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Development of a mercury-free ultraviolet high-pressure plasma discharge for disinfection

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Abstract

Ultraviolet (UV) disinfection is a critical and growing application for the disinfection of water. Current UV systems for disinfection applications are designed around the use of Low-Pressure and High-Pressure mercury-based lamps. Increasing demand to reduce and ideally remove the use of mercury requires innovative adaptations and novel approaches to current technology. A potential alternative technology could be Light Emitting Diodes (LEDs), with current low efficiencies, high costs and low operating powers, a development gap for a high-power mercury source has been identified. A mercury-free tellurium-based high-pressure plasma was developed and assessed. Though relatively low efficiencies were measured compared to current mercury-based technology rapid improvements are likely obtainable. Such an approach enables a novel adaptation to current technology utilising established: manufacturing facilities, approaches of UV system design and validation protocols. As a consequence, it offers the potential for a rapid low-cost transition to mercury-free UV disinfection where no alternative is currently available.

Introduction

Ultraviolet (UV) disinfection is a rapidly growing application in water disinfection because the discovery of inactivation of the chlorine-resistant pathogen *Cryptosporidium spp* with UV radiation at low UV fluences (UV doses) (Bolton *et al*., 1998). It is also effective against a wide range of microorganisms (Hijnen *et al*., 2006) including chlorine-resistant pathogens such as *Mycobacterium avium* (ubiquitous in biofilms in water distribution systems worldwide) (Shannon, *et al*., 2008). In contrast to many chemical methods such as chlorination and ozonation that can generate odorous substances and biohazardous materials, UV disinfection produces minimal disinfection by-products (Wurtele, *et al*., 2011).

As a result, the application of UV technology has increased during the early part of this millennium by more than 10% annually (Kolch, 2007). In 2014, the disinfection and sterilisation equipment section of the healthcare industry was estimated at \$3.1 billion USD (Jordan, 2015) and the total UV disinfection market is predicted to be valued at \$6.42 by 2022 (Anon, 2018). Regulatory UV disinfection validation protocols have been produced in Germany (DVGW, 2006), Austria (ÖNORM (2001) and ÖNORM (2003)) and the USA (EPA, 2006). Though each protocol varies in regards to specific detail, they all use the concept of biodosimetry originally proposed by Qualls and Johnson (1983) which relates a measured reduction in a non-pathogenic surrogate to key reactor performance indicators namely, lamp output (measured by sensor measurement), UV transmission of the water sample, flow through the reactor and required UV fluence (UV dose). All three protocols are based around the use of UVC lamps. The specific design of a reactor will impact its overall energy efficiency; however, in principle the lower the measured UVT, the higher the treated flow, and the higher the UV fluence required, the greater the UV radiation delivered to the system is required (i.e. the greater the UV power input to the system).

Current technology

Mercury-based lamps both Low Pressure (LP) and High Pressure (HP) (also referred to as Medium Pressure MP) are the principal radiation sources employed in the UV disinfection market to date (Dussert, 2005). These plasma radiation sources are used due to their simple and robust design, provision of high-density radiation (increasing with internal pressure) (Phillips, 1983) and suitable spectral efficiency for the purposes of disinfection.

LP lamps have a spectrally efficient output but are relatively inefficient electrically (ratio of lamp output to electrical input) at power densities used for disinfection (Heering, 2004). Increasing internal pressure (i.e. HP) increases electrical efficiency but decreases spectral efficiency for the purposes of disinfection. However, these inefficiencies can be reduced to some extent by having the mercury in the form of an amalgam which operates at a higher power in the LP region. The options for UV sources for disinfection are limited to the selection of either a lower energy density with higher germicidal efficiency (i.e. LP lamps) or a higher energy density with a lower germicidal efficiency (i.e. HP lamps), as summarised in Table 1.

An optimal UV system design accounting for financial and environmental cost would minimise the physical footprint of a system due to the reduction in operational space and raw material required for production. The system design needs would seem to favour a high-density UV source such as the HP lamp. In practice, a trade-off is made due to the reduced germicidal efficiency and so a whole life cost appraisal must be made on the basis of general application requirements and specific site conditions. In addition, concern for the potential release of mercury, that is, from broken lamps (Miller *et al*., 2013), the planned removal of mercury from general lighting in the European Union (EU) (Wetzstein, 2013) and the increasing countries committed to the Minamata convention on mercury suggest the need for alternatives. Ideally for the development of the application of UV treatment, a source that is both electrically and spectrally efficient whilst being of a high energy density for the municipal market is needed, with the materials used being less toxic than mercury.

Developing technology

Mercury plasma lamp technologies currently dominate the disinfection market. However, two alternative mercury-free sources are entering the market. The first being Dielectric Discharge Barriers (DBDs) which produce non-equilibrium plasmas (i.e. equivalent to an mercury LP lamp discharge) through a dielectric barrier (i.e. there is no contact between the plasma and electrodes) (Kogelschatz, 2003). UV generating plasmas are based on xenon (Xe) (147 nm and 150 + 172 nm from Xe_2) that can be altered with considerable flexibility using various phosphors, and due to the electrode position, it provides unparalleled flexibility in terms of geometric configuration (Kogelschatz, Eliasson, & Egli, Dielectric-barrier discharges. principle and applications, 1997). Practically, however issues with the high energy UV radiation and the necessary phosphor coatings can occur (Masoud and Murnick, 2013). Whether it is due solely to this or a combination of other practical aspects, DBDs have seen little impact on the wider market and in particular when high UVC energy is required.

The second are LEDs which are a solid state technology which emit photons of an energy dependent on the chemical composition of the diode material (Held, 2009). Wurtele *et al*. (2011) states that LEDs offer reduced disposal problems (i.e. no mercury), compact design, fast start-up (no warm up time), high cycling for just in time applications, potential for higher energy efficiency and potential longer lifetimes. Though considerable early performance predictions have been made (largely based on the development of visible LEDs) such as 75% efficiency by 2020 (Ibrahim, 2012 cited by Autin *et al.*, 2013), these have yet to be achieved. Current germicidal Wall Plug Efficiencies (WPE) of LEDs in the market are in the efficiency range of 1–2% such as that described by Mckain *et al* (2018) which has a WPE of 2% in spectral region of 270–280nm. Though larger efficiency figures can be noted such as 15% at 280 nm (Muramoto *et al*., 2014), care must be taken in their interpretation such as the stated values used (WPE vs External Quantum Efficiency (EQE)) and also the relative germicidal efficiency of the stated spectral output to ensure a basis for comparison. Practically this is a challenge when considering UVC source replacement for high UVC applications currently delivered using HP mercury lamps. An example using the lowest efficiency (10%) for a HP lamp provided by Autin *et al* (2013) a 10kW HP Hg would need approximately 44 444 diode arrays to produce an equivalent UVC output based on a 75% LED efficiency. Currently UVC LEDs match applications that have low UVC requirements such as Point-of-Use (POU) water disinfection systems which now exist (Pagan, Lawal, & Batoni, Hurdles and progress in UV-C LED technology for water disinfection, 2011). Though future improvements are expected it remains to be seen as to what level can practically be achieved and in addition to the stated challenges, the lack of validation protocols will also likely delay the application of such technology (Wurtele, *et al.*, 2011). There is a clear power limitation in UVC LEDs and a requirement for a high-power mercury-free alternative which is displayed in Fig. 1.

Table 1 Characteristics of LP, LP amalgam and HP lamps (adapted from Dussert (2005))

Characteristic	LP	LP Amalgam	HP
Hg pressure (atm)	0.01	0.01	1-2
Amount of Hg (mg)	5-50	35-100	40-400
Operational temperature (°C)	38-60	150-200	590-900
Emission spectrum	Monochromatic	Monochromatic	Polychromatic
Input power (W)	15-75	15-400	1,000-20,000
UVC efficiency (240-290nm)	32-38	30-36	12-16
Lamp life (hrs)	8000-15 000	8000-15 000	3000-9000

HP mercury-free plasma discharge concept

To produce suitable lamp plasma for use in UV disinfection an element or compound, Phillips (1983) states that the following characteristics must be achieved:

- Relatively low ionisation energy whilst having an excitation energy to produce resonance ER at desirable wavelengths
- Sufficient vapour pressure to produce optimal internal lamp pressures whilst having a low enough boiling temperature to be in gas/vapour phase whilst at lamp operating temperatures
- Chemically inert to lamp materials, that is, electrodes and lamp envelope

Mercury fulfils all these requirements splendidly, however, a direct elemental replacement is not available. The addition of a compound combining a Halogen and another emitting element (generally iodine) to a HP mercury lamp is use for visible lighting by producing a more suitable spectral output. The addition of a halogen reduces the vapourisation temperature and high temperatures (i.e. at the arc of the lamp) destabilises the compound enabling the spectral emission of the element alone (Phillips, 1983) as represented in Fig. 2.

Schafer (1976) proposed the use of antimony iodide alone as a mercury-free alternative; however, this apparently did not produce a practical output which is likely due to the internal pressure being too low to enable an efficient and practical discharge. Through assessing the elemental spectral lines, thermal characteristics both elementally and in a halogen form inclusive of thermal pressure relationships a potential solution may be devised. Three potential candidates were selected as shown in Table 2 namely iodine, tellurium and antimony all showing fairly comparable ionisation levels to mercury.

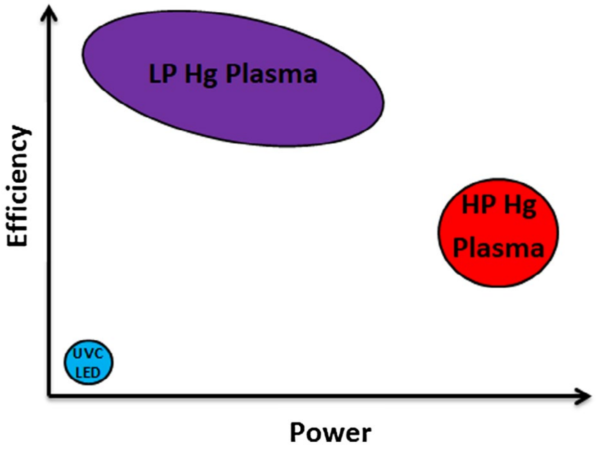

Fig. 1. A conceptual representation of the current status of UVC sources. [Colour figure can be viewed at wileyonlinelibrary.com]

The elemental spectral outputs (see Figs. 3 and 4) highlight antinomy as the most suitable spectral output; however, due to the potential pressure limitations incurred by Schafer (1976), tellurium may provide a suitable discharge using iodine to form the halide with the potential to add antimony as a dominant spectral radiator. No practical limitations of the materials were found within the literature and in practical support Turner (1994) describes the use of tellurium in an electrode discharge used for the generation of visible light (suggesting this is at a higher internal pressure than the technical proposition stated here).

In summary, the use of tellurium in the form of a halide and potentially the addition of a small proportion of antimony should enable a functional HP plasma emitting in the UV region. Theoretically, such a mercury-free discharge should be capable of delivering practical benefits as seen from conventional HP mercury lamps. Therefore research was undertaken with the following objectives:

(1) Assess whether a tellurium-based HP lamp can be produced

(2) Measure and assess the spectral output and functionality of such a source

(3) Review the suitability of such a plasma for UV disinfection and potential future improvements

Methods

Two stages of assessment were undertaken firstly the assessment of a tellurium iodide-based plasma and then

Table 2 Ionisation energies for mercury, iodine, antimony and tellurium adapted from Kramida *et al.* (2013)

Element	1st Ionisation (eV)	2nd Ionisation (eV)
Mercury	10.4375	18.7568
Iodine	10.45126	19.1313
Antimony	8.60839	16.63
Tellurium	9.0096	18.6

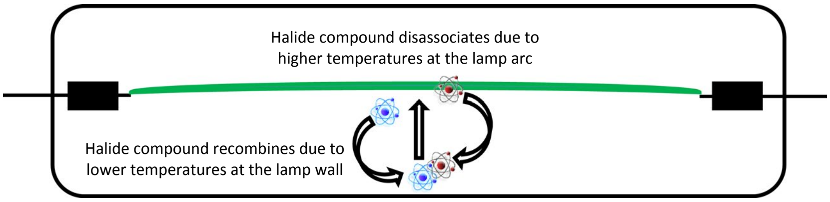

Fig. 2. Diagrammatic representation of halide cycle from lamp wall to lamp arc. [Colour figure can be viewed at wileyonlinelibrary.com]

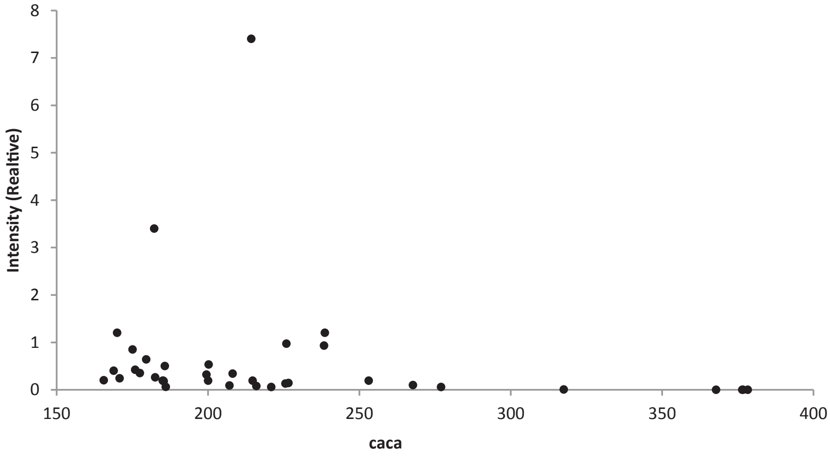

Fig. 3. Spectral data points for tellurium from all ionisation levels adapted from Kramida *et al*. (2012).

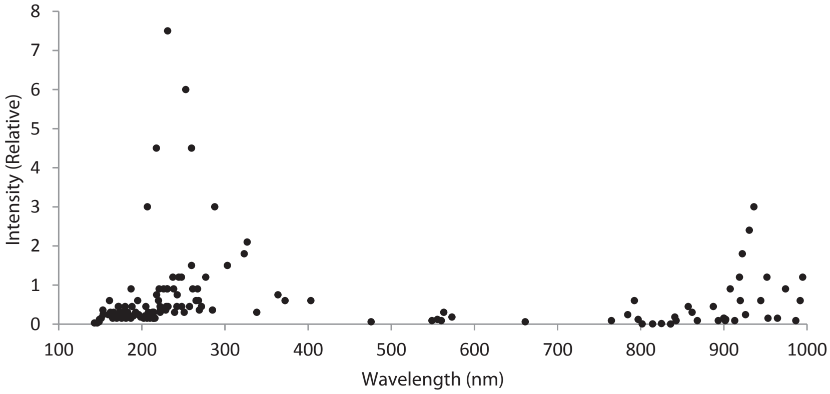

Fig. 4. Spectral data points for antimony from all ionisation levels adapted from Kramida *et al*. (2012).

secondly tellurium iodide with the addition of antimony iodide as shown in Table 3. Filling weights were based on reduced values compared to Turner (1994) with lamp geometries selected being in the region used for current HP Hg lamps (18 mm ID prototype lamps described by Phillips (1983)). However, due to a number of challenges encountered during the production of the prototypes and the final filling quantities used an additional third stage was required and described in Table 3.

The performance assessment was carried out in terms of three specific aspects: physical characteristics (i.e. arc stability), absolute spectral output and electrical characteristics. All prototypes were driven with an Eta+ (Nuertingen, Germany) X series electronic ballast with a 4kW power rating. If the prototype did not ignite, it was cooled using a freezer spray (Artic Products, Leeds UK or Electrolube, Leicestershire, UK) to reduce the internal gas pressure and consequently the strike voltage. The details of the lamp assessment are described below:

Physical characteristics

The first lamp of each prototype design was conducted in front of a viewing window (comprised of welding glass) to enable the viewing of the lamp when running the arc. Photographic images of the lamps running were taken through the viewing window using a Fujifilm (Fujifilm UK, Bedford, UK) s9600 bridge camera.

Spectral and electrical characteristics

The lamps were operated horizontally in air in a dark room with the lamp radiation passing through a collimating tube (500mm in length with internal baffles for collimation) with vertical entrance slit of 0.51mm in width. When the lamp had been stabilised, electrical characteristics were measured with a calibrated (traceable to national standard by TER Calibration Ltd, Wigan, UK) Voltech (Oxfordshire, UK) PM6000 power analyser. Germicidal efficiency was calculated from the spectral measurements accounting for the shaded slit width (0.53 mm), the measured distance from the lamp arc (0.5 m) and the arc length (0.1 m) and corrected for germicidal weightings. Two action spectra (AS) were used to calculate germicidal weightings: Spectrum B representing a target pathogen with no sensitivity below 230 nm (adapted from Meulemans (1987)) and Spectrum A representing a target pathogen with a high sensitivity below 230 nm. The AS used were adapted so relative values equalled one at 253.7 nm (AS are displayed in Fig. 5).

Table 3 Finalised lamp fillings for halide prototypes

Lamp Name	Te (mg)	TeI_4 (mg)	SbI_3 (mg)
1st Set of Prototypes			
15mm Lamp I	33	17	0
15mm Lamp II	66	33	0
18mm Lamp I	48	24	0
18mm Lamp II	96	48	0
2nd Set of Prototypes			
15mm Lamp III	40	19	0
15mm Lamp IV	70	35	0
15mm Lamp V	100	20	0
18mm Lamp III	50	25	0
18mm Lamp IV	100	50	0
18mm Lamp V	150	25	0
3rd Set of Prototypes			
15mm Lamp VI	20	4	5
15mm Lamp VII	100	20	21
18mm Lamp VI	20	10	5
18mm Lamp VII	100	50	21

Results

The performance results from all three prototype stages are provided below in Table 4 with related images to aid performance assessment provided in the subsequent figures. Practical challenges occurred during the prototype production, primarily the filling of halides and the associated sealing of the lamp which was the primary cause of prototype failures described in Table 4. In addition, the lamps that could not run were unable to reignited which was likely to an increase in the strike voltage required. Consequentially spectral and electrical measurements were taken from different individual lamp bodies to those which images were taken during running.

Benchmark Hg lamps

The Hg-based comparison lamps were made in a well-established process and were thus relatively simple to produce. The electrical performance of the lamps was extremely close and consistent (no greater then +/– 3V) to that of the designed running voltage (120V). The lamps themselves ran well in respect of starting and stability with observed centralised arcs in both the 18mm lamps and the 15 mm lamps (Fig. 6). There were indications (particularly on the left side of lamp) of the arc rising, suggesting that as per the design lamp voltage this is the maximum useable power density and consequently efficiency a Hg-based HP will deliver, thus making it an ideal benchmark. That being said, the lamps delivered only 6.6–7.5% germicidal efficiency (based on Action Spectrum A) compared

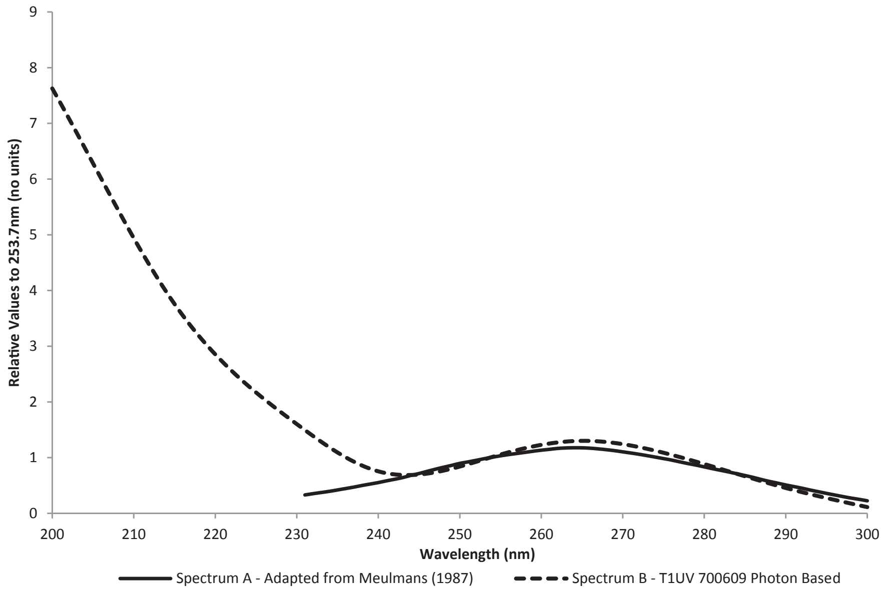

Fig. 5. Germicidal weightings for determination of lamp germicidal efficiencies.

Table 4 Electrical and spectral measurements from prototype lamps

Lamp details	Mean voltage (V)	Mean current (A)	Mean power (W)	200–300 nm (Integrated scan value W m^{-2})	Germicidal efficiency action spectrum A %	Germicidal efficiency action spectrum B %
Hg 18mm Lamp A	–	–	–	–	–	–
Hg 18mm Lamp B	122	5.67	657	10.2×10^2	6.6	13.4
Hg 18mm Lamp C	120	5.72	652	10.3×10^2	6.6	13.7
Hg 18mm Lamp D	118	5.78	649	10.9×10^2	7.2	14.4
Hg 15mm Lamp A	Ran and image taken					
Hg 15mm Lamp B	118	5.79	651	10.9×10^2	7.3	13.7
Hg 15mm Lamp C	117	5.89	651	11.3×10^2	7.5	14.3
Hg 15mm Lamp D	119	5.79	655	10.9×10^2	7.3	13.4
1st Set of Lamp Prototypes						
18mm ID Lamp I A			Did not complete production			
18mm ID Lamp I B	–80	–	–	–	–	–
18mm ID Lamp II A			Did not complete production			
18mm ID Lamp II B			Could not be run			
15mm ID Lamp I A			Could not be run			
15mm ID Lamp I B			Could not be run			
15mm ID Lamp I A			Could not be run			
15mm ID Lamp II B	95.7	7.7			–	–
2nd Set of Lamp Prototypes						
18mm ID Lamp III A	85	9.12	599	6.2×10^{-3}	0.4	0.5
18mm ID Lamp III B	88	8.9	580	Ran and image taken		
18mm ID Lamp IV A	92	8.15	660	8.9×10^{-3}	0.6	0.7
18mm ID Lamp IV B				Could not be run		
18mm ID Lamp V A	81	9.95	603	6.65×10^{-3}	0.4	0.6
18mm ID Lamp V B	80	10	600			
15mm ID Lamp III A	93	8.15	616	9.2×10^{-3}	0.6	0.7
15mm ID Lamp III B	Could not be run					
15mm ID Lamp IV A	95	7.6	605	10.45×10^{-3}	0.7	0.8
15mm ID Lamp IV B	Could not be run					
15mm ID Lamp V A	95	7.7	606	11.05×10^{-3}	0.8	0.9
15mm ID Lamp V B	90	8.3	575	Ran and image taken		
3rd Set of Lamp Prototypes						
18mm ID Lamp VI A	90	8.6	604			
18mm ID Lamp VI B	87	8.9	614	5.2×10^{-3}	0.4	0.5
18mm ID Lamp VI C	79	10.1	597	5.77×10^{-3}	0.4	0.6
18mm ID Lamp VII A	95	7.6	604	2.44×10^{-3}	0.2	0.2
18mm ID Lamp VII B	100	7.1	621	1.67×10^{-3}	0.1	0.1
18mm ID Lamp VII C	100	7.1	632			
15mm ID Lamp VI A	88	8.8	612			
15mm ID Lamp VI B	102	6.9	612	4.27×10^{-3}	0.3	0.4
15mm ID Lamp VI C	89	8.7	617	6.33×10^{-3}	0.5	0.7

(Continues)

Table 4 (Continued)

Lamp details	Mean voltage (V)	Mean current (A)	Mean power (W)	200–300 nm (Integrated scan value W m^{-2})	Germicidal efficiency action spectrum A %	Germicidal efficiency action spectrum B %
15mm ID Lamp VII A	95	7.0	559	–	–	–
15mm ID Lamp VII B	94	8.0	623	6.67×10^{-3}	0.5	0.6
15mm ID Lamp VII C	102	6.9	626	1.97×10^{-3}	0.1	0.1

to the published values in the region of 12–16% indicating significant contrast to generalised values (this being due to the length of the lamp and emitting proportion of arc to other losses such as from the electrodes) but enabling a direct like-for-like comparison of Hg HP lamps to that of the prototypes produced in stages 1, 2 and 3 below. The lamps both 15 mm and 18 mm provide a spectral output (Fig. 7) that would be expected for such an internal mercury pressure, though reduced spectral peaks are observed for the 18 mm lamps which could be due to additional absorption from the increased diameter.

Stage 1

The two initial prototypes illustrated that a lamp with a sustained plasma can be produced and run for a period of a least 20 min (the time limited by the need to carry out further scans rather than issues with the lamp), a voltage density of 9.57 V cm^{-1} can be produced (close to the comparative 12V cm^{-1} of the benchmark Hg lamps) and a non-stoichiometric Te and I lamp filling can be used to produce a functional plasma. The lamps that did not start could be visually identified as having halide dispersion near the stem removal which in conjunction with the fact that the lamps were unable to restart indicates the separation at least in part of the halogen into its elemental form.

Stage 2

The assembly yield of the second set of prototypes was increased to 75% largely due to improvements in lamp stem removal. This also enabled identification of halide residual in the lamp stem and lamp positioning post stem removal as the causes of the 25% of the failures. Lamps III, IV and also lamp V (containing a reduced percentage of TeI_4 to Te) produced voltages in a narrow region between 85 and 95V. The production of similar voltages rather than an expected change proportional to the amount of lamp filling used could indicate either a restriction of Te entering the gas phase cause by non-stoichiometric quantities of lamp fillings, or saturation of lamp filling in the gas phase, that is, increasing the lamp filling will not result in further fillings entering the gas phase and a proportional increase in lamp voltage (hence the production of second lamp design with significantly reduced fillings in stage 3).

The germicidal efficiencies of the stage 2 prototypes were significantly lower than the design target, ranging from 0.4 to 0.9% (depending on lamp and germicidal weighting). This can in part be attributed to the spectral output produced for both 18 mm (Fig. 8) and 15mm lamps (Fig. 9) which is minimal at 220 nm and displays a gradual increase towards 300 nm. Though this is not an ideal spectral output it is approximately one-tenth that of the Hg equivalent

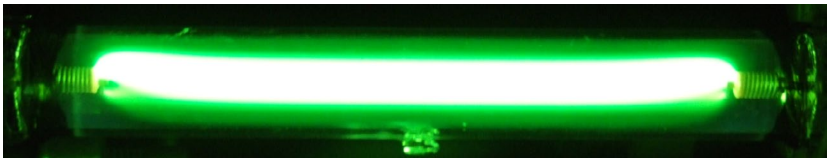

Fig. 6. Mercury lamp 15 mm Lamp A. [Colour figure can be viewed at wileyonlinelibrary.com]

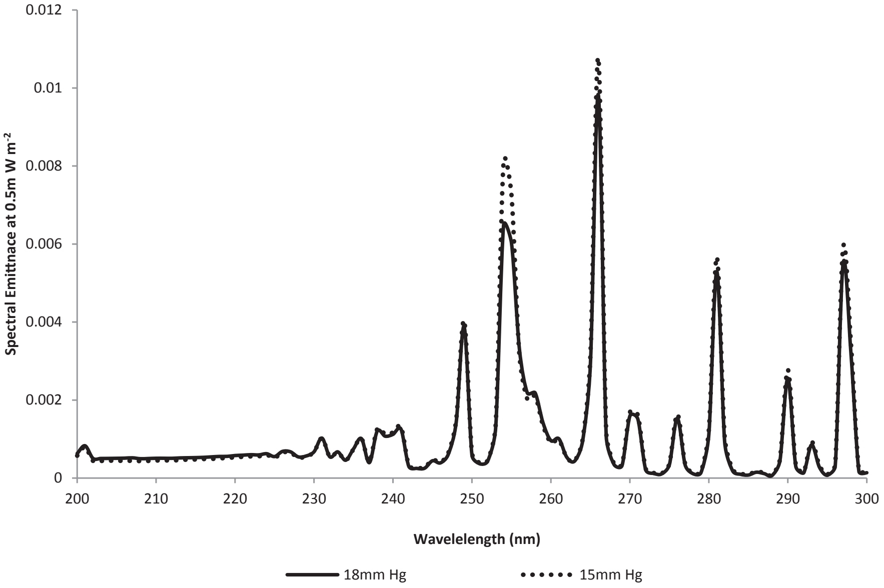

Fig. 7. Mean spectral output of mercury lamps.

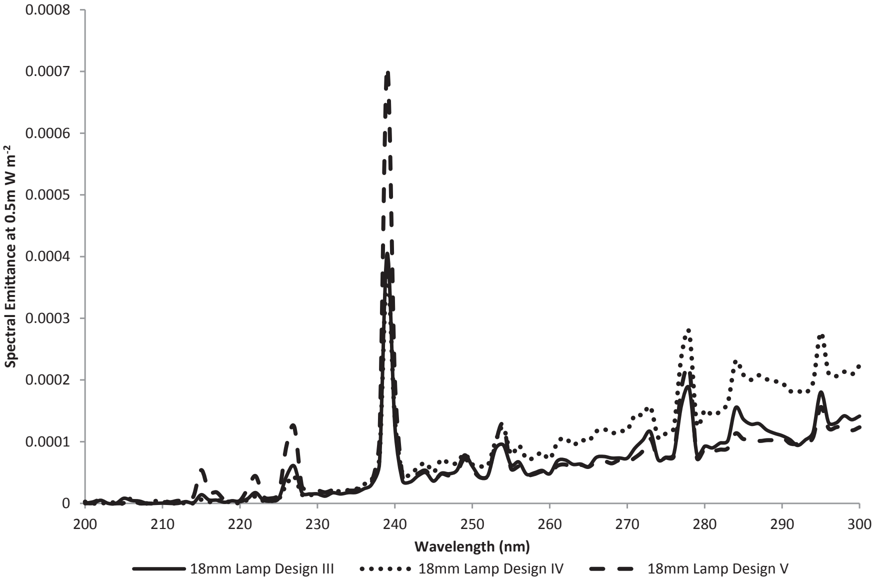

Fig. 8. Mean spectral output of 18mm diameter prototype lamps of design III, IV and V.

lamp and thus further losses must be occurring elsewhere in the lamp; the lamp driver being a contributory factor is ruled out due to the use of the measured power factor in power calculations which measured to the lamp (not inclusive of power supply losses). Noticeable features of both prototype sets 1 and 2 are the bright arcs displayed

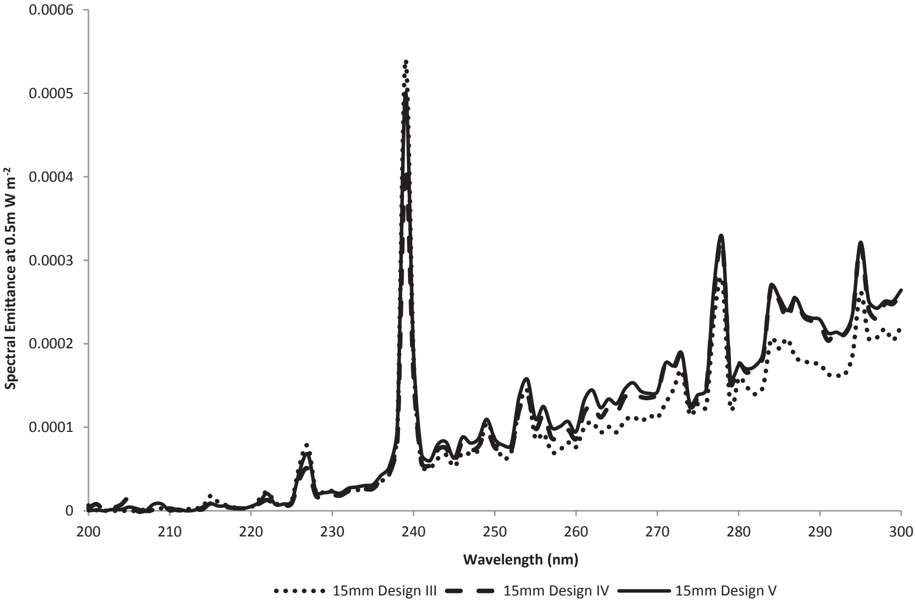

Fig. 9. Mean spectral output of 15mm diameter prototype lamps of design III, IV and V.

images indicating a high visible or output other than 200–300 nm and also the `gas pockets' particularly visible near the electrodes (Fig. 10 label B) with considerable convection currents being displayed. These latter points could be indicative of losses through unintended photon emission (not in the UV region) and/or additional thermal losses.

Stage 3

The spectral outputs of all of the prototypes in stage 3 changed considerably, with numerous peaks developing throughout the previously established continuum in stage 2 (Figs. 11 and 12). Furthermore both 15 mm and 18 mm lamps with design VI show a small but increased output below 220nm; however, this is not the case with the 18 mm lamps. In fact in contrast to the proposed increase in lamp efficiency with Sb as a dopant the prototypes produced in stage 3 are lower than that of stage 2.

The lamp design VI for both 15 mm and 18 mm lamps was based on one-fifth of the lamp fillings for lamp VII; however, minimal change in voltage was measured especially for the 15 mm lamps. This indicates that the Te in the gas phase is saturated; however, it appears I continues to enter the gas phase. This can be seen in the transition from the straight stable arc with a low visible output and no gas pockets to that of the final often turbulent lamp (as described in results stage 2). This was most clearly demonstrated in lamp 18 mm VII C shown which transitioned to a raised upper arc with a dark lower section forming from gas pockets to encompass the bottom half of the lamp. During this transition, the lamp voltage increased by one-third, suggesting that I was entering the gas phase and is the cause of the undesirable lamp characteristics after lamp warm up. The physical changes were even clearer with lamp 15 mm VIA (Fig. 13) which displayed a minimal visible output during lamp warm and a straight arc later transforming into a discharge with a high visible output yet noticeably less turbulence and ‘gas pocket’ collection around the electrodes. Because lamp design VI was designed with a reduced lamp filling and shows the same response for both 15 mm and 18 mm lamps, it suggests that reducing the amount of lamp filling particularly that of the iodide contribution is likely to increase UV output.

Discussion

The prototypes lamps all produced a sustained HP plasma discharge without the need for Hg as a filling as theoretically predicted. The lamps also produced a spectral continuum in the desired 200–300nm region and the lamp physical structure remained intact in all prototypes. Interestingly, the spectral output is limited below 230nm which is a spectral region of high absorbance for nitrate photolysis to nitrite (Mack and Bolton, 1999) and as such could be more desirable than that of the conventional HP mercury discharge spectrum. These findings are not only novel but are also critical characteristics of any future lamp to improve the performance of high UV density radiation sources.

The overall measured efficiency, however, was considerably lower than expected, with the highest performing variant producing approximately one-tenth of the germicidal output compared to their Hg counterparts. However, at

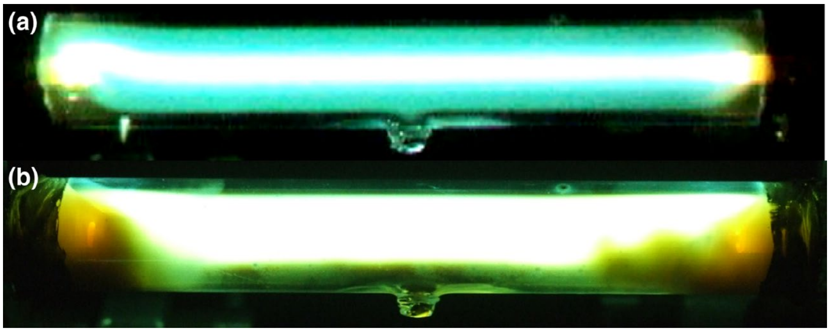

Fig. 10. Lamp 15mm IIB. (Image A (Top) taken during the warm up stages of the lamp and Image B (Bottom) taken when the lamp had warmed up). [Colour figure can be viewed at wileyonlinelibrary.com]

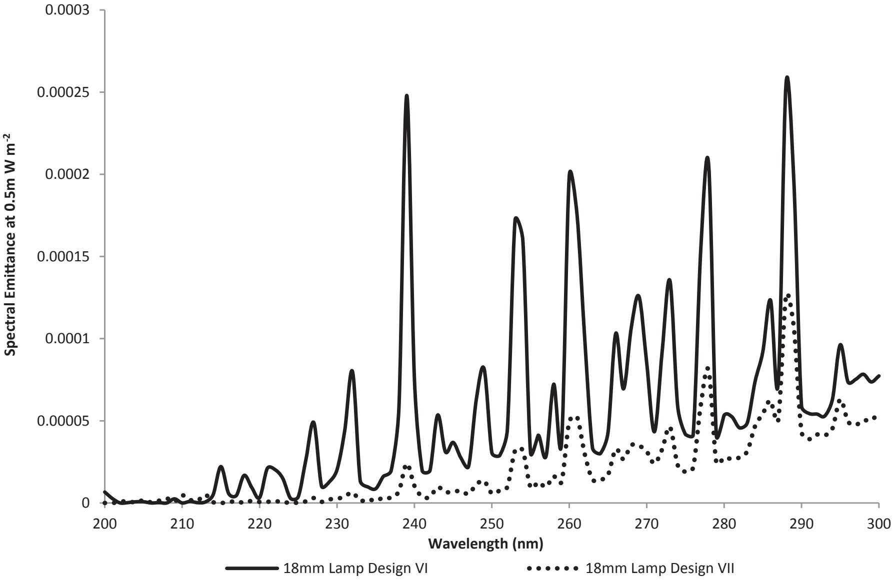

Fig. 11. Mean spectral output of 18mm diameter prototype lamps of design VI and VII.

0.9% efficiency at 606W (54540 mW of UVC), the best performing variant would be equivalent to 90900 UVC LED arrays based on a 2% efficient 30mW output. As such the concept which is not yet optimised has potential merit, however, some practical challenges on plasma stability and the inability to reignite need to be addressed.

Lamp instability is likely due to the halide transition above 600°C ($2TeI_{2(g)} \rightleftharpoons Te_{2(g)} + 2I_{2(g)}$) (Holleman and Wiberg, 1995) either producing arc instability from additional I_2 or condensation of Te (dependent on the delivered ratios). The inferred saturation of lamp filling (suggested by both voltage limitation and stability) suggests that condensation did not appear to be occurring, or if so, not to the detriment of the functionality of the lamp. In contrast I_2 did appear to affect the stability of the arc as the lamp voltage was stable; however, excess iodine would likely stimulate visible emission and therefore contribute to a lower UVC efficiency. If the lamp filings can be optimised this should in theory improve both lamp stability and efficiency considerably.

All of the prototypes which did strike got to full running power (not including the transition into the turbulent stage) in approximately 1 to 2 minutes, a similar timeframe to that of a Hg HP lamp making system design allocations (e.g. duty/standby requirements), the same as that for HP mercury lamps currently in use. Additionally, as the lamp has nominally the same physical and emissive characteristics to Hg-based lamps, it would easily be applied in current methods of design and system validation smoothing its route to market compared to other technologies such as UVC LEDs.

Possible design improvements

Further practical improvements to develop such a lamp for the commercial market will involve several additional

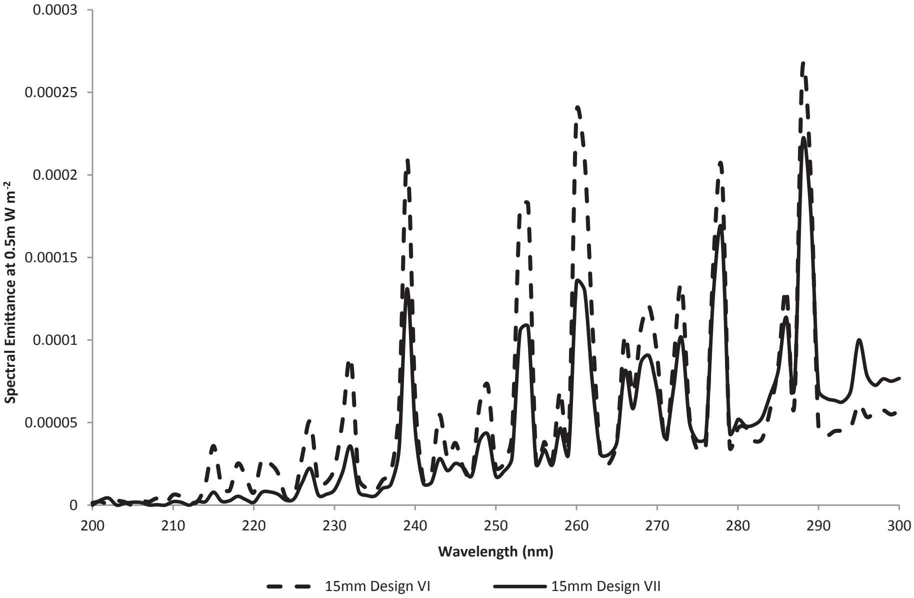

Fig. 12. Mean spectral output of 15mm diameter prototype lamps of design VI and VII.

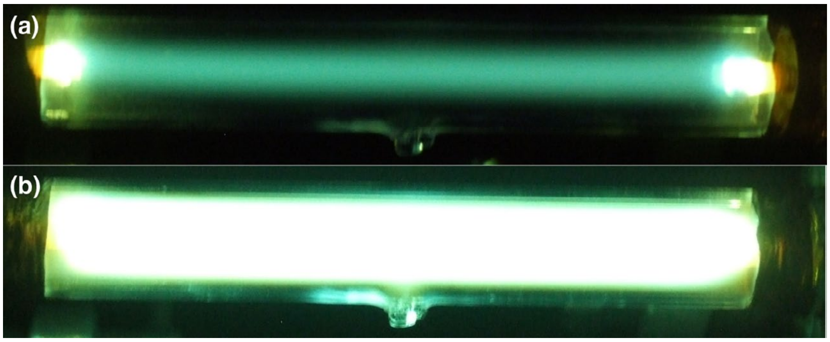

Fig. 13. Lamp 15mm VIA. [Colour figure can be viewed at wileyonlinelibrary.com]

development steps. The core requirement being to reduce the amount of iodine used to the point where it has little to no adverse effect on the arc stability and output above and beyond forming and maintaining the plasma. This will be critical to improve the performance of the lamp and once achieved for an optimal discharge the further following practical considerations can be assessed:

- This will likely include optimisation of the electrical lamp driver which may require optimisation of electrical frequency (Yan and Hui, 2000) but will almost certainly require a configuration providing a higher strike voltage for successful lamp reignition.
- The lamp strike voltage could be reduced with the use of a 'penning gas' (Phillips, 1983) replacing argon with a dual gas combination with differing ionisation levels leading to a greater production of ions (Penning and Addink, 1934).
- The lamp running voltage could be increased by raising the buffer gas pressure to increase the lamp impedance and/or changing the buffer gas used (though this would further increase the necessary strike voltage) and also offer the potential for optimisation of spectral output.

Ultimately a number of subtle design iterations will be required following fundamental plasma improvements to produce a lamp to meet a design specification based on market requirements. Implementing such proposals in conjunction with the lengthening of the lamp may well produce considerable efficiency improvements leading to its practical implementation.

Potential market future considerations

The UV market is multifaceted and comprised of many sub sectors that address not only the needs of large municipal markets but also industrial applications of varying size, as well as domestic markets. Such differing market requirements with respect not only size but also to other critical criteria such as, but not limited to, intermittent processes requirements (cycling), space limitations, specific customer requirements (e.g. absence of mercury, system pressure requirements and environmental temperatures), capital cost drivers and operational cost drivers as well as a variation of validation specifications from strict specifications to none. It this therefore that unlikely that one UV source will be optimal for all these applications and consequently the Te lamp presented will likely suit a number of market segments and will be affected by the continuous developments of all the other UV generating technologies. Those markets which are primarily driven by operating costs or electrical requirements will be best matched by traditional LP Hg-based sources as shown in Fig. 1. However, for those applications where the removal of Hg is a primary driver the selection between the presented Te lamp or LEDs will be separated by application. LEDs currently will compete favourably with low power requirements (e.g. lower flows, higher UVT) and high intermittent processes and where cost is a lower driver. The Te lamp, however, would compete more favourably in the reverse conditions with higher power requirements (e.g. higher flows and lower UVT) and low intermittent processes and where cost and potentially space are higher priorities. Importantly the 'traditional' lamp design used by the Te lamp design has the potential to utilise established and field-tested electronic driver technology as well as being able to utilise already established and widely accepted validation protocols providing a unique offering for UV applications.

An important next step throughout the development of UV source technologies is a wholistic review of technologies in respect of not only direct whole life costs but also carbon costs, sustainability (including the potential for developments), supply chain vulnerability of critical components (i.e. rare earth elements) and of course toxicity. In respect of toxicity this is a potential concern in regard to either a lamp break through handling or in the event of lamp and quartz sleeve breakage in a UV system, that is, into the treated fluid.

In regard to toxicity, mercury is well known for its impact on human health because the 1950's following the Minamata disaster and is linked to various disorders including: neurological, nephrological, immunological, cardiac, motor reproductive and genetic (Zahir *et al*., 2005). Though negative health impacts can occur from ingestion another key method of absorption into the body occurs in its vapour phase where approximately 80% of inhaled gases can be absorbed (Ha, *et al.*, 2017). However, numerous elements and their associated compounds can have a degree of toxicity and both tellurium and iodine are not excluded from this. Toxicity-related data for mercury, tellurium and iodine are shown in Table 5.

Tellurium apart from the described application is used widely in industry and agriculture with many further new applications arising and studies to date have shown to have negative impacts due to its toxicity (Vij and Hardej, 2012). Primary concerns for its transmission route is via ingestion (as per many of the trials) or more practically through absorption of gas or dust via industrial processes (Taylor, 1996). Though iodine toxicity can occur (Angel *et al*., 2008), it has been and continues to be widely utilised in relation to direct human contact applications primarily as an antiseptic agent and this practice is expected to continue (Vermeulen *et al*., 2010). Iodine can also be utilised as disinfectant for water in remote settings; however, a restriction on the quantity consumed daily is advised (Backer and Hollowell, 2000). Thus, though iodine toxicity has been identified with appropriate limits, its use in applications has been deemed to be acceptable.

The routes of potential exposure are critical when considering the toxicity of lamp fillings. This will primarily be either through ingestion if the contents enter the treatment fluid or through inhalation via user maintenance (assuming gloves are used when handling). In both cases, the practical challenges of tellurium as a lamp filling having a higher melting and vapourisation temperature than mercury become beneficial. Following loss of lamp function tellurium will rapidly cool solidifying on nearby material (i.e. the lamp quartz) which in the case of handling simplifies the task as well as having a lower proportion in the vapour phase compared to liquid mercury which is highly advantageous. System pipework design will often

Table 5 Elemental information relating to toxicity adapted from CDC (2018)

	Boiling Point (°C)	Vapour pressure	Solubility	Exposure limits	IDLH (Air)
Mercury	357	0.0012 mmHg	Insoluble	Hg Vapour: TWA 0.05 mg/m^3 [skin] Other: Concentration in Air 0.1 mg/m^3 [skin]	10 mg/m^3
Iodine	185	(25°C): 0.3 mmHg	0.01%	NIOSH REL Concentration in Air: 0.1 ppm (1 mg/m^3)	20.76 mg/m^3
Tellurium	990	~0 mmHg	Insoluble	NIOSH REL TWA 0.1 mg/m^3	25 mg/m^3

(IDLH = Immediately Dangerous to Life or Health).

enable debris collection either through a filter (post UV system) or a collection point within the pipework. Having the material attached in the case of tellurium rather than free liquid mercury would be far more desirable in such a process. In regard to vapour inhalation both liquid mercury and iodine need to be considered and treated with care during handling. However, as can be seen in Table 5, the Immediately Dangerous to Life or Health (IDLH) figures are over twice that for both iodine and tellurium compared to mercury and the wide use of iodine suggests a preferable handling quality to that of mercury.

The extensive practical use of iodine and the high vapourisation temperature of tellurium the lamp filling as a functional alterative to mercury is worth considering for further studies for a conventional HP plasma design. Following design improvements, a full design comparison should be undertaken to assess the suitability of the presented technology, however, initial performance suggests that it may well match a market need and application.

Conclusions

A Hg-free HP plasma concept primarily using tellurium was proposed and assessed with the following findings:

(1) The plasma as a proof of concept functioned well albeit with some instability and lower UV levels than predicted.

(2) The tellurium-based UV radiation source, which if continued to be developed could have significant and far-reaching effects on the industry due to the demand to reduce or remove the use of mercury.

(3) A developed lamp could provide a much-needed tool for UV disinfection applications that require a mercury-free source and high levels of UV radiation, benefiting from the commonality of current lamp geometry in respect to UV system design and validation to enable rapid application of the technology.

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