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Wastewater disinfection by combination of ultrasound and ultraviolet irradiation

V. Naddeo[a,*], M. Landi[a], V. Belgiorno[a], R.M.A. Napoli[b]

[a] Department of Civil Engineering, University of Salerno, Via Ponte don Melillo, 1, 84084 Fisciano (SA), Italy
[b] Department of Environmental Science, University of Napoli Parthenope, Via Amm. F. Acton, 38, 80133 Napoli (NA), Italy

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ABSTRACT

Reclamation and reuse of wastewater is one of the most effective ways to alleviate water resource scarcity. In many countries very stringent limit for chlorination by-products such as trihalomethanes has been set for wastewater reuse. Accordingly, the use of alternative oxidation/disinfection systems should be evaluated as possible alternative to chlorine. Recently ultrasound (US) was found to be effective as pre-treatment for wastewater disinfection by UV irradiation.

The aim of this work is to investigate the wastewater advanced treatment by simultaneous combination of UV and US in terms of bacteria inactivation (*Total coliform* and *Escherichia coli*) at pilot-scale. The pilot plant was composed of two reactors: US–UV reactor and UV reactor.

The influence of different reaction times, respective US and UV dose and synergistic effect was tested and discussed for two different kinds of municipal wastewater.

An important enhancement of UV disinfection ability has been observed in presence of US, especially with wastewater characterized by low transmittance. In particular the inactivation was greater for *T. coliform* than for *E. coli*.

Furthermore, the results obtained showed also that the fouling formation on the lamps was slower in US–UV reactor than in UV reactor both with and without solar radiation.

1. Introduction

Disinfection plays a key role in reuse of wastewater for eliminating infectious diseases. Hazardous chlorination by-products restricted the use of chlorine for the disinfection of water and wastewater [1–3]. In Italy, a very stringent limit for chlorination by-products such as trihalomethanes (THMs < 30 μg/L) was set for wastewater reuse. Accordingly, the use of alternative oxidation/disinfection systems should be evaluated as possible alternative to chlorine.

Ozonation and UV have lately emerged as a viable alternative by virtue of their operational costs and accurate maintenance operation. In particular the UV disinfection is affected by suspended solid matter and by formation of organic fouling on the lamps [4–8]. Many studies tested wastewater and drinking water ultrasound disinfection ability, with performance of about 90% and with low frequency and high density [9–15].

The reason why ultrasound power can produce chemical and physical effects is due to the phenomenon of cavitation. Ultrasound cavitation refers to the production of microbubbles in a liquid which are formed when applied a large negative pressure [10,16–18]. High ultrasound power produces strong cavitation in aqueous solutions causing shock waves and reactive free radicals (e.g. $^{\bullet}OH$, $HO_2^{\bullet}$ and $O^{\bullet}$) through the violent collapse of the cavitation bubbles [19,20].

These effects should contribute to the physical disruption of microbial structures [21,22] and inactivation as well as the decomposition of toxic chemicals [23–25]. These may include organic compounds as: surfactants [26], pharmaceutical compounds [27,46], dyes [28], humic acids [15], cyclic hydrocarbons [29] and aromatic compounds in which phenols are also included [30–32].

The physical–chemical water characteristics significantly contribute to ultrasound disinfection efficiency [10,19,20]. One of the most recent ultrasound disinfection application observed that cell elimination induced by ultrasound irradiation is permanent, as denoted by the zero reappearance rates of the disrupted bacteria [14].

An alternative to chlorination is the use of multiple disinfectants which can enhance inactivation of pathogens and reduces the chlorination by-products (i.e. trihalomethanes and haloacetic acids). The advantages of combined treatments are: the direct exposure of bacteria to biocide due to a mechanical break-down of bacterial clumps; their dislodgement from surfaces/places difficult to reach; the increased permeability of the cell walls of the bacteria to the biocide; the reduction in their temperature resistance [33,19].

Indeed, one of the most interesting topics in the recent advances in sonochemistry is the possibility of double or more excitations with ultrasound and other types of energy as UV [34–36]. Therefore,

* Corresponding author. Tel.: +39 089964139; fax: +39 089964100.
E-mail address: vnaddeo@unisa.it (V. Naddeo).

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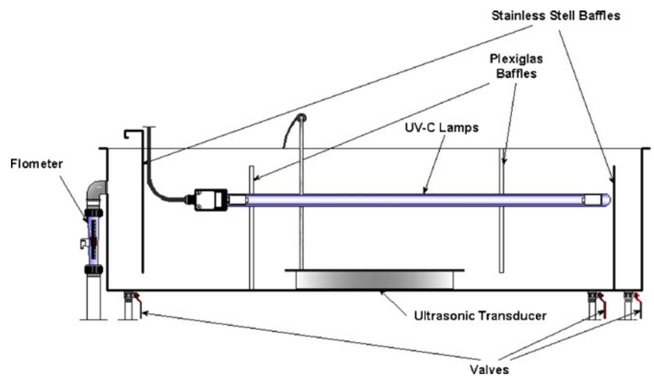

Fig. 1. Schematic longitudinal section of the US–UV reactor (reactor 1).

recently ultrasound (US) was found to be effective as pre-treatment for wastewater disinfection by UV irradiation [37] as well as the combined effect of ozone and ultrasound providing good efficiency for disinfection of well water [38].

This research investigated the advanced wastewater treatment by combination of UV and US in terms of bacteria inactivation (*Escherichia coli* and *Total coliform*) at pilot-scale. The influence of retention time, relative US and UV dose and synergistic effect was also tested and discussed.

2. Materials and methods

2.1. Pilot plant set-up

The US–UV reactor (Fig. 1) was designed and realised to investigate the disinfection effectiveness; the process and the reactor is patented by University of Salerno.

The whole pilot plant was composed by two reactors: the US–UV reactor and a UV reactor.

The pilot plant was set up for use at the wastewater treatment plant (WWTP) of Mercato San Severino (Salerno, Italy). Reactor 1 (US–UV reactor, Fig. 1) was composed of ultrasonic transducer TD-US 1400 (CEIA S.p.A., Italy) at low frequency (39 kHz), variable power from 350 to 1400 W and two low pressure UV-C lamps (Trojan Technologies, Canada) of 150 W each. Reactor 2 (UV reactor) was composed of only two low pressure UV-C lamps (PROCOM s.r.l., Italy) of 200 W each. In both reactors, a volume of 80 L has been designed for disinfection zone. The influent flow rate has been controlled by two valves and two flowmeters (OPPO s.r.l., Italy).

2.1.1. Characteristics of treated wastewater

The Mercato San Severino WWTP is based on conventional biological process scheme, and presently treats both domestic and industrial discharges. The pre-treatment of this plant is based on screens and grit removal, while secondary treatment is performed with activated sludge. Tertiary treatment is carried out by conventional deep bed sand filter [39]. Disinfection is obtained by chlorination. A physico-chemical characterization of the influent wastewater treated by Mercato San Severino WWTP is shown in Table 1.

A pilot plant was installed downstream of the full-scale sand filtration unit of the WWTP. The tests were carried out at two wastewater streams having different characteristics; one of these with a very high pathogen concentration (Type-B), both with a very low transmittance ($UV_{254} > 0.6\ cm^{-1}$). It is well known that UV disinfection performances are reduced by low transmittance values [40]. This is the reason why in these works, different retention times were tested. The physico-chemical characterizations of the wastewater, treated by pilot plant, are summarized in Table 2.

Table 2
Characteristics of the wastewater treated by pilot plant.

Parameter	Pilot plant influent	
	Type-A	Type-B
Total coliform (CFU/100 mL)	270,000	–
Escherichia coli (CFU/100 mL)	14,400	310,000
pH	7.91	7.36
Redox potential (mV)	65.42	138
Conductivity (mS)	2.65	2.88
COD (mg/L)	76	84
TSS (mg/L)	43.6	58.4
Turbidity (NTU)	2.27	14.9
UV_{254} (1/cm)	0.72	0.81

2.1.2. Experimental conditions

The tests with a Type-A influent, carried out in reactor 1, are performed at 2, 5, 10, 15 and 30 min of retention times with both disinfection technologies turned on. The US and UV disinfection tests were also carried out separately in the same conditions. The *E. coli* and *T. coliform* inactivation was evaluated. According to previous studies [13,41], in these tests the ultrasound energy power was set at 1400 W and UV dose at 1656 mJ/cm^2. At 15 and 30 min of retention times a 350 W of US energy power was also tested.

The tests with a Type-B influent, carried out in both reactors, were performed at around 30 min of retention times in both UV and US–UV disinfection, guaranteeing a constant flow by the employment of valves and flowmeters. The disinfection effectiveness was evaluated by *E. coli* inactivation. In reactor 1 US power was set at 350 W and UV dose at 1656 mJ/cm^2. In reactor 2 UV dose was set to 1656 mJ/cm^2, too.

These tests were carried out with solar radiation ('sun' tests) and without solar radiation ('dark' tests). These tests were conducted continuously for four days.

2.2. Analytical methods

Analytical measurements were conducted at Environmental Engineering Laboratory of the University of Salerno, Fisciano (SA), Italy. The membrane filter method was used for microbiological analysis according to Standard Methods [42]. Acetate cellulose type filter with 0.45 mm pore size (Millipore, USA) was used for water sample filtration as well as *m-endo* medium (Oxoid, Italy) and TBX (Oxoid, Italy) were used respectively for *T. coliform* and *E. coli* retention. The results were expressed in colony forming units per 100 mL (CFU/100 mL). Absorbance measurements were performed using a λ12 UV-Vis spectrophotometer from PerkinElmer. Turbidity was detected by HACH turbidimeter (Model 2100N). BOD_5, COD and TSS were detected following the Standard Methods [40]. The measurements of pH, conductivity and redox potential were carried out by three probes (Hanna Instruments®).

Table 1
Chemical–physical characteristics of the influent of Mercato San Severino WWTP (mean of daily average values, from 1/1/2007 to 31/12/2007).

	Average value	Standard deviation	Maximum value	Minimum value
BOD_5 (mg/L)	171.4	135.9	570	31
COD (mg/L)	594.7	415.4	2659	119
TSS (mg/L)	846.3	857	5520	39.6

Table 3
Effect of US dose on bacteria inactivation at different retention times by US, UV and US–UV disinfection process (tests in reactor 1 with WW Type-A).

Bacteria species	Retention time (min)	US (350 W) (CFU/100 mL)	US (1400 W) (CFU/100 mL)	UV (2× 150 W) (CFU/100 mL)	US (350 W) + UV (2× 150 W) (CFU/100 mL)	US (1400 W) + UV (2× 150 W) (CFU/100 mL)
Total coliform	2	–	140,000	110,000	–	50,000
	5	–	125,000	35,000	–	20,000
	10	–	65,000	30,000	–	10,000
	15	87,000	30,000	15,000	7500	0
	30	43,500	15,000	4	0	0
Escherichia coli	2	–	7,600	6,200	–	4,400
	5	–	7,400	2,200	–	1,800
	10	–	4,200	1,800	–	1,400
	15	6,720	2,400	400	320	2
	30	2,880	900	0	0	0

3. Results and discussion

The results obtained with Type-A influent show that the disinfection efficiency by UV increased from 30% to 98% as retention time increased from 2 to 15 min respectively (Table 3). According to the investigated conditions and wastewater characteristics, the UV process alone was not sufficient to decrease the *E. coli* colonies under the limit set for wastewater reuse in Italy (10 CFU/100 mL). The use of ultrasound improved UV disinfection; in wastewater sample Type-A, characterized by initial *E. coli* density of 14400 CFU/100 mL, the combined process US–UV allowed to decrease the *E. coli* colonies under 10 CFU/100 mL with a retention time of 15 min (Table 3). The formation of free radicals (e.g. •OH and O•) with a high oxidative power and the disruption of microbial cells by shock waves represent the great disinfection power generated by US [20].

In the combined disinfection system (US–UV) the synergistic effect was strong in terms of *T. coliform* inactivation compared to *E. coli* removal (Table 3). However the initial concentration (C_0) of *T. coliform* was higher than the C_0 of *E. coli*; considering the ratio C/C_0 the inactivation is comparable for both of the bacteria species.

The effect of US dose on bacteria inactivation was investigated at two different contact times (15 and 30 min) by US, UV and US–UV disinfection process (Table 3). The synergistic effect in term of both effectiveness of the disinfection system was obtained at 15 min of treatment with the higher US dose (Table 3).

According to the previous results, the tests with a Type-B influent were carried out for consecutive three days with a retention time of 30 min. In these tests the *E. coli* inactivation, after some hours of treatment, was about 94% in both reactors. This behaviour was imputable to high turbidity and high concentration of suspended solid in the wastewater Type-B (Table 2). Both these parameters are very relevant for UV disinfection. Solid particles impair UV disinfection through three effects; light beam scattering, particle shading and shielding the bacteria harboured therein [43,44]. In Fig. 2 it is possible to appreciate qualitatively the high turbidity in wastewater.

On the third day, after about 55 h of continuous treatment, while in the UV reactor the inactivation went down until 77%, in the US–UV reactor the disinfectant power was still up 90% (Fig. 3). The tests showed the influence of ultrasound on lamps fouling formation. In fact, while during the tests, the lamps in UV reactor were becoming dirtier day by day, in US–UV reactor the UV lamps were perfectly clean even after three days of treatment (Fig. 2). The US cleaning effects was guaranteed by the collapse of cavitation bubbles which produce liquid jets on the lamps' surface. In this way, the US breaks the cake layer on the lamps making the UV beans emission achievable in wastewater.

US irradiation in combined process has a double key role; US increase the disinfection performance not only by its disinfection power but also by providing the constant cleaning of the UV lamps, guaranteeing constant disinfection performances.

In the 'dark' tests the initial inactivation was about 97% in both reactors. At the fourth day, after about 80 h of continuous treatment, the *E. coli* inactivation decreased to 80% in UV reactor, while in US–UV reactor the inactivation was still higher than 92% (Fig. 4).

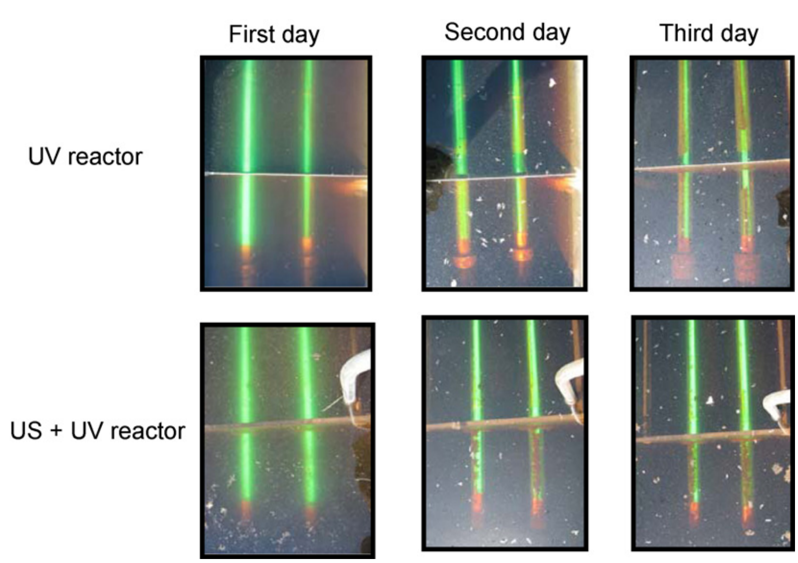

Fig. 2. Comparison between fouling on the lamps in the UV reactor (up) and in the US–UV reactor (down) versus time.

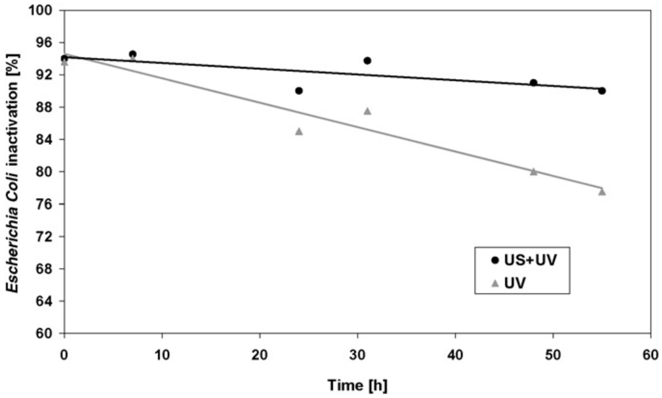

Fig. 3. *Escherichia coli* inactivation versus length of the test in "sun" condition (30 min of hydraulic retention time, WW Type-B).

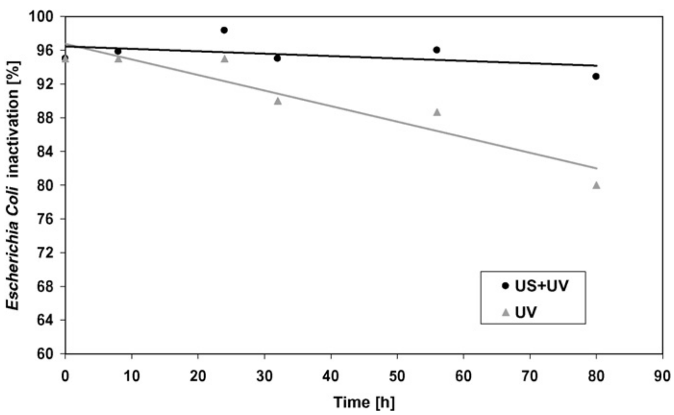

Fig. 4. *Escherichia coli* inactivation versus length of the test in "dark" condition (30 min of hydraulic retention time, WW Type-B).

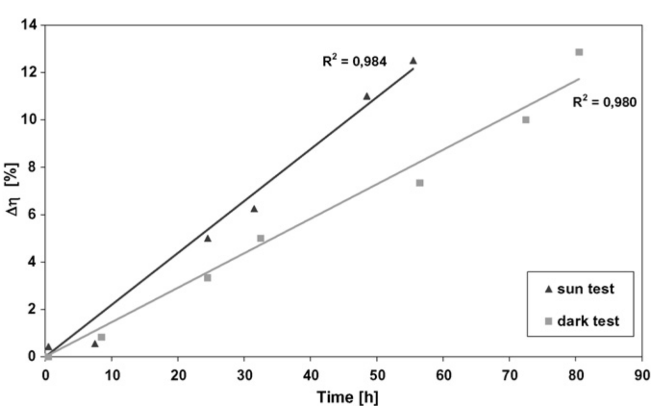

Fig. 5. Relative disinfection efficiency ($\Delta\eta$) of *Escherichia coli* between US–UV and UV efficiency versus the length of the test, with and without solar irradiation (WW Type-B).

Fig. 5 shows the remainder of the disinfection efficiencies ($\Delta\eta$) versus the time (h) plot, in both 'sun' and 'dark' conditions, and where $\Delta\eta$ was definite with the following formula:

$$\Delta\eta = \eta_{\text{US+UV}} - \eta_{\text{UV}}$$

It represents the difference between *E. coli* percentage inactivation in US–UV ($\eta_{\text{US+UV}}$) and in UV (η_{UV}) reactors. Fig. 5 shows an appreciable linear trend ($R^2 = 0.98$) in both 'sun' and 'dark' conditions. Apart from that, it shows that the regression line of 'sun' test has slop greater of the 'dark' test; at the same time the 'dark' condition has a lower inactivation power difference, given the lower fouling formation. The higher fouling formation is explained by the fact that in 'sun' tests was presented the light emission in visible light range; this solar irradiation stimulates, on the quarts sleeves, the growth of microorganisms, typically fungal and filamentous bacteria, which constituted the biofilm [45].

Overall the results show the synergistic effect in terms of both effectiveness of the disinfection system and operational/maintenance benefit related to the self-cleaning of the UV lamps.

4. Conclusions

In this work advanced ultrasound disinfection process was investigated at pilot-scale with an ultrasound and ultraviolet combined process in a US–UV reactor, patented by University of Salerno.

The tests were conducted with wastewater characterized by low transmittance, where generally UV disinfection was not suitable. Instead, this innovative combined treatment is able to guarantee high performances also with low transmittance wastewater.

An important enhancement of UV disinfection ability has been observed in presence of US, especially.

Sonication effects also increase the UV disinfection efficiency in terms of reduction of big particles and cleaning lamps. What is more, the analyses show the effects of solar radiation on UV lamps fouling formation and the specific possibility to remove fouling by US.

Thus the combined process US–UV can be considered as a valuable alternative to conventional oxidation/disinfection processes when less expensive solutions such as chlorination cannot be applied because of very stringent limits set by regulations (e.g. trihalomethanes). Indeed, the combined process US–UV allowed decreasing the *E. coli* colonies under 10 CFU/100 mL (wastewater reuse Italian limit) with a retention time of 15 min.

The advanced ultrasound disinfection (US–UV), applied under such conditions, may be an effective technique in all WWTP where the wastewater reuse is an important integrative/alternative resource for not drinking purposes. Nonetheless, further studies should be performed to evaluate better the disinfection effectiveness on a different bacteria species and in continuous operation, subsequently in terms of formation of unknown ultrasound disinfection by-products (UDBPs).

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