The effectiveness of a multi-spark electric discharge system in the destruction of microorganisms in domestic and industrial wastewaters

A. M. Anpilov, E. M. Barkhudarov, N. Christofi, V. A. Kop'ev, I. A. Kossyi, M. I. Taktakishvili and Yu. V. Zadiraka

ABSTRACT

The aim of this work was to investigate the effectiveness of a high voltage multi-spark electric discharge, with pulse energy of 1 Joule, in killing microorganisms in wastewater. Wastewater from primary treated effluent arising from domestic and industrial sources was abstracted for continuous pulsed discharge disinfection. The wastewater contained a large mixed population of microorganisms ($\sim 10^7$ CFU ml^{-1} [10^9 CFU 100 ml^{-1}] total aerobic heterotrophic bacteria) including vegetative cells and spores. The electrical conductivity of the wastewater ranged from 900–1400 μS cm^{-1} and it was shown that a specific energy of 1.25–1.5 J cm^{-3} was required to achieve 1 log reduction in bacterial (faecal coliforms/total aerobic heterotrophs) content. This is higher than that previously shown to reduce the population of *E. coli* in tap water of low conductivity, demonstrating the role of total wastewater constituents, including dissolved and particulate substances, water colour and the presence of microbial spores, in effective disinfection. The system can be engineered to eradicate microbial populations to levels governed by legislation by increasing treatment time or energy input.

Key words | electric discharge, microbial disinfection, pulsed-plasma, slipping surface discharge (SSD), UV, wastewater

A. M. Anpilov
E. M. Barkhudarov
V. A. Kop'ev
I. A. Kossyi
M. I. Taktakishvili
General Physics Institute of the Russian Academy of Sciences,
38 Vavilov Street, Moscow,
Russia

N. Christofi (corresponding author)
Pollution Research Unit,
Napier University,
Edinburgh EH10 5DT,
Scotland, UK
Tel: +44 131 455 2490
Fax: +44 131 455 2291
E-mail: *n.christofi@napier.ac.uk*

Yu. V. Zadiraka
SPLITS Technologies Ltd,
Edinburgh,
Scotland, UK

INTRODUCTION

High voltage electrical discharge in water (Goryachev *et al*. 1998; Anpilov *et al*. 2001) has been considered as a potential method of water treatment to kill microorganisms, negating the use of chemicals such as chlorine that leads to disinfection by-products (acetaldehydes, haloacetic acids, haloacetonitriles, haloketones and trihalomethanes) which may additionally compromise human health (Sketchell *et al*.1995; Van Leeuwen, 2000; von Gunten *et al*. 2001). Factors favouring their use in microbial disinfection include the generation of UV radiation (especially effective near wavelengths of 254 nm), acoustic, shock waves, chemically active substances (e.g. hydroxyl ions and hydrated electrons), cavitation processes in an acoustic wave field, pyrolysis and hydrolysis (Yutkin 1986; Anpilov *et al*. 2002). There are also possible synergetic effects following physical and chemical reactions.

The main literature on microbiological disinfection of water deals with the use of electric discharges in microbial destruction under physiologically relevant conditions (solutions), and in drinking and natural waters utilizing specific microbial populations (Goryachev *et al*. 1998). Such studies, carried out under laboratory conditions, show the effectiveness of the killing process with values of 0.5–1.0 J cm^{-3} for a 1 log microbial reduction (Goryachev *et al*. 1998). Anpilov *et al*. (2002) used pulsed electric discharges utilizing a multi-electrode Slipping Surface Discharge (SSD) treatment system to kill *E. coli* and viruses (coliphages) in tap water of low conductivity. The energy required for a 1 log reduction of coliphages (0.15 J

cm^{-3}) was less than that required to inactivate *E. coli* (0.3 J cm^{-3}). Wastewater disinfection is expected to be more problematic as it is associated with a high electrical conductivity, has a complex chemical and biological (microbiological) composition, can be turbid due to suspended particulates and can suffer from colouration. The energy requirement for effective microbiological disinfection of wastewaters can be much higher.

Two approaches to water disinfection using high voltage pulse discharge are possible and have been used. High energy pulses of ≥1 kJ can be used where the main killing factor involves shock waves (Bogomaz *et al*. 1991). The advantage of this approach is that killing is practically independent of conductivity and other water characteristics. Serious drawbacks are associated with shock loads on the system and the design of a reactor and its power source are complicated. An alternative approach uses pulses of comparatively low energy (~1 J) (Goryachev *et al*. 1998; Anpilov *et al*. 2002). Here shock loads are practically absent and the design of a treatment system can be rather simple. This kind of system can be designed as a module, enabling its safe use in hospitals, homes and other low water volume usage situations. The main factors affecting the killing of microorganisms are UV radiation, chemically reactive free radicals and other oxidising species and cavitation processes in the acoustic wave field. It is evident that conductivity, colour and dissolved and particulate substances in the water affect treatment using low energy discharges. The literature on the use of pulsed electric discharges for wastewater treatment is scarce (Goryachev *et al*. 1998) and this is especially true for systems using low energy pulses.

The possibility of using low energy (~1 J) discharge technology for the disinfection of real wastewaters is an interesting and realistic proposition. In this study we have devised an experimental system based on SSD devices to disinfect wastewater following primary treatment (it can also be used following secondary sewage treatment). The SSD system differs from well-known 'needle-plane' discharge devices (Efremov *et al*. 2000). The multi-electrode design reduces wear of the electrodes and can be used to affect treatment of large volumes of water (PCT 1999; Anpilov *et al*. 2001, 2002). The absence of sharp edges on electrodes additionally increases the durability of the system. This discharge is achieved in a water–gas mixture that is formed when gas bubbles emanate through the inter-electrode spaces. Liquid and gas phases and plasma–liquid boundaries in the discharge generate a variety of chemically active substances. Gas bubbles tend to significantly lower the breakdown threshold and can easily produce discharges in waters with high conductivities.

The main aim of the present study was to determine the effectiveness of microbiological disinfection of real wastewater arising from domestic and industrial sources. This work deals with specific energy input into water necessary to achieve a 1 log reduction in the mixed bacterial population of this natural effluent. In the study, the physics of the discharges was investigated as well as the microbiological aspects. This report deals primarily with the microbiological effects of treatment under the different conditions imposed.

MATERIALS AND METHODS

The SSD treatment system

A diagrammatic representation of the experimental system is presented in Figure 1. It comprises the following main units: two reactor chambers through which wastewater flows, discharge units and two five-channel high voltage pulse power units (one controlling 5 and the other 4 discharge units). The reactor chambers are each constructed of stainless steel as a rectangular tube with a total volume of 10 l (treating 5 l wastewater) and a 100 cm^2 cross-section. Each discharge unit is similar to that described by Anpilov *et al*. (2002). Six ring electrodes made of titanium or stainless steel are placed coaxially around a Teflon tube with 2 mm gaps. Gas (air) is fed into this tube and emanates through the gaps. The power source (5 channels) generates the following: pulse initial voltage (U_o), 20 kV; pulse frequency (f), 0–100 Hz; energy of a storage capacitor (W), 2 J per channel; current pulse duration (τ), 4–5 μs. Control of discharge from the capacitor in the discharge unit to the electrodes is achieved using one thyratron. Discharge current and voltage were measured with a Rogovsky belt and divider accordingly.

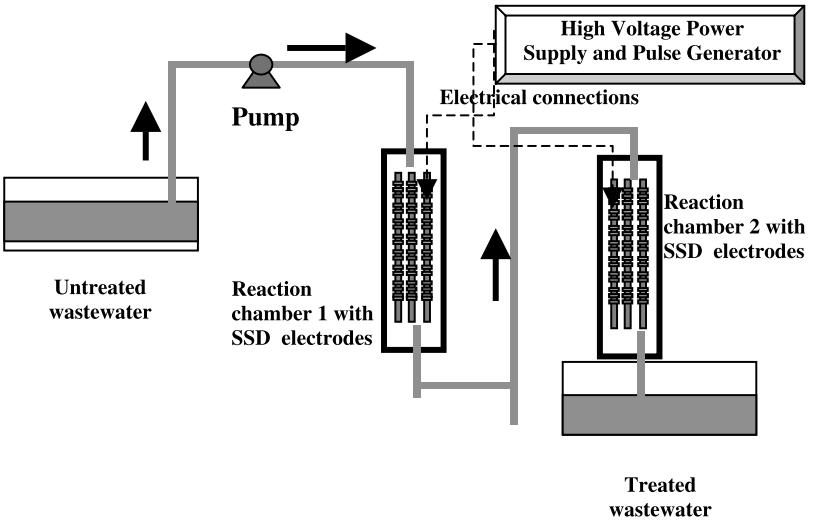

Figure 1 | Diagrammatic representation of continuous wastewater treatment using the SDD system of plasma discharge electrodes.

Their signals were directed to the inputs of a Tektronix TD S3012 oscillograph and UV radiation in the range $200 \leq \lambda \leq 350$ nm was registered with a ''blind'' in the visible light area photo-electric multiplier PEM-142 having a caesium antimonide photocathode. Energy input into water was determined using current and voltage oscillograms. The PEM signal shows the dynamics (duration) of the ''hot'' period of the discharge. Typical oscillograms are shown in Figures 2(a) and (b). Energy contribution into the discharge during a single pulse is determined by:

$$W_p = \int I_i(t) U_i(t) \mathrm{d}t$$

Effectiveness of the discharge energy transformation is calculated by:

$$\lambda = W_p / W$$

Wastewater treatment

Water treatment was carried out using wastewater directly abstracted from the final effluent stream at the Livingston Wastewater Treatment Plant in West Lothian, Scotland, UK. This wastewater only received primary treatment. Water was pumped from the waste stream through the inlet of reaction chamber (RC) 1 receiving treatment from discharge units in both RC1 and RC2 prior to discharge back into the wastewater stream through the outlet of RC2. RC1 contained 5 and RC2 contained 4 discharge units.

The effectiveness of water disinfection was determined using different wastewater flow rates of 2.0, 2.5, 3.33 and 5.0 l min^{-1}. These represented hydraulic retention times (HRT) of 2.5, 2, 1.5 and 1 min for each chamber. Variation in flow rates made it possible to change specific energy input into the water using values calculated from oscillograms. Three samples for microbiological analyses were taken at the reactor inlet and outlet at intervals between 30–60 s. This procedure was repeated 30 times over a time interval of several minutes for a single treatment run. Air to facilitate effective discharge was introduced through the inter-electrode gaps of the discharge units using pumping with flow control.

Microbiological analyses

Sampling for microbiological (bacterial) analyses was done from both the inlet and outlet ports of the reaction chambers. Wastewater treatment was carried out over a period of time to allow 30 samples each of inlet

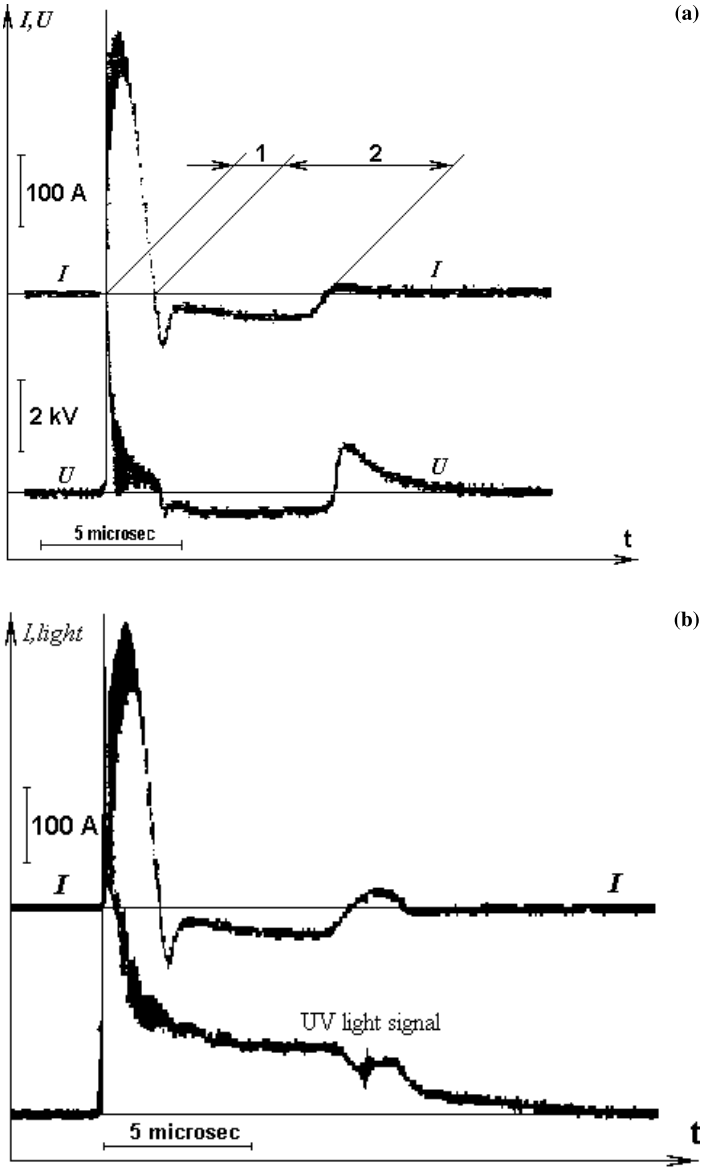

Figure 2 | (a) Typical oscillograph trace of SSD current and voltage. (b) Typical oscillograph trace of SSD current and UV light at λ=200–350 nm.

(untreated) and outlet (treated) water to be recovered at intervals representing the HRT. In calculating microbial destruction, the average populations in the replicate inlet and outlet samples were utilised. Temporal variations in wastewater populations were expected and indeed found for experiments utilising different flow rates and carried out on successive dates, each over a period of 1.5–2.5 h. The experiments were done in triplicate for the 2.5 min HRT treatment; in duplicate for the 2 and 1.5 min HRT treatments and as a single analysis for the 1 min HRT treatment.

All samples for microbiological analyses were analysed by Scottish Water Scientific, Edinburgh, Scotland (NAMAS Accredited). They were dispatched for processing within 1–2 h of sampling. Bacterial enumerations were made using spread plating techniques, for total

Table 1 | Variations in wastewater physicochemical factors in the inlet and outlet streams of the SSD reactors during microbial treatment. All values are in mg l^{-1} unless otherwise stated

Physicochemical factor	Exp. 1 inlet (1)	Exp. 1 inlet (2)	Exp. 2 inlet (1)	Exp. 2 inlet (2)	Exp. 2 outlet
Turbidity (NTU)	16.10	15.40	18.10	24.00	16.20
Iron	5.95	5.37	5.08	8.34	4.65
Cadmium	<0.005	<0.005	<0.005	<0.005	<0.005
Chromium	1.26	0.091	0.138	0.731	0.060
Copper	0.793	0.176	0.462	0.492	0.162
Nickel	0.888	0.074	0.127	0.535	0.048
Lead	>0.01	>0.01	<0.01	<0.01	<0.01
Zinc	0.075	0.124	0.154	0.090	0.097
5 day BOD	3.1	4.9	3.8	2.5	9.3
COD	33	44	49	39	54
Ammonia	<0.5	<0.5	<0.5	0.8	<0.5
Nitrite	0.14	1.28	0.85	1.27	1.90
Nitrate	12.95	18.22	15.80	15.48	19.08
pH	7.20	7.20	7.30	7.90	7.40
Soluble reactive phosphorus (as P)	0.53	<0.40	1.30	1.09	0.44
Total phosphorus (as P)	1.10	0.70	1.40	1.10	0.80
Suspended solids	25	25	24	40	24
Total nitrogen (as N)	12.7	21.1	16.9	17.5	22.4
Total oxidised nitrogen (as N)	13.09	19.50	16.65	16.75	20.98

aerobic heterotrophic bacteria (TAB) using total plate count agar (incubations at 22°C) and faecal coliform (FC) bacteria using McConkey's Agar (incubations at 37°C). All results are expressed in Colony Forming Units (CFU) ml^{-1}.

Chemicals analyses

Natural wastewater, which showed temporal changes in physicochemical factors, was treated by the SSD system. Wastewater characteristics including conductivity, metal analyses and other physicochemical factors were monitored and analyses carried out by Scottish Water Scientific (Table 1 shows selected analyses).

RESULTS

Figures 3 and 4 show the changes in bacterial populations before (inlet) and after (outlet) treatment

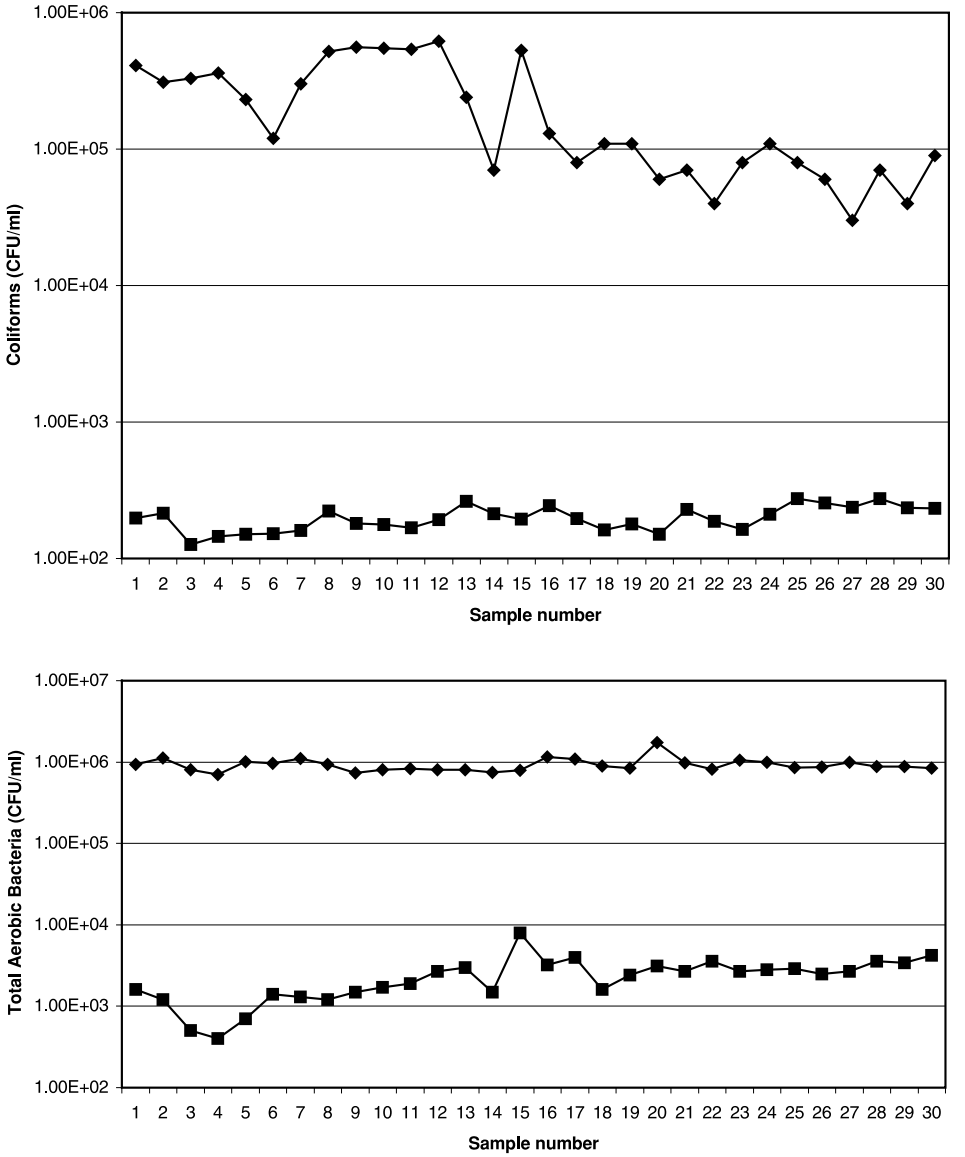

Figure 3 | (a) Temporal variation of faecal coliform bacteria in inlet (◆) and outlet (■) samples of wastewater treated by the SSD system. Experiment 1, Hydraulic Retention Time was 2.5 minutes. (b) Temporal variation of total aerobic heterotrophic bacteria in inlet (◆) and outlet (■) samples of wastewater treated by the SSD system. Experiment 1, Hydraulic Retention Time was 2.5 minutes.

by the SSD system. Only the data for wastewater with a 2.5 min hydraulic retention time (HRT) are presented for experiments carried out on different days. It is evident that up to 3 log reductions in populations were achieved for FC (Figures 3(a) and 4(a)) and TAB (Figures 3(b) and 4(b)) although this was not consistent. The inlet microbial populations were variable and these were significant over the ~1.5–2.5 h treatment period.

Figure 5 shows the conductivity changes of the wastewater during experiments carried out for the different HRT. Thirty samples were taken for each experiment. The conductivity varied between 900–1400 μS cm^{-1} and not more than 10–15% for any given day.

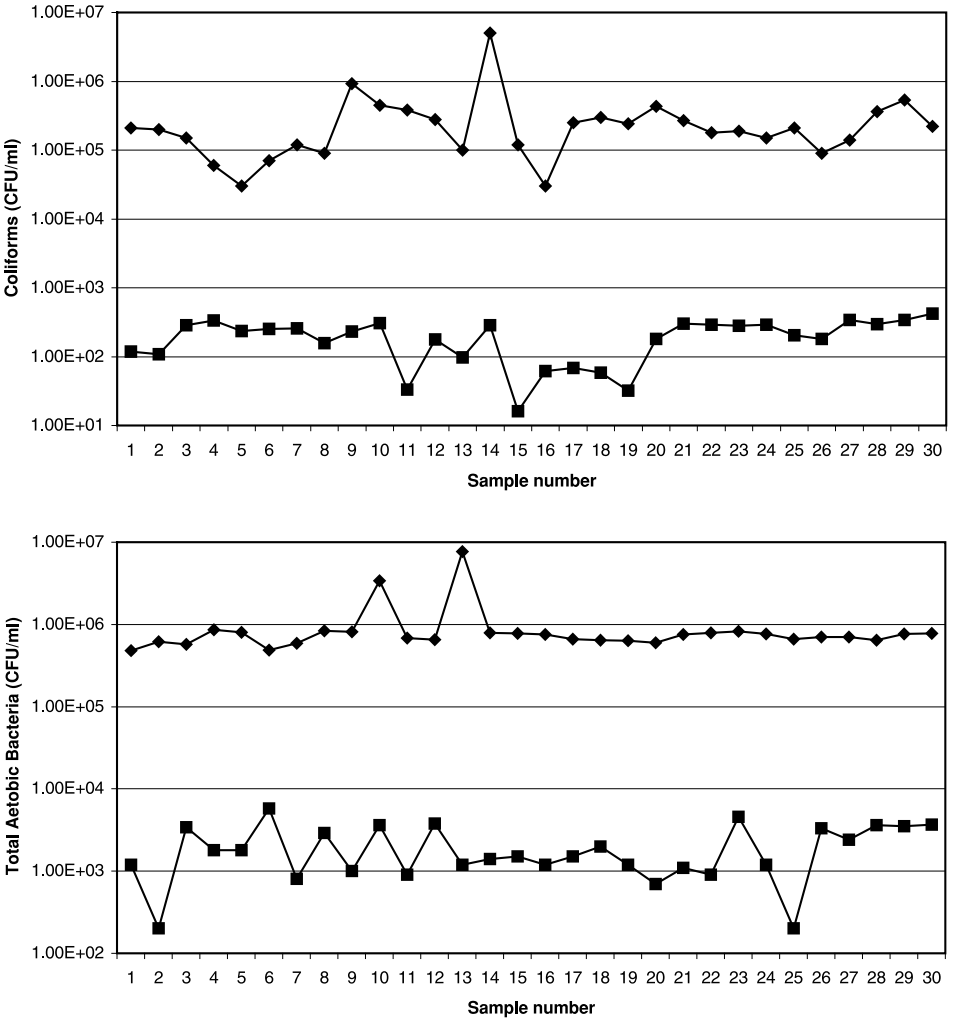

Figure 4 | (a) Temporal variation of faecal coliform bacteria in inlet (◆) and outlet (■) samples of wastewater treated by the SSD system. Experiment 2, Hydraulic Retention Time was 2.5 minutes. (b) Temporal variation of total aerobic heterotrophic bacteria in inlet (◆) and outlet (■) samples of wastewater treated by the SSD system. Experiment 2, Hydraulic Retention Time was 2.5 minutes.

Dependence of bacterial decay ($\log N/N_0$) on specific energy input is presented in Figure 6. All points represent the mean of 3 replicates with 30 sampling points. Specific energy input y was determined by:

$$y = \frac{W_p f \tau}{V} = \frac{W_p f}{U}$$

where f = frequency of pulse sequence in Hz; τ = duration time in seconds (s); V = volume of the treated water in cm^3 and U = water flow rate in $cm^3\ s^{-1}$. In the systems used 1 pulse energy contribution into discharge (W_p) was ~0.4 J cm^{-3}. It follows from Figure 6 that the specific energy input (y_0) necessary for 1 log reduction in the number of faecal coliforms and total aerobic heterotrophic bacteria differs slightly and is within the range 1.25–1.5 J cm^{-3}.

DISCUSSION

The sterilising influence of high-voltage pulse discharges on microbial cells can be approximated by the following exponential decay equation:

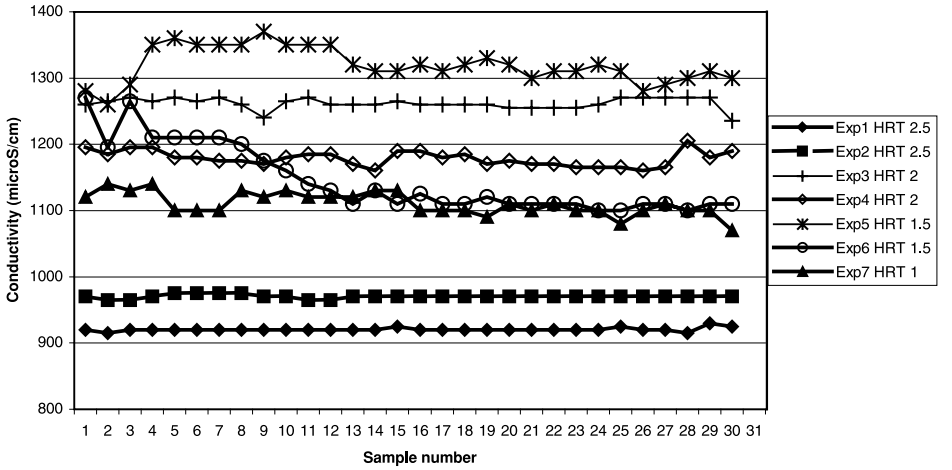

Figure 5 | Temporal variations in electrical conductivity of wastewater treated by the SSD system for various hydraulic retention times (HRT).

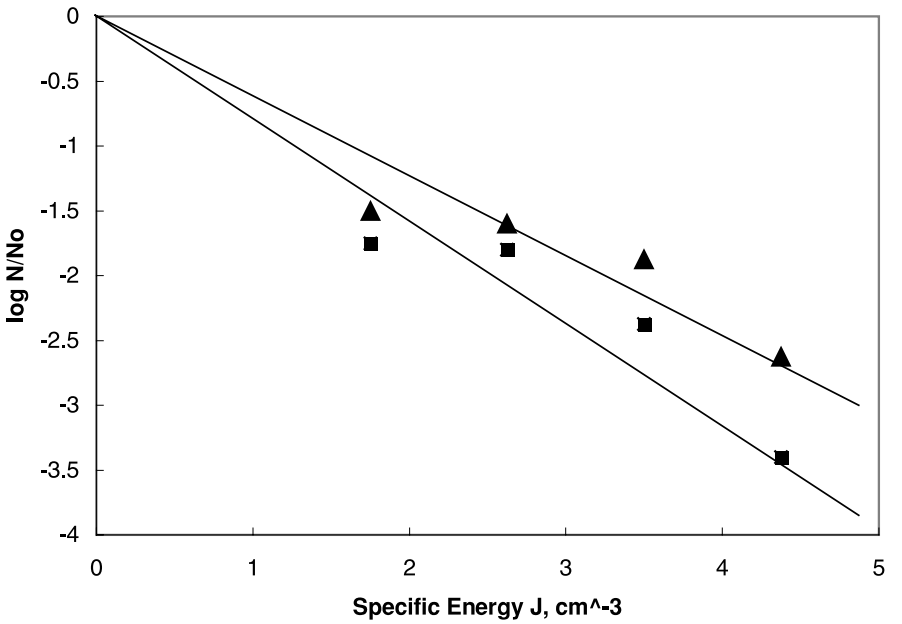

Figure 6 | Log bacterial population (N/N_0) changes versus energy for the SSD-treated wastewater. ▲, total aerobic heterotrophic bacteria (22°C); ■, faecal coliforms (37°C).

$$N = N_0 \exp(-\alpha y)$$

where N_0 is the initial microbial population, N is the concentration following a specific energy input (y) and α is the coefficient of the reaction rate. This relationship has been confirmed in laboratory experiments with drinking water and physiologically relevant solutions (Goryachev *et al.* 1998; Anpilov *et al.* 2002). Experimental data presented in Figure 6 correspond to values obtained for different HRT experiments and represent the average results of a large number of measurements (each with 3 replicates for 30 sampling points) obtained on different days. Nevertheless these results follow the exponential decay equation rather well. It is evident that, with the wastewater used, physicochemical factors, such as electrical conductivity, suspended material, colour, metal

content, etc. (Table 1), do have a small influence on the decay but do not have a dramatic influence on the effectiveness of disinfection. It follows from typical current I, voltage V and UV radiation ($y = 200$–350 nm) traces presented in Figures 2(a) and (b) that two main stages of pulsed arc discharges are observed corresponding to the existence of high-temperature plasma channels. The first stage is associated with a peak current ~300 A, a mean voltage ~800 V and duration ~1.75 μs. The second stage has mean current ~40 A, mean voltage ~500 V and duration 5.5 μs. It is seen that the specific yield of UV radiation (radiation energy per unit discharge input energy) in stage 2 is several times higher than in stage 1. It can also be seen that UV radiation remained practically constant, indicating nearly isothermal characteristics of the processes under the conditions imposed. It is important that this fact be taken into account when selecting discharge mode, as UV is mainly responsible for microbial killing. Temperature change of the wastewater during discharge treatment is negligible and undetected and thus unlikely to be a factor in microbial killing.

One of the best-known methods of non-chemical water disinfection is based on the use of UV lamps, mainly of the mercury electrode type, producing a biologically active spectrum in the range 180–300 nm. The presence of mercury in these lamps is dangerous for the environment. In addition, the quartz walls of these lamps act as a barrier to the short wavelength spectrum of UV radiation and glass surfaces are subject to fouling, reducing sharply the UV lamp's effectiveness with time. Both low- and medium-pressure mercury lamps are used for the microbial disinfection of potable water and wastewater (Gehr & Wright 1998; George *et al*. 2002; Huffman *et al*. 2002; Modifi *et al*. 2002). The low-pressure lamps are primarily used, emitting non-ionising UV radiation with a wavelength of 254 nm that is absorbed by and causes damage to DNA through the formation of photoproducts (mainly thymine dimers (von Sonntag & Schuchmann 1995)). Medium-pressure lamps are not common and they generate a broader UV spectrum.

Microorganisms differ in their ability to resist damage to DNA following a dose of UV and this is dependent on the physical structure of the microorganisms (affecting UV penetration) and their ability to repair the damage to genetic material. Bacterial repair mechanisms involve the SOS response activated by the *RecA* genes (Friedberg *et al*. 1995; Little & Mount 1982). It has been shown that, following UV treatment, photochemical damage of DNA can be reversed in microorganisms under visible light irradiation (photoreactivation). It is therefore possible, following inactivation, for microorganisms to regain viability.

In the SSD systems used to generate UV in this study it has been shown that oxidising species including ozone, H_2O_2 and free radicals such as hydroxyl radicals and hydrated electrons are formed due to the UV spectrum produced (Anpilov *et al*. 2001). These factors can have additional effects on microbial constituents through oxidation processes. In DNA, for example, additional hydrolysis of the phosphodiester backbone can take place initiated by ozone. Wavelengths between 180–220 nm constitute the ozone-generating region and 185 nm is used to produce ozone and H_2O_2 by what are termed ozone lamps. Studies have also shown that advanced oxidation technologies (AOT) utilising ozone, ozone with UV and H_2O_2 with UV treatment of water leads to the production of oxidising species affecting the degradation of organic substances including disinfection by-products (Williams *et al*. 2002).

Using lamps, it has been determined that the UV doses necessary to inactivate microorganisms depend on the type of organism and the required rate of inactivation with values ranging from 2 to 130 mJ cm^{-2} (Wedemeyer 1996). Bacteria such as *E. coli* are inactivated (four log [99.99%] reduction) by a UV dose of 5–10 mJ cm^{-2} (Hargy 2001). Spore-forming bacteria require higher UV doses such as 25–30 mJ cm^{-2}. Protozoans of the genera *Giardia* and *Cryptosporidium*, causing gastro-intestinal disorders and which resist breakthrough chlorination, exist as vegetative cells and cysts (oocysts). The two life-cycle stages exhibit different resistances to UV irradiation. The cysts are considered more resistant to inactivation, with doses of 3 mJ cm^{-2} (Mofidi *et al*. 2002) and 10 mJ cm^{-2} (Campbell & Wallis 2002) required for a 2 log reduction in *Giardia* cysts. Higher doses of 20–40 mJ cm^{-2} achieved a 3 log reduction in cysts (Campbell & Wallis 2002) but Hargy (2001) reported studies where 2–10 mJ cm^{-2} produced a 4 log inactivation of *Cryptosporidium* and *Giardia*.

Hassen *et al.* (2000) demonstrated a 3 log reduction in coliforms and streptococci with UV lamps functioning in wastewater with UV transmission of 45% and an effective UV dose of 100 mJ cm^{-2}. These treatment conditions are not common for many wastewaters, particularly those in the present study, which were brown in colour and associated with suspended solids. The proposed low energy SSD treatment system can be an effective alternative to UV lamp disinfection. The SSD system generates UV directly in water and the multi-spark electrodes facilitate multi-point UV discharge able to affect most microorganisms within the flowing wastewater. As disinfection relies on UV dose and microbial flocculation in wastewater (Parker & Darby 1995), acoustic shockwaves generated by the SSD system, which are also able to break up flocs within wastewater, enable better UV contact and effects on microorganisms. The shockwaves can additionally act to inactivate the bacteria (see Loske *et al.* 2002).

If one ignores the drawbacks inherent in UV lamps, making a comparison of their energy efficiency in wastewater treatment with the SSD system is rather problematic, as this comparison must be done under the same experimental conditions, which is difficult. According to Shmeliov *et al.* (1996), UV lamps with energy consumption up to 30 Wh m^{-3} (0.11 J cm^{-3}) and treating drinking water containing 10^4 CFU cm^{-3} *E. coli* gave a bacterial reduction equal to 99.7–99.99% (~4 log), which is more effective than the SSD system (Anpilov *et al.* 2002). In the case of turbid wastewaters, the problems of using UV lamps become more important and the proposed low energy SSD treatment system can be a viable alternative as a disinfection technology. In some cases a combination of the SSD system and UV lamp treatment can provide an effective and more efficient treatment alternative.

This study has demonstrated the effectiveness of the SSD system in microbial disinfection of wastewater. It utilises a combination of effectors of microbial viability including pulsed UV plasma, acoustic shockwaves, ozone, free radicals, hydrated electrons and other chemically active substances. The system can be engineered to eradicate microbial populations to levels governed by legislation by increasing treatment time or energy input.

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