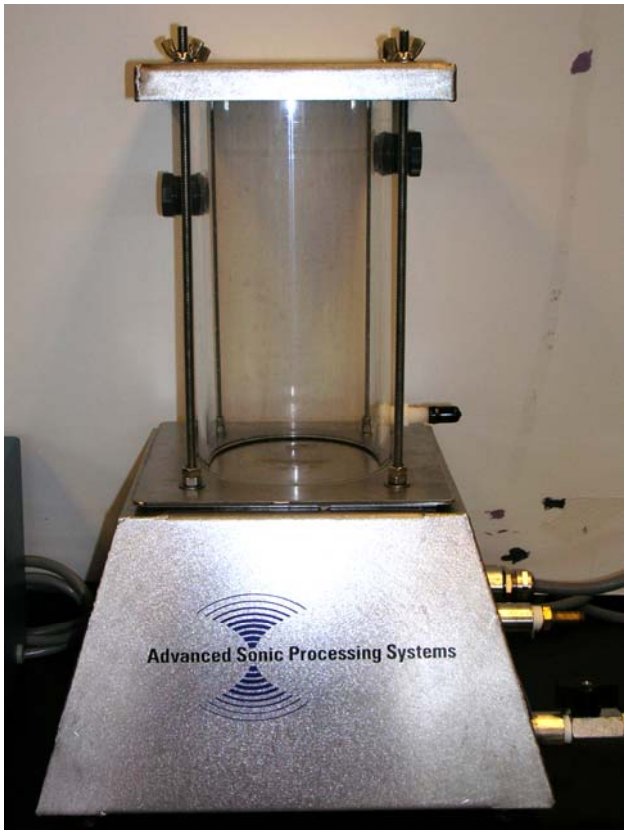


Figure 3.3 Ultrasound reactor

3.6. Experimental Procedure

3.6.1. UV dose response curve (UV-RDC)

To investigate the effect of kaolin particles addition on the sonication and subsequently, on the UV dose response curve, effluent was sieved between two sieves with opening sizes of 32 and 150 µm and the collected particles were diluted to obtain a suspension consists of approximately 10000 particles per liter. Each sample was treated in three ways:

1- The control test: disinfection of the wastewater sample with no sonication
2- Sample was sonicated for 60 s at 300 W power and 20 KHz frequency, and then subjected to UV light for disinfection.
3- Kaolin (Kentucky-Tennessee Clay Company) with the average size of 5µm was added and homogenized in the test solution before the sonication pretreatment and then exposed to UV light for disinfection.

3.6.2.Chemical Actinometry test (Iodine Dosimetry)

In this study, 400 mL of sample containing distilled water and various amounts of kaolin (0, 10 and 100 mg/L) was sonicated for 6 minutes. Solutions of KI and ammonium molybdate were utilized to measure the effect of kaolin addition on the sono-chemical effects of ultrasound. Samples were collected from ultrasound reactor chamber every 2 minutes and filtered by syringe filter (0.2 μm, VWR , Mississauga, Ontario) to remove all the kaolin particles within the sample, then 0.5 ml of KI solution (0.1 M) and 20 μl of ammonium molybdate (0.01 M) were added to 2 ml of filtered sample in UV cuvette . Several experiments were carried out to optimize the sonication time, various volume fractions of samples and chemicals for this test .The amount of produced iodine was measured by Lambda 35 UV/Vis Spectrometer (Perkin Elmer, Wellesley, MA, USA, Wellesley, MA, USA) at wavelength of 350 nm, concerning to the reactions below the concentration of I_3^- is measured by spectrometer that is equal to hydrogen peroxide concentration. In this case, H_2O_2 concentration is calculated based on the Beer-Lambert law, it is concluded that the absorbance increases by formation of H_2O_2.

$H_2O \rightarrow OH^\circ + H^\circ$ $\quad 2OH^\circ \rightarrow H_2O_2$ **During sonication through water**

$2I^- + H_2O_2 \rightarrow I_2 + 2OH^-$ **In the cuvette**

$I_2 + I^- \rightarrow I_3^-$

In this study ammonium molybdate is used as the catalyst for the chemical reactions. Regarding the significant sensitivity of chemical reactions to the temperature, this parameter was controlled constantly by thermometer during sonication to avoid the considerable effect of temperature increasing on formation of H_2O_2.

3.6.3. Particle Size Fractionation

1 L of diluted mixed liquor sample was passed through sieves with the opening sizes of 32 and 150 μm and then collected on the sieve with the size of 32 μm, after that it was sonicated in

absence and presence of kaolin particles (100 mg) for 60 s. The sample was then used for particle size distribution analysis.

Chapter 4

4. Results and Discussion

4.1. Effect of Sonication on Particle Size distribution

4.1.1. Effect of sonication on particle size distribution of activated sludge flocs in mixed liquor sample

Figure 4.1 and 4.2 illustrate the breakage effect of sonication on large particles, they both indicate the reduction in the amount of large particles and increasing in the amount of small particles due to sonication. These figures indicate that sonication breaks wastewater flocs into smaller sizes. In this case UV disinfection would be more efficient after sonication. Similar results were obtained by Yong [48] in 2007.

Figure 4.1 shows the breakage of particles based on number percentage. Figure 4.2 indicates the same effect based on quantity of particles (number).For example in figure 4.1, approximately 1% of the whole effluent sample (mixed liquor sample) contains particles in size of 30 μm that is corresponding to around 260 particles in the given size in figures 4.2 and 4.3, subsequently it can be observed in figure 4.1, approximately less than 0.2% of the whole effluent sample consists of particles in size of 30 μm after sonication that corresponds to 20 particles in the same size in figures 4.2 and 4.3. The reduction in the amount of particles due to sonication indicates its significant capability to break wastewater flocs.

The cut-off at 8 μm in the figures happens due to the detection limit of the particle size analyzer. However, since large particles are mostly responsible for the tailing effect [19], the particles smaller than 8 μm are not expected to cause any effect on the results.

Figure 4.3 illustrates the same phenomenon begins at particles size of 20 μm to focus on breakage of particles greater than 20 μm, the figure indicates a significant reduction in the amount of particles greater than 20 μm due to sonication.

The three figures show a significant effect of sonication pretreatment prior UV disinfection to break wastewater flocs and consequently make them more amenable to UV disinfection.

The reduction percentage of large particles can be calculated from:

$$b = 100 \times (1 - \frac{N_{Ps}}{N_{P0}})$$

Where,

$N_{Po=}$ number of large particles/volume before sonication;

N_{Ps} = number of large particles/volume after sonication.

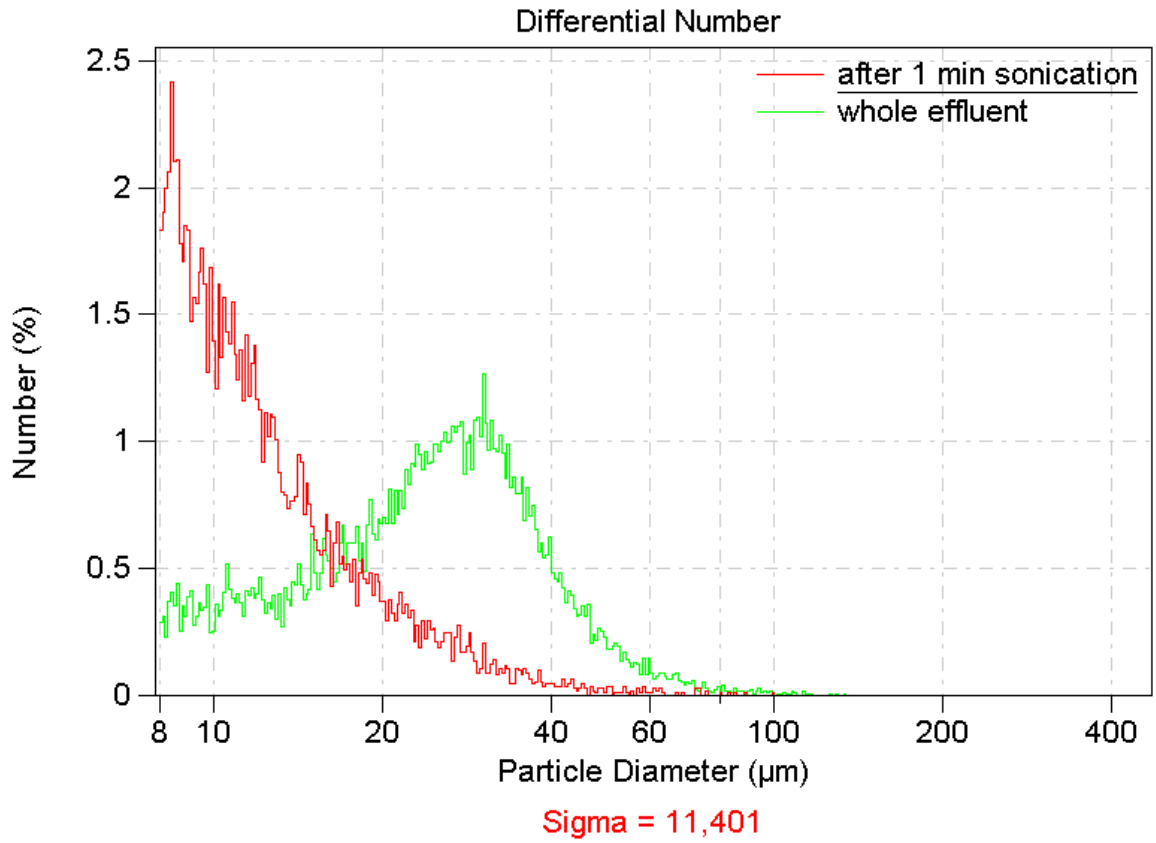


Figure4.1 Effect of 1 min sonication on particle braekage in mixed liqour samples (number%)

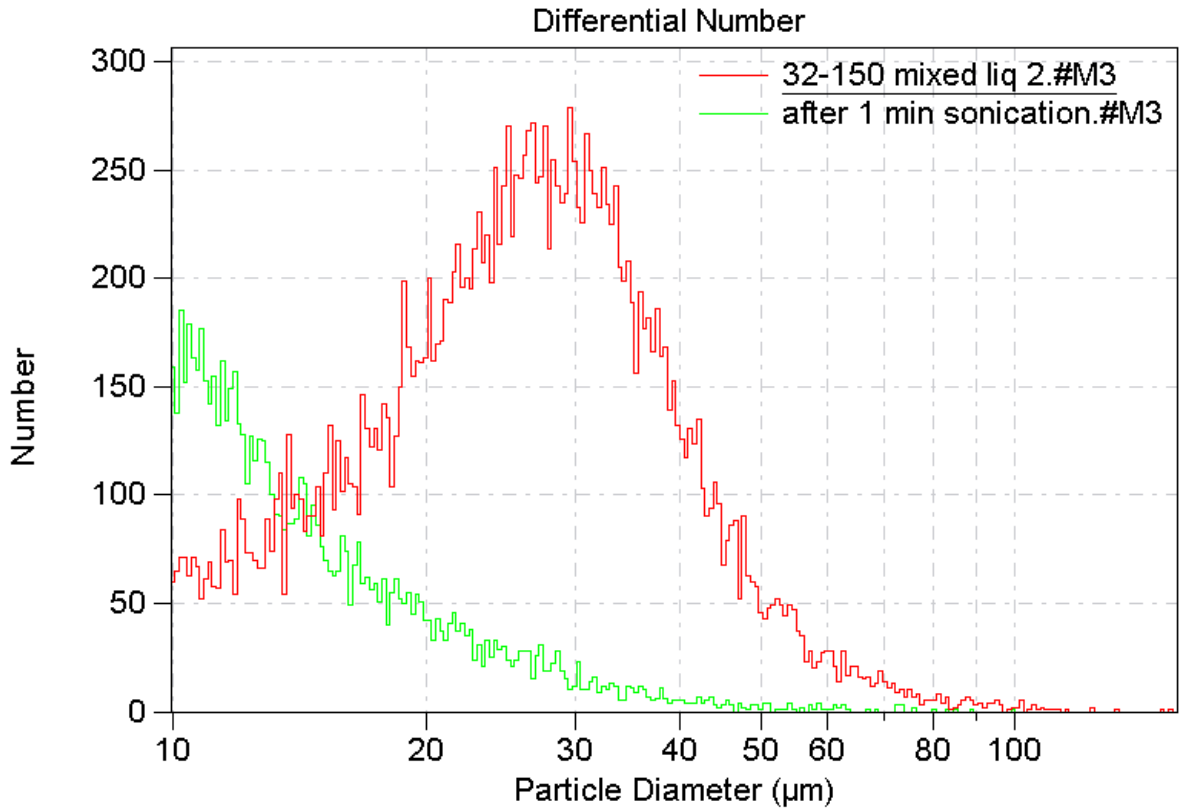


Figure 4.2Effect of sonication on particle breakage in mixed liqour samples (number)

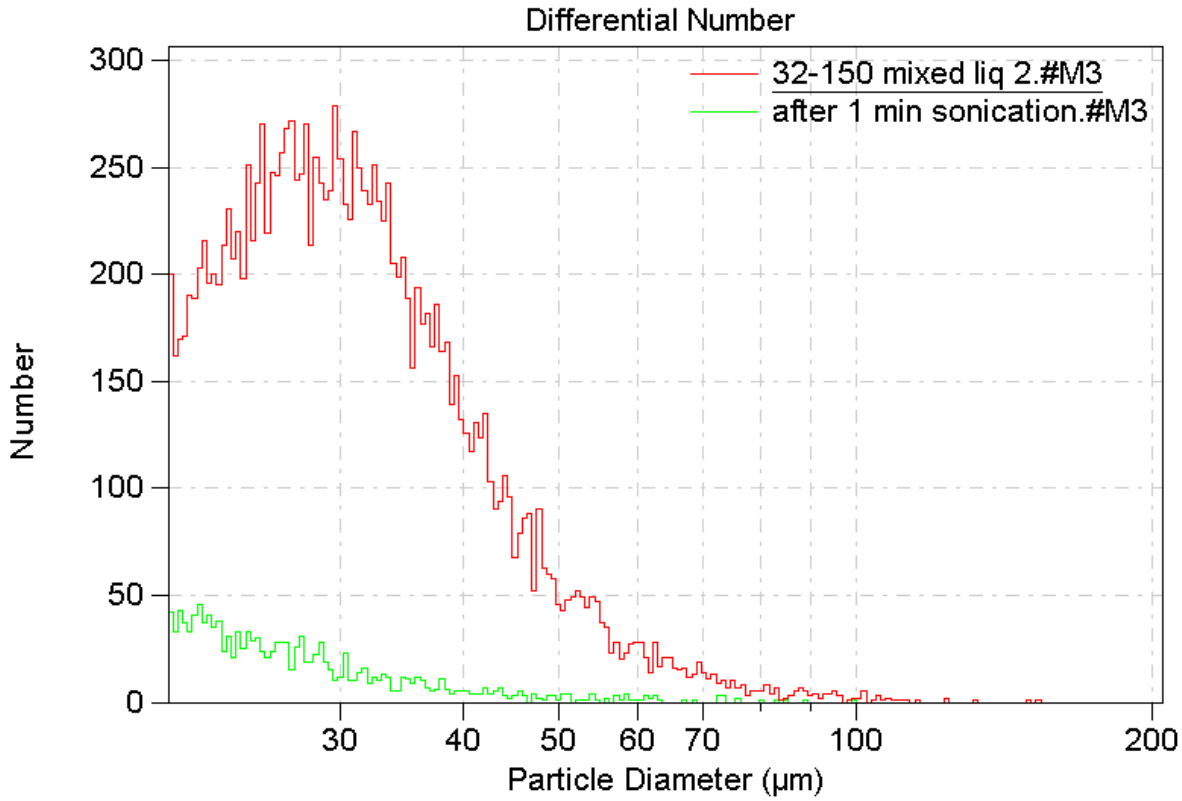


Figure 4.3Effect of sonication on particle breakage with the cut off at 20 µm to consider larger particles breakage

4.1.2. Effect of sonication on particle size distribution of activated sludge flocs in secondary effluent

Given that large particles are mostly responsible for the tailing effect this work is primarily focused on the breakage of large particles into smaller ones. Since secondary effluent is collected at the end of the secondary clarifier, it does not contain plenty of large particles.

Figure 4.4 indicates size distribution of activated sludge flocs and the effect of sonication on breakage of particles in secondary effluent. This figure shows that there is little effect after 1 minute sonication on the particle size distribution of activated sludge effluents.

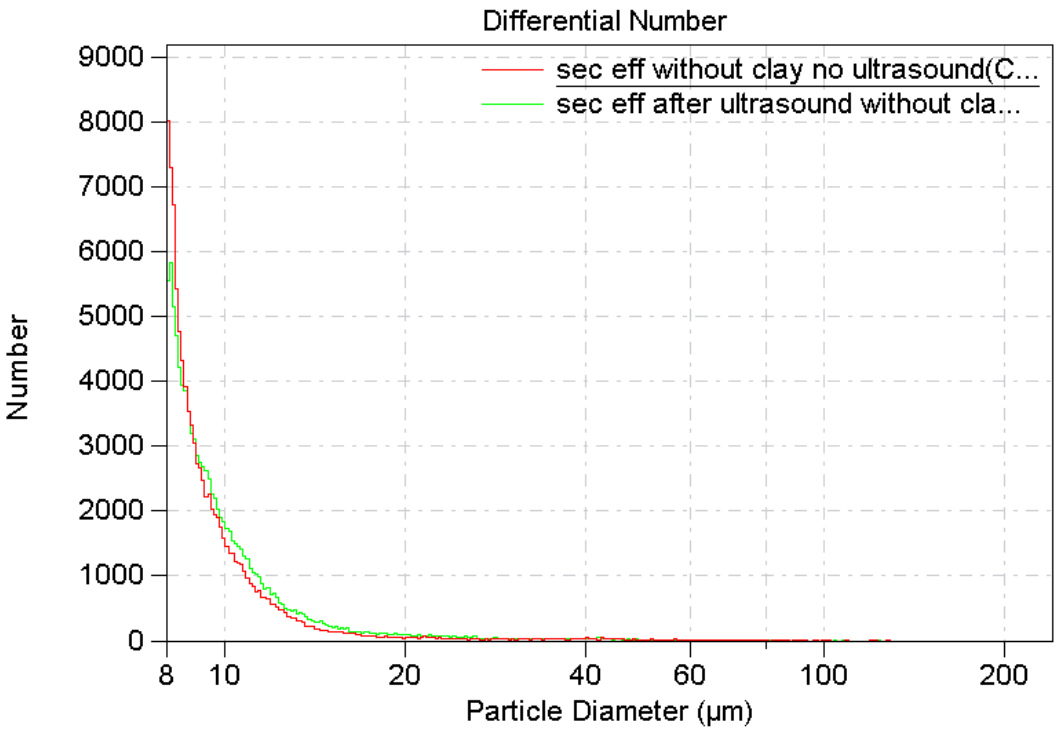


Figure 4.4 Effect of 1 minute sonication on breakage of secondary effluent particles

4.2. Effect of kaolin addition on sonication particle breakage

4.2.1. Effect of kaolin addition on breakage of activated sludge flocs in mixed liquor by sonication

Figure 4.5 illustrates the effect of kaolin addition on breakage of large particles; the great amounts of small particles in size ranges of 8-10 μm indicates the amount of kaolin particles with the mean diameter of 5 microns.

Figure 4.6 indicates the particle size distribution with the cut off at 20 μm to consider large particles breakage.

Figures 4.5 and 4.6 indicate breakage of activated sludge flocs due to sonication in similar appearance to figures 4.2 and 4.3. However the effect of addition of kaolin particles on breakage of wastewater flocs is rarely clear in the figures. Regarding figure 4.6, kaolin particles do not significantly affect the breakage of activated sludge flocs in wastewater samples.

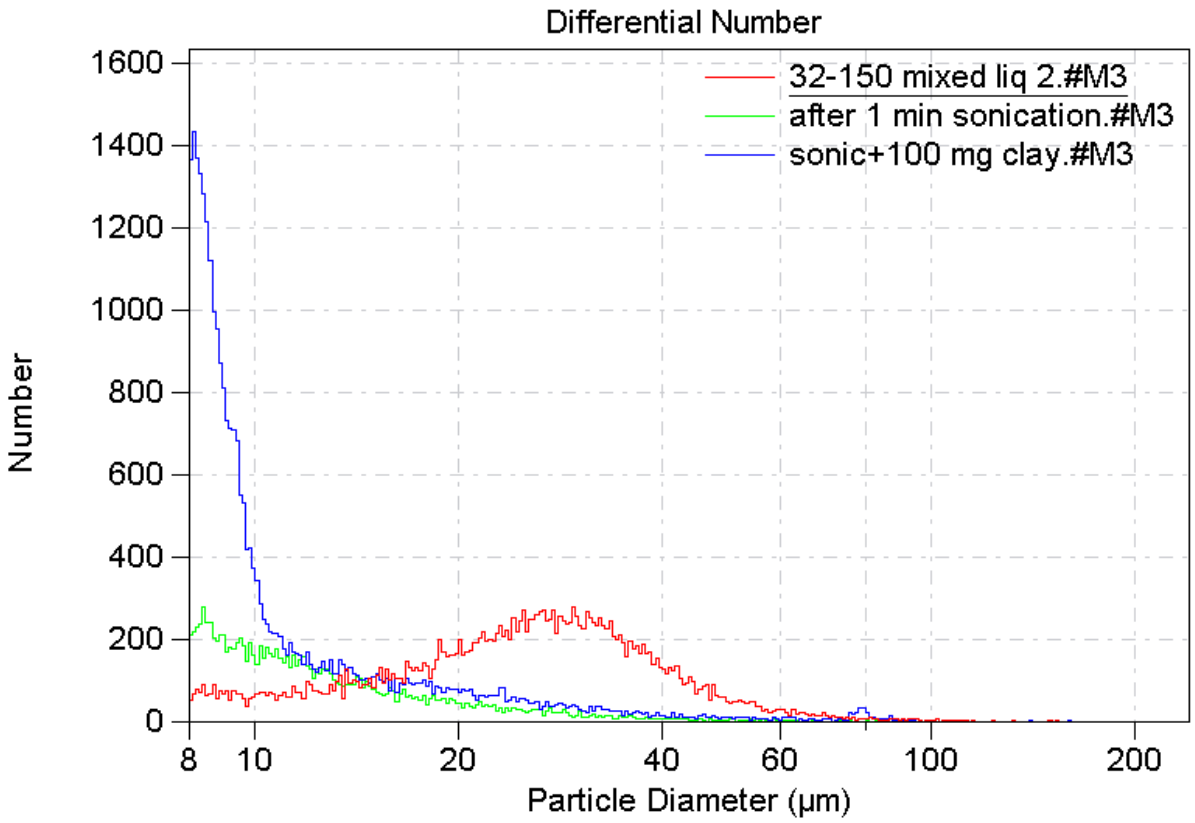


Figure 4.5Effect of 1oo mg/L kaolin addition on sonication particle breakage

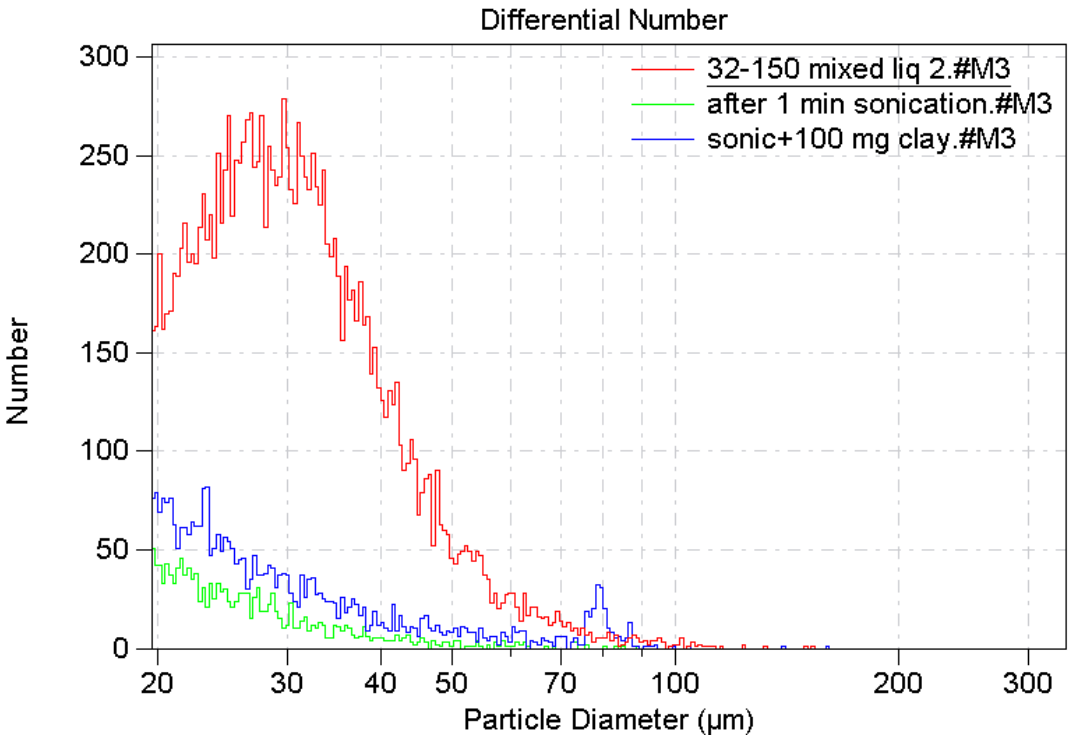


Figure 4.6 Effect of 100 mg/L kaolin addition on 1 minute sonication

4.3. Effect of sonication on the breakage of kaolin particles

Kaolin pareticles themselves may be broken during the sonicaiton process. In order to cosider the effect of sonication on the breakage of kaolin particles, 100 mg of kaolin was dissolved in 1 L of distilled water and homogenized before sonication, then the solution was sonicated in ultarsound reactor with 300 W power and 20 KHz frequency for 1 and 4 minutes respectively. Following this step, the particle size distribution was analyzed to indicate the kaolin particles breakage.

Figure 4.7 illustrates the effect of sonication on breakage of additional kaolin particle. Based on this figure, there is no evidence of the breakage of kaolin particles after 60 s sonication. However, increasing the sonication time to 4 minutes shows a detectable reduction in the concentration of large particles.

Figure 4.8 and 4.9 illustrate the effect of 4 minutes sonication on the breakage of kaolin particles; they show that after 4 minutes of sonication through the solution the kaolin particles would break. Regarding to the figures 4.7 and 4.8, sonication is capable to break kaolin particles. As a result in this study, to avoid the breakage of kaolin particles in a solution of wastewater flocs and kaolin particles, sonication time did not exceed 60 s.

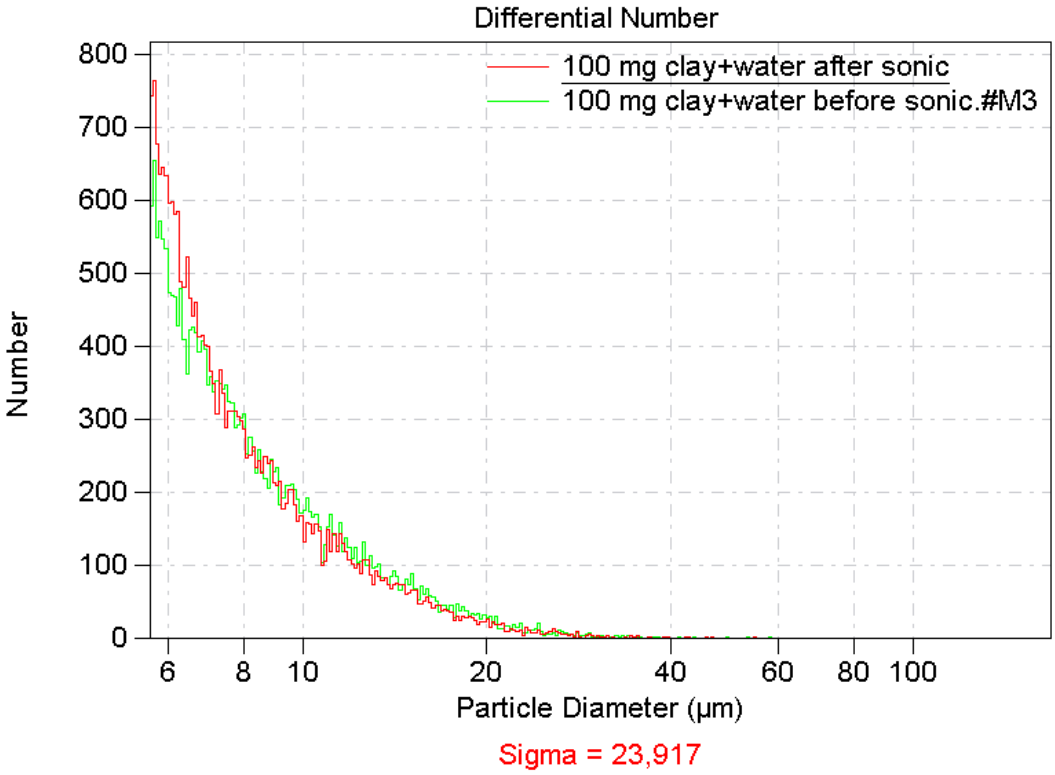


Figure 4.7Effect of 1 minute sonication on kaolin particles breakage

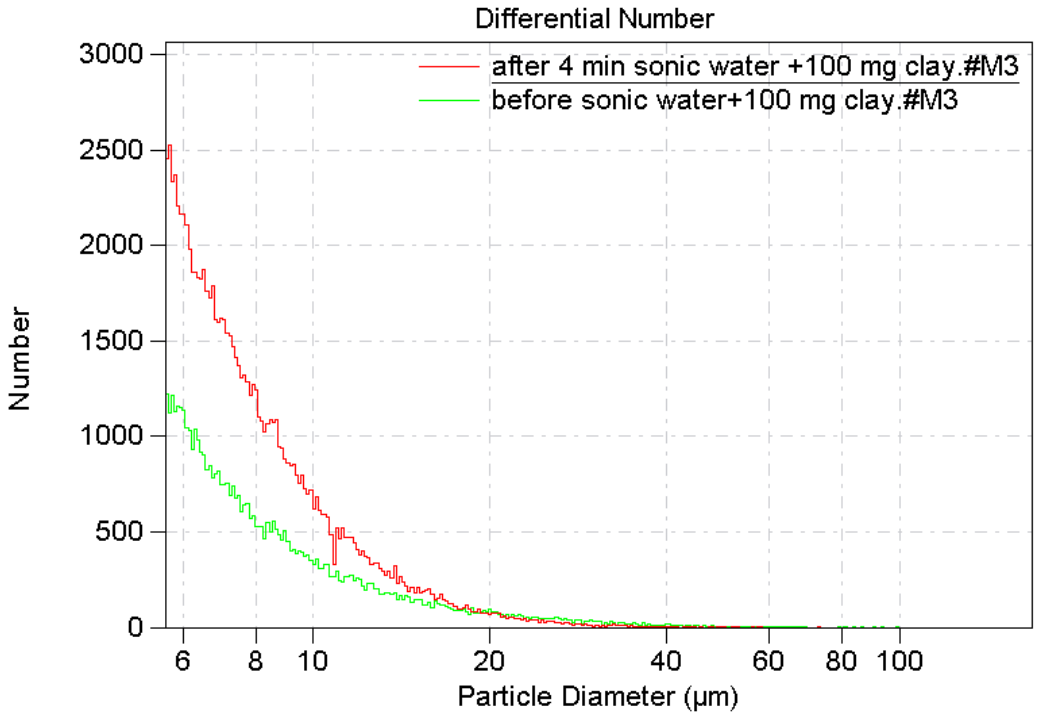


Figure 4.8Effect of 4 minutes sonication on kaolin particles breakage

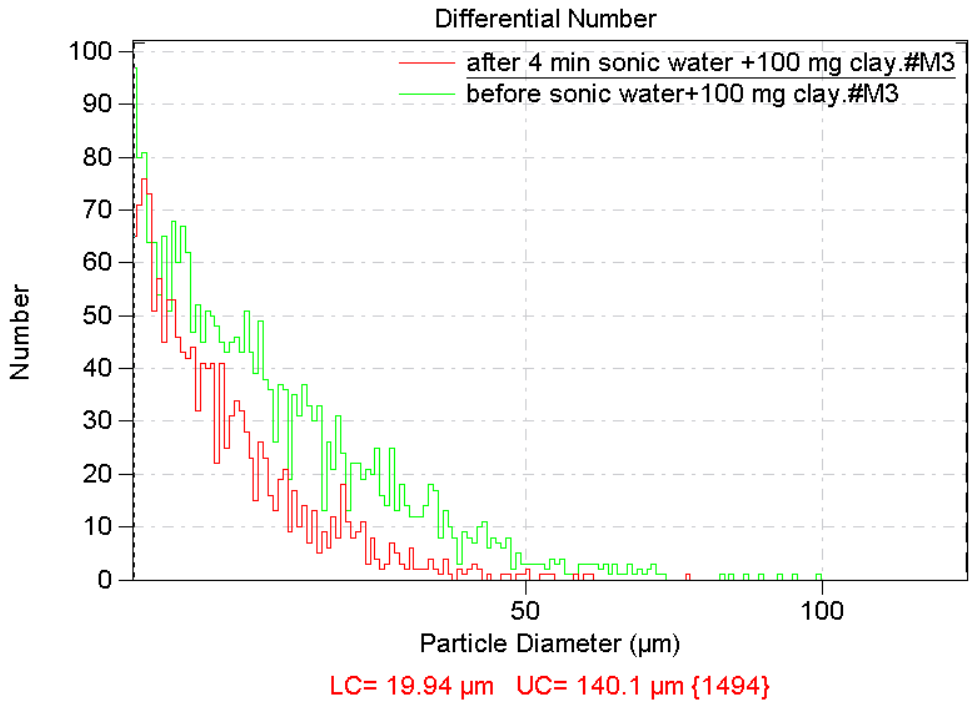


Figure 4.9Effect of 4 minutes sonication on kaolin particles breakage with the cut off at 20 µm

4.4. UV Dose Response Curves (UV-DRC)

4.4.1. Effect of sonication on UV response curve

Figure 4.10 illustrates the effect of sonication on initial slope and tailing level of UV response curve, as it was proved in previous studies sonication increases the initial slope and decreases the tailing effect [48, 14].

In figure 4.10, it can be concluded that after 1 minute sonication, there is an approximately one log decrease in number of surviving bacteria colonies compared to the control test (no sonication) at tailing level. Also, the initial slope of coliform removal is increased by 1.4 log units after 1 min sonication.

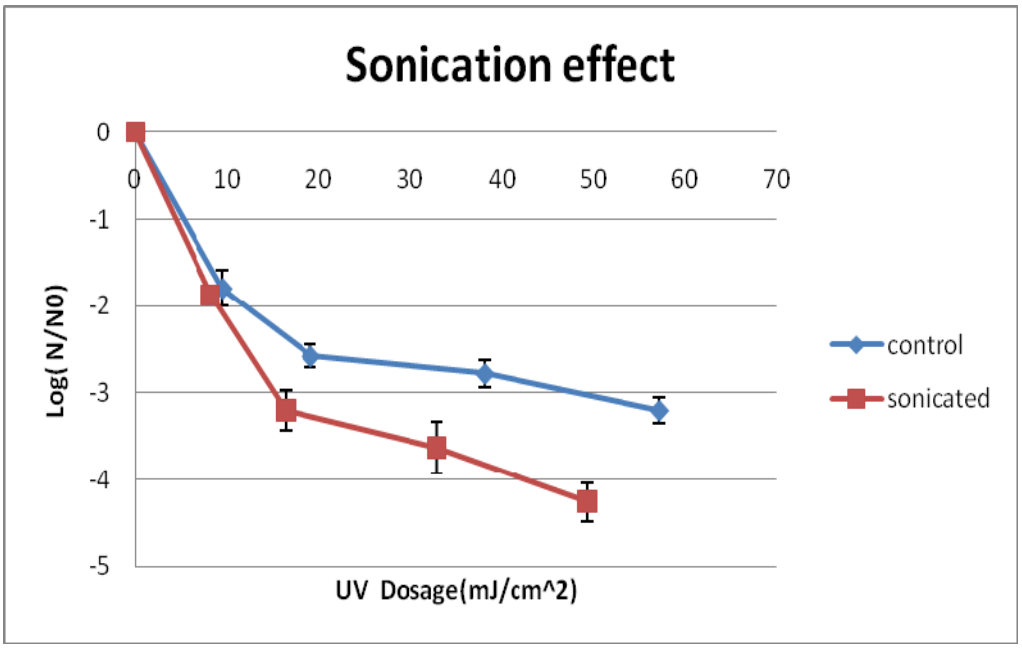


Figure 4.10Effect of sonication on UV dose response curve

4.4.2. Effect of kaolin addition on sonication in UV response curve

Earlier studies have shown that the addition of 100 mg/L kaolin can reduce the tailing level of UV-DRC. However, this reduction was statistically not significant [Torres, 2010]. Figure 4.11 shows that after 1 minute sonication in the absence of kaolin particles there is an approximately one log decrease in number of surviving bacteria colonies compared to the control test (neither sonication nor kaolin particles addition) at tailing level. Moreover, the coliform removal initial slope is increased by 1.4 and 1.9 log units after sonication and sonication in presence of kaolin, respectively.

Further tests are required to better examine the effect of kaolin addition on the UV-DRC of the effluent.

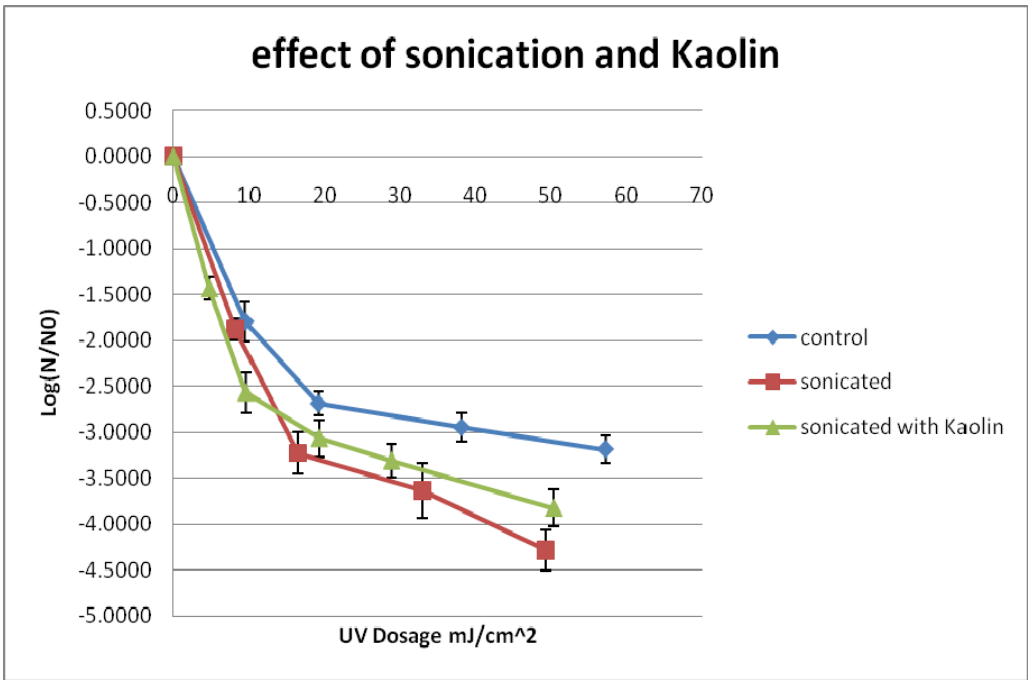


Figure 4.11 Effect of kaolin addition on sonication in UV response curve*[internal communication with Dr.Ricardo Torres(2010) ,Environment Canada,Burlington,Canada]*

4.5. Chemical Actinometry test (Iodine Dosimetry)

In this study the Iodine Dosimetry (Chemical Actinometry) was considered to evaluate the effect of kaolin addition on cavitation, this method is based on the fact that sonication generates hydroxyl radicals and subsequently hydrogen peroxide (H_2O_2) through water which can quickly react with the iodine ion (I^-) to liberate I_2, the amount of produced iodine indicates the sonochemical cavitation efficiency.

Figures 4.12 and 4.13 show the formation of peroxide due to sonication with and without kaolin addition. Based on these results, kaolin did not have any significant effect on the formation of H_2O_2. Hence, it can be concluded that kaolin particles did not enhance the caviation intensity in the sample.

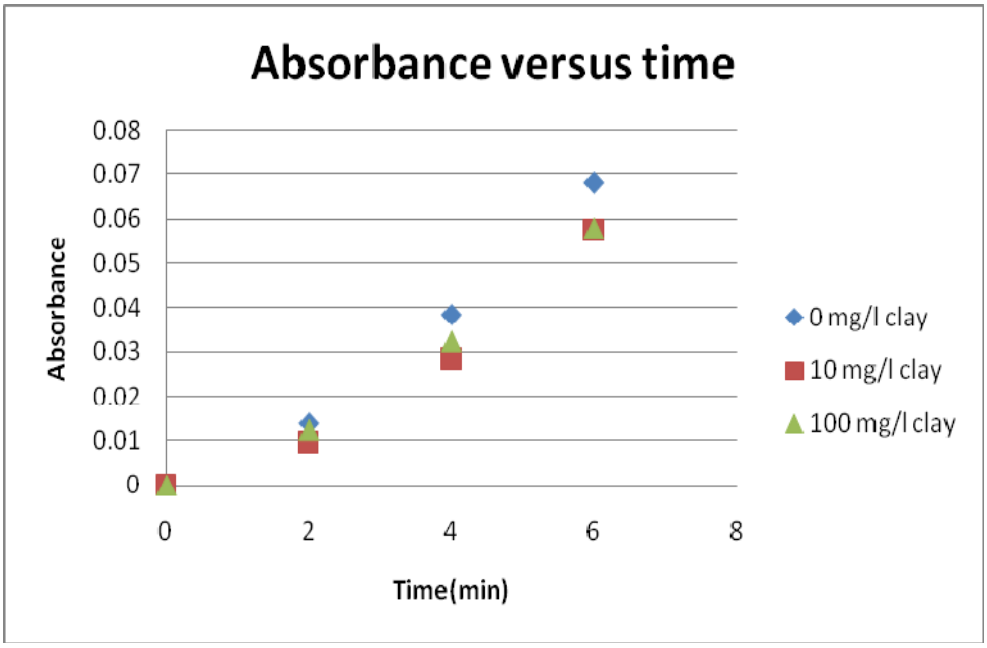


Figure4.12 Effect of kaolin addition on cavitation(absorbance)

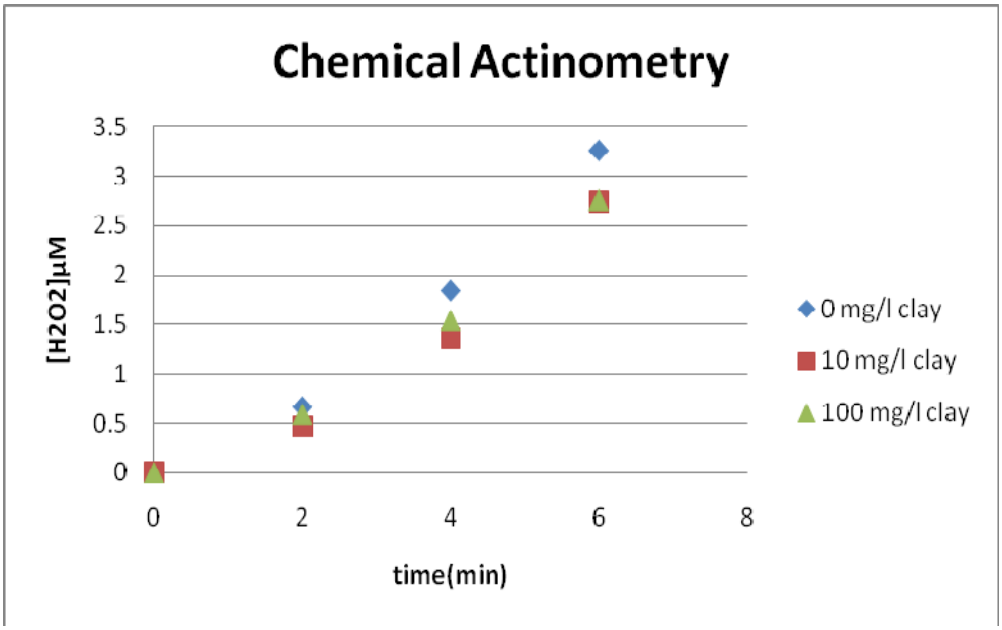


Figure 4.13 Effect of kaolin addition on cavitation(H_2O_2 concentartion)

Chapter 5

5. Conclusion

In this study, the effects of sonication on particle breakage of mixed liquor and secondary effluent have been investigated. More specifically, addition of kaolin particles on the performance of sonication step is assessed in terms of enhancing the breakage of effluent suspended particles. A more efficient particle breakage readily corresponds to more feasible treatment process. This study shows that kaolin addition had no significant effect on the breakage of effluent suspended particles. Similar to earlier studies, in this work sonication of wastewater samples for 60s resulted in reduction of CFUs number at the tailing level and increasing at the initial slope of coliform removal in UV dose response curve, however addition of kaolin particles prior sonication did not significantly affect the UV dose response curve. Chemical actinometry showed that kaolin particles would not have a noticeable impact on the cavitation intensity .The presented results are preliminary and further detailed experiments should be conducted to provide a more fundamental understanding about the exact influence of such particles.

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