

24 with DSA anodes opens the door to novel efficient disinfection processes, limiting the
25 occurrence of hazardous disinfection by-products.

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35 **Keywords:** Ultrasound, dimensionally stable anode (DSA), electrodisinfection,
36 wastewater reclamation, integrated process.

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47 **1. Introduction.**

48 In recent years, electrochemical disinfection has attracted the interest of several
49 researchers, being the main target the development of economic and safe disinfection
50 technologies which prevent the occurrence of hazardous disinfection by-products
51 (Cotillas et al., 2015; Haaken et al., 2014; Mezule et al., 2013). One of the milestones of
52 these studies has been the applicability of diamond (BDD) anodes for the treatment of
53 synthetic wastewater polluted with *Escherichia coli* (*E. coli*) (Cui et al., 2013; Gusmão
54 et al., 2010; Lacasa et al., 2013; Li et al., 2010). These anodes are classified into the
55 group of non-active electrodes and exhibit high performance in disinfection and in the
56 removal of several type of pollutants (Garcia-Segura et al., 2015; Marselli et al., 2003;
57 Martínez-Huitle et al., 2012; Panizza, 2014).

58 Unfortunately, the electrochemically-assisted disinfection with BDD anodes favours the
59 generation of chlorates and perchlorates (Bergmann, 2010; Bergmann and Rollin, 2007;
60 Vacca et al., 2011) which are known to be harmful for human health. This fact limits the
61 maximum value of current density and electric charge applicable in disinfection
62 processes (Llanos et al., 2014). Opposite to electrolysis with diamond coatings,
63 production of chlorate and perchlorate is expected to be limited with the well-known
64 Dimensionally Stable Anodes (DSA) based on RuO₂, widely used in chlor-alkali
65 industry for the production of hypochlorite from concentrated brines (Kiros et al., 2006;
66 Mohaddes, 2008).

67 Another novel technology that may exhibit good features for disinfection of urban
68 wastewaters is the application of ultrasound irradiation (Antoniadis et al., 2007).
69 Disinfection can be achieved by different mechanisms: 1) the production of free radicals
70 in the bulk (Juretic et al., 2015); 2) shearing forces produced by ultrasonic cavitation
71 (Drakopoulou et al., 2009); 3) breakage of bacteria agglomeration, what favours the

72 reaction between oxidants species and microorganisms and therefore, the disinfection
73 processes (Hughes and Nyborg, 1962).

74 In this context, in a previous research it was found that ultrasonic cavitation
75 dramatically increases the efficiency of the inactivation of *E. coli* working with BDD
76 anodes and low current densities (in order to prevent the formation of chlorate and
77 perchlorate) (Llanos et al., 2015). This improved performance was explained in terms of
78 a more efficient production of oxidants and to the promotion of the dispersion of
79 agglomerated cells, favouring the attack to microorganisms.

80 Thus, the aim of the present work is to study the potential synergistic effect of coupling
81 ultrasound irradiation and electrolysis with DSA anodes to the disinfection of actual
82 effluents from municipal wastewater treatment plants (WWTP). This study would
83 widen the application of the combined sono-electrodisinfection process to higher values
84 of current densities and applied electric charges, limiting the formation of hazardous
85 disinfection by-products as chlorate. In a first approach, the efficiency of single
86 sonodisinfection and electrodisinfection processes was studied, evaluating the influence
87 of the applied ultrasound power and current density on the process performance. Next,
88 US irradiation was coupled with the electrochemical disinfection with DSA anode in
89 order to evaluate the potential synergies of both techniques.

90

91 **2. Material and methods.**

92 **2.1 Analytical techniques.**

93 Faecal coliforms were estimated using the most probable number (MPN) technique
94 (APHA-AWWA-WPCF, 1998). Microorganism counts were carried out by the
95 multiple-tube-fermentation technique (24 h of incubation at 44 °C) using 5 tubes at each
96 dilution (1:10, 1:100, and 1:1000).

97 Nitrogen and chloride inorganic anions (NO_3^- , NO_2^- , Cl^- , ClO^- , ClO_2^- , ClO_3^- , ClO_4^-)
98 were measured by ion chromatography using a Shimadzu LC-20A equipped with
99 Shodex IC I-524A column; mobile phase, 2.5 mM phthalic acid at pH 4.0; flow rate, 1.0
100 mL min^{-1}). The peak of hypochlorite interferes with that of chloride; therefore, the
101 determination of hypochlorite was carried out by titration with As_2O_3 in 2 M NaOH.
102 The same ion chromatography equipment (Shodex IC YK-421 column; mobile phase,
103 5.0 mM tartaric, 1.0 mM dipicolinic acid and 24.3 mM boric acid; flow rate, 1.0 mL
104 min^{-1}) was used to measure the nitrogen inorganic cation (NH_4^+).

105 Inorganic chloramines were measured following the DPD standard method described in
106 the literature (APHA-AWWA-WPCF, 1998).

107 Occurrence of trihalomethanes (CHCl_3 , CHBrCl_2 , CHBr_2Cl , CHBr_3) was followed by
108 gas chromatography with an electron capture detector (ECD). The column used was a
109 SPB 10 column (30 m x 0.25 mm; macroporous particles with 0.25 μm diameter). The
110 flow rate was 50 ml min^{-1} and the initial temperature was 50°C during 5 minutes until
111 reach a final value of 150°C (6°C min^{-1}). Injection volume was set to 1 μL during 5
112 minutes and hydrogen was used as carrier gas.

113 **2.2 Experimental setup.**

114 The sonodisinfection, electrodisinfection and sono-electrodisinfection processes were
115 carried out in a single-compartment electrochemical cell. Dimensionally Stable Anode
116 (DSA) based on RuO_2 (Tianode, India) was used as anode and stainless steel (SS) (AISI
117 304) (Mervilab, Spain) as cathode. Both electrodes (anode and cathode) were circular
118 with a geometric area of 78.5 cm^2 and the electrode gap was 10 mm.

119 Wastewater was stored in a glass tank (2.5 dm^3). The ultrasound generator (UP200S,
120 Hielscher Ultrasonics GmbH, Germany) is equipped with a horn (40 mm diameter, 100

121 mm length, 12 Wcm^{-2} , 24 kHz) that is located inside the glass tank. The maximum
122 power of ultrasound irradiated is 200 W and the output can be continuous or pulsed with
123 duty cycle ranging from 10 to 100%.

124 The system works in total recirculation mode, with a peristaltic pump (JP Selecta
125 Percom N-M328) continuously recycling the target wastewater. The power supply is a
126 Delta Electronika ES030-10. Temperature of the system is kept constant by means of a
127 thermostatised bath (JP Selecta, Digiterm 100) and a heat exchanger. A schematic of the
128 experimental set-up has been described elsewhere (Llanos et al., 2015).

129 **2.3. Experimental procedure.**

130 Bench-scale electrolyses and sonoelectrolyses of $2,000 \text{ cm}^3$ of wastewater were carried
131 out under galvanostatic conditions. The current density applied ranged from $6.37\text{--}11.46$
132 A m^{-2} . The cell voltage did not vary during electrolysis, indicating that RuO_2 layers did
133 not undergo appreciable deterioration or passivation phenomena. Prior to use in
134 galvanostatic electrolysis assays, the electrode was polarized for 10 min in a $5,000 \text{ mg}$
135 $\text{dm}^{-3} \text{Na}_2\text{SO}_4$ solution at 150 A m^{-2} to remove any kind of impurity from its surface.

136 Sonochemical disinfection of $2,000 \text{ cm}^3$ was carried out at ultrasound power ranged
137 from 0-200 w.

138 **2.4. Target effluents.**

139 The target wastewater used in this study is the effluent of the secondary clarifiers of the
140 municipal WWTP of Ciudad Real (Spain). The WWTP treats the wastewater produced
141 in an average-sized town (75,000 p.e.) located in the centre of Spain. The influent is
142 domestic wastewater without a significant industrial contribution. However, the effluent
143 obtained after the treatment presents high concentration of microorganisms and prevents

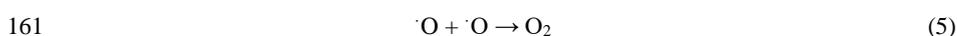
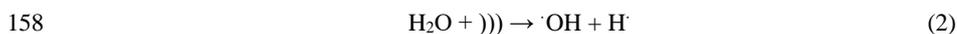
144 the direct reuse of treated wastewater. The average characteristics of the samples used in
145 this work are shown in Table 1.

146

147 3. Results and discussion.

148 3.1. Single sonochemical and electrochemical disinfection processes for the 149 treatment of urban treated wastewater.

150 Fig. 1a shows the changes in concentration of *E. coli* with the irradiation time during
151 several sonochemical disinfection tests, in which different US power are applied to
152 wastewater. As it can be observed, the concentration of microorganisms decreases with
153 irradiation time until it reaches a final value from which it remains constant. This fact
154 has to be explained in terms of the different processes that occur when ultrasound waves
155 come into contact with a liquid, in particular: 1) shear forces generated for ultrasonic
156 cavitation; 2) generation of free radicals or hydrogen peroxide (Eqs. 2-5) (Adewuyi,
157 2005; Antoniadis et al., 2007).



162 The occurrence of these processes depends on the frequency of the ultrasonic radiation
163 applied. In this context, ultrasound irradiated is typically classified in two general
164 groups: low frequency (20-100 kHz) and high frequency (100 kHz-10 MHz). The main
165 difference between these two groups is related to the production of radicals. At low
166 ultrasound frequencies, the bubbles formed are mainly bigger and their implosion

167 occurs after some cycles of compression/expansion. This behaviour could facilitate the
168 recombination of the radicals formed (Eqs. 3-5) and therefore, it could hinder their
169 transference to the liquid. On the other hand, when the frequency is increased, the
170 generation of free radicals can be favoured due to the short time of the expansion step
171 (Thompson and Doraiswamy, 1999). Hence, high frequency UV irradiation is much
172 more efficient than low frequency. The frequency of the ultrasound used in this work is
173 24 kHz (low frequency). Thus, the decrease observed in the concentration of *E. coli*
174 should be mainly related to the shear forces produced from the collapse of bubbles
175 during the cavitation and to the recombination of hydroxyl radicals to form hydrogen
176 peroxide, rather than to the direct action of the radicals formed.
177 Furthermore, it can be observed that the higher the US power irradiated, the higher the
178 inactivation of microorganisms. In order to evaluate the rate of the process, the data
179 obtained in the first stage (corresponding to the decrease in the *E. Coli* population)
180 were fitted to a first order kinetics (Eq. 6). The disinfection kinetic constants calculated
181 in this stage are shown in Fig. 1b.

$$\text{Ln} (E. coli / E. coli_0) = - k \cdot t \quad (6)$$

182
183 As it can be seen, the disinfection rate increases with the ultrasound power (a trend
184 previously described in literature (Antoniadis et al., 2007)), reaching a maximum value
185 of 0.016 min⁻¹ for a power of 200 W (maximum value supplied by the equipment used).
186 However, results indicate that there is a recalcitrant amount of *E. coli* that is not
187 inactivated by the application of US irradiation. This result does not agree with others
188 published in literature that reported a high efficiency in the inactivation of faecal
189 coliforms by US irradiation (Drakopoulou et al., 2009). Nevertheless, the low specific
190 US power irradiated in the present work (1,500 W L⁻¹ in Drakopoulou et al. vs. 100 W
191 L⁻¹ in the present work) help to explain this residual concentration of *E. coli* that is not

Comentado [MARR1]: Supongo que lo habréis sacado de algún sitio. La realidad es que la energía que se transmite y llega a especies que pueden ser precursores de radicales no es suficiente como para romper las moléculas

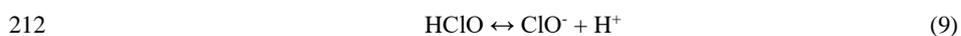
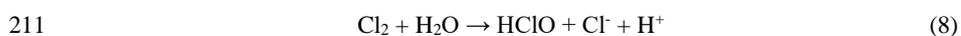
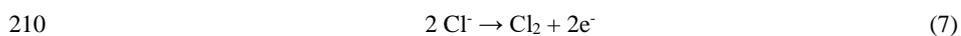
Comentado [MARR2]: Hubieses estado bien medir oxidantes para ver si hay agua oxigenada

192 inactivated by the application of US irradiation. At this point, it is important to take in
193 mind that even the energy applied in our work is huge for an efficient treatment and
194 hence lower doses of US should be applied in order to study feasible technologies.
195 Finally, it is important to note that the pH was remained constant during the
196 sonochemical process at a value around 8.

197 The next step of this research consisted in the study of electrochemical oxidation with
198 DSA anodes of the same target wastewater. Fig. 2a shows the changes in the
199 concentration of *E. coli* with the applied electric charge during the electrodisinfection
200 tests carried out at different current densities.

201 As it can be observed, the concentration of microorganisms decreases with the applied
202 electric charge, reaching the total disinfection of the effluent (within the duration of the
203 test) only at the highest current density evaluated (11.46 A m⁻²). Fig. 2b shows the
204 disinfection constants obtained when experimental data are fitted to a first order model
205 (Eq. 6).

206 The disinfection rate increases exponentially. The most probable mechanism of
207 disinfection is based on the action of disinfectant species electrochemically generated
208 from the ions naturally contained in wastewater (Cotillas et al., 2015), mainly chlorine
209 species as hypochlorite (Eqs. 7-9) (Cotillas et al., 2013; Lacasa et al., 2012b).



213 In this context, Fig. 3a shows the concentration of free chlorine with the applied electric
214 charge during the electrodisinfection at different current densities. As it can be
215 observed, hypochlorite concentration increases with the applied electric charge due to
216 the electrooxidation of chloride. At low current densities (6.37 A m⁻²), this species

Comentado [MARR3]: Solo hay tres puntos!!!!. No se sabe que ocurre a j mayores a esta. Es especulación

Comentado [MARR5]: Esto, conjuntamente con lo de las cloraminas se conto en lo de anaid

217 increases until reaching a final value around 0.04 mmol Cl dm⁻³. However, from current
218 densities of 8.91 A m⁻², hypochlorite behaves as a reaction intermediate, with maximum
219 concentrations of 0.024 and 0.058 mmol Cl dm⁻³ at 8.91 and 11.46 A m⁻², respectively.
220 Hypochlorite generated in the cell can be involved in different reaction pathways. First,
221 it can inactivate *E. coli* cells resulting in the disinfection of the effluent. In addition, the
222 reaction of hypochlorite with ammonium may lead to the production of combined
223 chlorine compounds (Eqs. 10-12). Although the initial concentration of ammonium is
224 low, it is important to highlight that the electrochemical reduction of nitrates over
225 stainless steel cathodes is a very efficient process which develops at a high rate during
226 the electrolysis of nitrate-containing solutions like the wastewater samples used in this
227 work (Lacasa et al., 2012a). This reduction produces the source of ammonium for the
228 production of chloramines and, obviously, its effect is expected to be higher at higher
229 current densities.



233 Fig. 3b shows the concentration of chloramines as a function of the current density
234 applied during the electrodisinfection tests. As expected, chloramines increase with the
235 applied electric charge but different trends can be described. At low values (6.37 A m⁻²),
236 the formation of chloramines is still not heavily favoured, reaching a maximum final
237 value of 0.004 mmol Cl dm⁻³. However, when current density increases above values of
238 8.91 A m⁻², higher concentrations of chloramines are obtained due to the reaction
239 between hypochlorite and ammonium (j = 8.91 A m⁻², chloramines = 0.037 mmol Cl
240 dm⁻³; j = 11.46 A m⁻², chloramines = 0.060 mmol Cl dm⁻³).

241 Chlorine species at higher oxidation state (chlorite, chlorate and perchlorate) were not
242 detected at the current densities studied. This fact was somehow expected because
243 electrolysis with DSA anodes requires current densities higher than that used in this
244 work to promote this chlorine speciation (Neodo et al., 2012).

245 According to the data obtained during chlorine speciation, it is important to highlight
246 that it is required concentrations of free and combined chlorine species higher than 0.05
247 mmol Cl dm⁻³ and current densities of 11.46 A m⁻² to reach the total disinfection of
248 urban wastewater within the reaction time used in this work (XX minutes). Likewise,
249 pH remained constant in values around 8, favouring the formation of hypochlorite anion
250 and chloramines (mainly **monochloramine**).

251 Regarding the formation of trihalomethanes during the electrodisinfection (data not
252 shown), it has been found a negligible concentration of these species for all the current
253 densities studied. The maximum concentration generated was 0.1832 μmol dm⁻³ at the
254 highest current density. This concentration is far below the most restrictive values found
255 in environmental **regulations**.

Comentado [MARR6]: Las líneas de tendencia de las cloraminas son muy feas. Probablemente se deban al efecto envolvente de las tres cloraminas. Esto hay que ponerlo y hacer un línea más bonita en el gráfico

256 **3.2. Integrated sonoelectrodisinfection process for the treatment of urban treated** 257 **wastewater.**

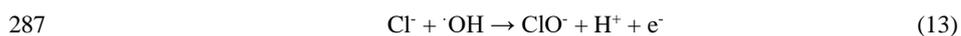
258 Once the sonodisinfection and electrodisinfection processes were assessed for the
259 inactivation of microorganisms, the goal of this section is to evaluate an integrated
260 sono-electrodisinfection process. The current density chosen to evaluate the
261 performance of this coupled process was 8.91 A m⁻², a value at which the complete
262 disinfection was not reached with the electrodisinfection process during the length of
263 the tests carried out. Fig. 4 shows the changes in the concentration of *E. coli* during the
264 sono-electrodisinfection process at increasing US power applied (8-200 W).

Comentado [MARR7]: Comentar valores

265 As it can be observed, the effluent is efficiently disinfected during the duration of the
266 test (xx minutes) when applying ultrasound power over 32 W and E. Coli is completely
267 depleted. Likewise, it is worth noting that it is possible to carry out the disinfection of
268 urban wastewater at lower current densities (8.91 A m^{-2}) than those necessary during the
269 electrochemical process (11.46 A m^{-2}) for the complete removal of pathogens within the
270 test.

271 As in the single electrodisinfection process, free and combined chlorine compounds
272 (represented in Fig. 5) play a key role in the disinfection process.

273 As it can be observed, hypochlorite (Fig. 5a) behaves as a reaction intermediate, as it
274 was the case of electrodisinfection process. When profiles of hypochlorite generation of
275 electro- and sonoelectro- disinfection processes are compared, it can be observed that
276 the irradiation of US increases the production of hypochlorite. Nevertheless, the
277 maximum concentration of hypochlorite follows two different trends depending on the
278 range of US power applied. Within the range 8-72 W, the maximum concentration of
279 hypochlorite increases with US power. On the contrary, from 72 W to 200 W, the
280 maximum concentration detected decreases with US power applied. This dissimilar
281 evolution of the detected hypochlorite can be explained by considering the two
282 overlapped effects that can be observed when US irradiation and electrolysis processes
283 are integrated. Thus, on the one hand, it has been described that coupling both
284 techniques leads to an enhanced production of oxidants due to the generation of
285 hydroxyl radicals by water sonolysis (Eqs. 1-2), as it is the case of the production of
286 hypochlorite from chloride according to Eq. 13 (Souza et al., 2014).



288 Furthermore, it has been described that the application of US can suppress the
289 agglomeration of microorganisms and thus increases the disinfection rate by an

Comentado [MARR8]: No estoy seguro. A 24 khz no es cierto. Si podría ser descomposición de agua oxigenada generada en el catodo

290 enhanced mass transfer of disinfectant species to *E. coli* cells (Jin et al., 2013; Llanos et
291 al., 2015). As commented previously, one of the main routes of hypochlorite reduction
292 is the disinfection process itself, so an increased mass transfer (higher US power
293 applied) can lead to a lower concentration of hypochlorite detected. In this case, the
294 lower hypochlorite concentration measured is due to the higher rate of hypochlorite
295 reduction and not to a lower formation rate.

296 On the other hand, the concentration of chloramines (Fig. 5b) increases with the applied
297 electric charge and with ultrasound irradiated. This increase is explained by the higher
298 hypochlorite concentration and by the enhanced mass transfer, which facilitate reactions
299 of chloramines formation (Eqs. 10-12). In addition, it is worth mentioning that chlorate
300 and perchlorate were not detected, so it can be stated that the production of these
301 hazardous disinfection by-products is avoided by the use of DSA anodes.

302 Finally, a synergy coefficient (%) is used to evaluate the rate increase when coupling
303 both processes. The calculation of this synergy coefficient is based on the constant rates
304 previously calculated in the sono- (SD), electro- (ED) and sonoelectro- (SED)
305 disinfection processes (Eq. 14).

$$306 \quad \text{Synergy coefficient (\%)} = \frac{k_{SED} - k_{SD} - k_{ED}}{k_{SD} + k_{ED}} \cdot 100 \quad (14)$$

307 These synergy coefficients are gathered in Fig. 6. As it can be observed, the synergy
308 coefficient increases for higher US power applied, reaching a maximum value of 236%
309 for a US power of 200 W. These synergistic effects are partly explained by the higher
310 amount of hypochlorite measured in the combined process. Furthermore, as previously
311 commented the application of US irradiation produces the suppression of agglomeration
312 of *E. coli* cells in wastewater (Hughes and Nyborg, 1962), enhancing the attack of
313 disinfectants to microorganisms. This synergistic effect was described in a previous

314 work in which sono-electrolysis with diamond electrodes was assessed (Llanos et al.,
315 2015), although the constant rates obtained with that anode material were higher to
316 those measured for DSA. However, it is worth noting the importance of confirming this
317 behaviour with active anodes as commercial DSA, with which it would be possible to
318 operate at higher current densities without the generation of hazardous chlorine
319 compounds such as chlorate or perchlorate. This fact, together with the lower cost of
320 DSA, would lead to compact, economic and smart devices with improved disinfection
321 capacity.

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325 **4. Conclusions**

326 The main conclusions that can be drawn from the present manuscript are:

327 -The disinfection rate of a sonochemical process increases with ultrasound power
328 irradiated. However, it is not possible to achieve the complete disinfection of the
329 effluent in the range of US power applied in the present work.

330 - Electrochemical disinfection with DSA anodes is an efficient technology to inactivate
331 the microorganisms in wastewater. It is necessary a minimum current density and
332 applied electric charge to completely disinfect the effluent.

333 - Synergies higher than 200% of disinfection rate are found when coupling ultrasound
334 irradiation and electrodisinfection with DSA anodes. Chlorate and perchlorate were not
335 detected, being hypochlorite and chloramines the main responsible of the disinfection
336 process.

337 - Ultrasound enhances mass transfer processes and therefore, it improves the production
338 of disinfectant species in the bulk. Likewise, ultrasound favours the suppression of the
339 agglomeration of *E. coli* cells in the bulk, allowing an efficient attack of disinfectant
340 species to microorganisms. The result obtained allow designing efficient disinfection
341 devices that avoid the production or hazardous disinfection by-products.

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346 Regeneration. The authors are also grateful to “Departamento Administrativo de
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