Design of the Photovoltaic Solar Electro-Oxidation (PSEO) process for wastewater treatment

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A B S T R A C T

The Photovoltaic Solar Electro-Oxidation (PSEO) process combines the effectiveness of the electrochemical oxidation based on boron-doped anodes to mineralize organic matter, with the autonomy and environmentally friendly characteristics of photovoltaic solar energy. The PSEO process constitutes an attractive technology for the treatment of urban wastewaters of isolated communities where standard solutions cannot be applied. The aim of this work is the design of this process, which implies the sizing of the electrochemical reactor and the photovoltaic modules. The input variables for the design are the characteristics of the effluent, the wastewater flow rate (which depends on the number of equivalent inhabitants for whom the plant operates), the required yield and the solar irradiance. The solar irradiance depends on the location in which the PSEO process is applied, since differences in the required PV module area higher than 170% are obtained in several locations in Spain. Moreover, the residence time distribution in the reactor also significantly affects the dimensions of the electrode (A_a) and solar module (NA_{PV}) areas.

1. Introduction

Nowadays in the European Union, urban wastewaters must be collected and adequately treated even in villages with fewer than 2000 equivalent inhabitants, according to Directive 91/271/EEC (EC, 1991). However, in isolated communities, the wastewater treatments used in large cities are not usually applicable, due to technical and financial limitations. Innovative technologies based on small-scale treatment systems can be adapted to the needs of the isolated areas (Fahd et al., 2007).

The Photovoltaic Solar Electro-Oxidation (PSEO) process appears as an attractive alternative to treat the urban wastewaters of isolated communities. On the one hand, electrochemical oxidation based on boron-doped diamond (BDD) is an advanced technology which permits an efficient removal of organic matter of very different nature and origin, without the generation of sludge or the consumption of chemicals, which are characteristic of different conventional treatments (Dominguez-Ramos et al., 2010). The main drawback of electrochemical oxidation is its high investment and

operational (due to energy consumption) costs in comparison with conventional technologies, such as activated-sludge processes. However, electro-oxidation with BDD electrodes provides higher pollutant removal and it can compete satisfactorily with other advanced oxidation processes such as Fenton oxidation or ozonation in the treatment of several types of wastewaters in terms of operation and investment costs (Anglada et al., 2010; Cañizares et al., 2009). This technology is also characterized by simple equipment, easy operation and versatility (Anglada et al., 2009), especially when filter-press reactors are considered, due to their simple and widely used design (Chen, 2004; Martínez-Huitle and Brillas, 2009).

On the other hand, the use of a renewable source of energy such as photovoltaic (PV) solar energy contributes to the reduction of the economic and environmental impact associated with the high energy demand of electrochemical oxidation (Panizza et al., 2001). This source of energy also provides autonomy for the electro-oxidation, since it does not require any connection to the grid, which constitutes an important advantage in isolated rural areas. The PSEO process establishes a direct integration of the PV modules to the electrochemical reactors so that energy storage systems are

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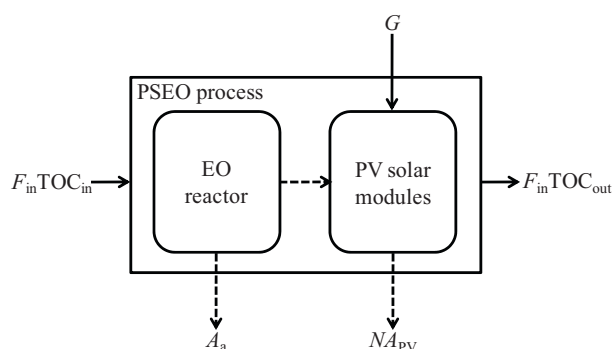 E-mail address: alvarezge@unican.es (E. Alvarez-Guerra).

Fig. 1 – Schematic diagram of the PSEO process.

no longer required. The use of batteries introduces several problems, like the loss of energy in charge–discharge cycles (efficiency is around 70–80%), their short lifetime, the control of the state of the charge that must be implemented, the investment and operating costs of this equipment, as well as the environmental impact of battery disposal (Thomson and Infield, 2005; Valero et al., 2010). The technical feasibility of electrochemical oxidation processes carried out with PV modules directly connected to the electrochemical reactor has been demonstrated in several examples that can be found in the literature (Ochiai et al., 2010; Park et al., 2008; Valero et al., 2010).

The aim of this paper is the design of the PSEO process for wastewater treatment. The sizing of the main equipment of the process (electrochemical reactor and PV modules as required anodic area and effective area, respectively) is carried out. An analysis of the variables that affect the design is showed. Special attention is paid to the geographical location of the plant to treat urban wastewater, which showed noticeable influence in the required areas, so location constitutes one of the main reasons that justify the application of the PSEO process.

2. Conceptual design

The schematic diagram of the PSEO process is shown in Fig. 1. The electric power in the electrochemical reactors is delivered by a suitable PV solid-state energy conversion device such as PV solar modules. Therefore, this electric current is used by electrochemical reactors to treat the wastewater effluent.

The conversion of luminous energy into electric energy is described by means of the following equation that represents the performance of the PV modules (Alvarez-Guerra et al., 2011):

$$I = \alpha NA_{PV} G \tag{1}$$

where I is the total applied electric current, G is the incident solar irradiance and NA_{PV} is the total effective area of the modules (N is the number of available modules and A_{PV} the effective area of an individual selected module). G is a variable that changes during the day and throughout the year, that is, it is a time-dependent function. The linear coefficient, α, is obtained as the quotient of the short-circuit current of the module (I_{sc}) and the product of A_{PV} and the solar irradiance in standard reference conditions (G_{ref}).

A conventional I–V diagram of a PV solar module at a fixed G should exhibit a region, below maximum power volt-

age, in which voltage between terminals, V, has a negligible influence over I. Eq. (1) models the PV module performance under the operation in this region (Dominguez-Ramos et al., 2010) and it allows the estimation of the electric current in a given PV system, which implies that NA_{PV} and G are known.

Regarding the voltage established between the anodes and cathodes of each electrochemical reactor, V, and therefore, voltage between terminals of the PV modules due to their direct connection, it is modeled by the following expression (Chen, 2004):

$$V = a + b \cdot \log(i) + c \cdot i \tag{2}$$

where i is the applied current density. Coefficients a and b are related to the equilibrium potentials and overpotentials of the electrodes, whereas c represents the ohmic term and it is dependent on the effluent conductivity, λ (Alfafara et al., 2004). In this way, when the conductivity is relatively low, the electric resistance of the effluent increases and, consequently, the value of c also does.

A minimum voltage to each cell, V_{min}, has to be established in order to assure that the desired oxidation reactions with participation of hydroxyl radicals take place at the anodic surface. In this way, if a value of V_{min} is selected, the current density, i, is also fixed by Eq. (2), for a given effluent conductivity, which has an influence on V–i relationship by means of c. Once I and i are known, the required anodic area, A_a, can be calculated by means of the following equation:

$$I = A_a i \tag{3}$$

On the other hand, the organic matter mineralization in the PSEO process has been described by means of a kinetic equation which states a first-order dependence regarding the concentration of organic matter expressed as total organic carbon, TOC (Alvarez-Guerra et al., 2011):

$$(-r_{TOC}) = -\frac{V_s}{A_a}\frac{dTOC}{dt} = kTOC \tag{4}$$

where k is the rate constant and V_s is the effluent volume treated in the electrochemical system. The rate constant, k, includes both the $V_s \cdot A_a^{-1}$ ratio (m) and the first order kinetic constant of the TOC mineralization process (min^{-1}) obtained under batch operation. Eq. (4) can be applied when the TOC$\cdot i^{-1}$ ratio is relatively low, so the current efficiency, η_c, is kept below 100% (Boye et al., 2006; Choi et al., 2010; Panizza and Cerisola, 2010). In this work, the current efficiency is defined as follows:

$$\eta_c = \frac{(-r_{TOC})}{(-r_{max})} = \frac{kTOC}{i/nF} \tag{5}$$

where $(-r_{max})$ is the maximum reaction rate that can be achieved with a given current density; n, the number of electrons involved in the oxidation reaction; and F, the Faraday constant (96,485 C mol^{-1}).

Despite the fact that the mineralization rate is not dependent on the current density, mass balance is related to the energy conversion due to the influence that the restriction of the minimum voltage has in A_a. A minimum G that allows establishing V_{min} is required to oxidize the wastewater pollutants.

The kinetic equation is required to apply the mass balance. This balance relates the TOC of the outlet effluent (TOC$_{out}$) to
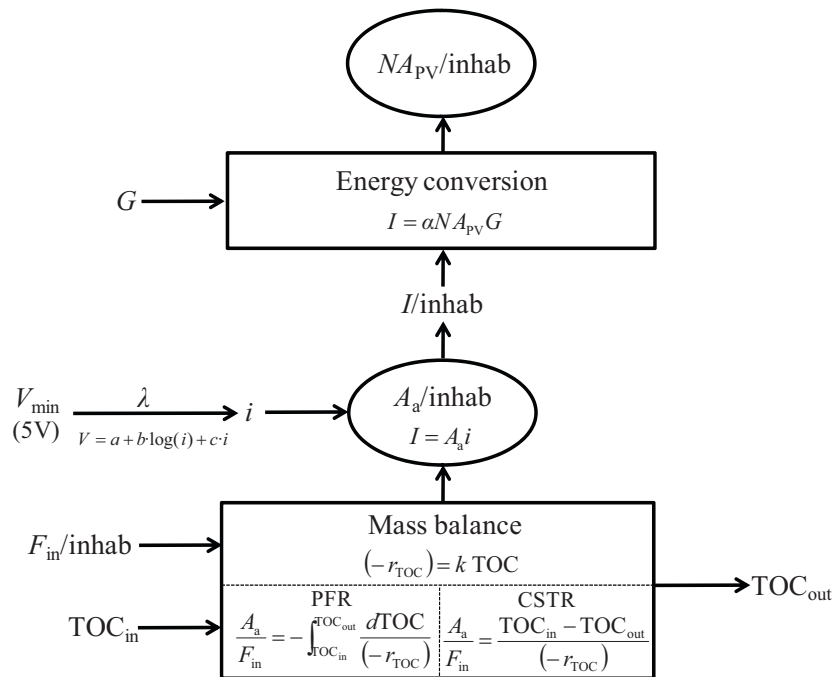
Fig. 2 – **Flow diagram for the PSEO process design.**

the inlet TOC (TOC_{in}), the wastewater flow rate (F_{in}) and the anodic area (A_a). F_{in} represents the total wastewater flow rate that enters into the process, whereas the flow rate through each electrochemical cell is not considered as a variable since there is an optimal flow rate recommended by the manufacturer of the electrochemical cell.

The PSEO process design involves the sizing of the main items of equipment required to perform the treatment. The main items are the electrochemical reactors (considering anodic area) and the power supply (Cañizares et al., 2009). In this case, the device for power supplying is the set of PV modules (expressed as effective PV module area), with which the electric energy that electrochemical cells demand is generated. Fig. 2 represents the flow diagram of the PSEO process. Therefore, in the design process, NA_{PV} and A_a must be the output variables, so the treatment efficiency expressed as TOC removal has to be fixed by means of TOC_{out}. The daily mean wastewater generated by one inhabitant is selected as the flow rate to be treated, so the equipment sizes represent the values of NA_{PV} and A_a demanded by each inhabitant.

As a consequence, the input and output variables, parameters and operational constraints for designing a PSEO installation are:

- Input variables: G, F_{in}, TOC_{in}, TOC_{out}.
- Output variables: NA_{PV}, A_a.
- Model parameters: α, a, b, c, k.
- Operational constraints: V_{min}.

The parameter α is defined as a function of data provided by manufacturers of photovoltaic modules, so it can be obtained theoretically. On the other hand, a, b, c and k must be determined experimentally: a, b and c by means of cell voltage–current density data fitting; and k by means of $(-r_{TOC}) - TOC$ data fitting (Alvarez-Guerra et al., 2011).

3. Model development

Once the required equations, variables and parameters of the PSEO process have been stated in Section 2, the way in which they are used for the design is explained below.

Regarding the operational constraint that V_{min} represents, the minimum voltage at which the electrochemical oxidation has been carried out in the experiments reported by Alvarez-Guerra et al. (2011) is fixed at 5 V. However, in the experiments in which energy was supplied by the photovoltaic modules, the mineralization rate was almost negligible when irradiance was relatively low, so $V < 5$ V. In this way, the same conclusion is derived from the experimental data showed by Dominguez-Ramos et al. (2010), since data at $V < 5$ V corresponds to the periods of time in which the mineralization rate was near zero (the TOC-time profile remained practically constant). Moreover, this value is in accordance with the cell voltages stated by Dominguez-Ramos et al. (2008) and Zhu et al. (2009), for a similar experimental set-up and experimental conditions, so 5 V is selected as V_{min}. According to Eq. (2), this constraint is equivalent to imposing a minimum current density, i_{min}, which also implies that G must exceed a minimum value, G_{min}, to become the PSEO installation operational:

$$G_{min} = \frac{i_{min}(V_{min})}{\alpha} \frac{A_a}{NA_{PV}} \qquad (6)$$

For a given PSEO installation (A_a and NA_{PV} are fixed), the plant could not treat the effluents during the periods of time in which solar irradiance, G, does not reach the value of G_{min}.

Eq. (6) states an inverse relationship between the $(NA_{PV}) \cdot A_a^{-1}$ ratio and G_{min}. Therefore, the $(NA_{PV}) \cdot A_a^{-1} - G_{min}$ relationship is defined by a hyperbolic function. This fact implies that the total applied current I necessary to reach V_{min} (so the PSEO installation becomes operational) can be obtained with multiple combinations of different minimum solar irradiations, G_{min}, and PV modules areas, NA_{PV}. When solar irradiances are relatively high, a lower number of mod-
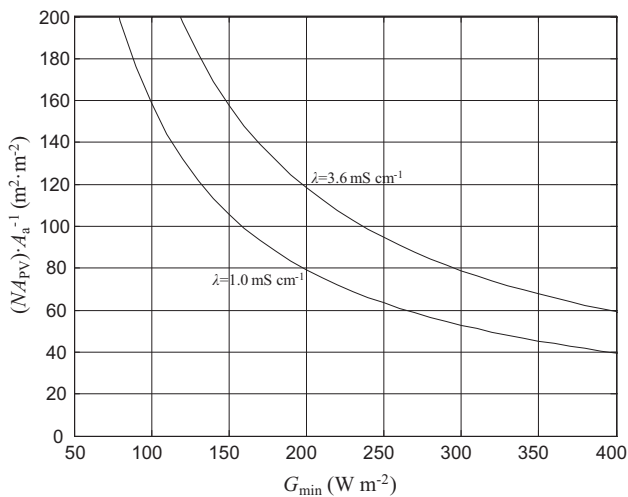
Fig. 3 – Relationship between $(NA_{PV})\cdot A_a^{-1}$ and G_{min} for effluents with different conductivity, λ, and $\alpha = 4.44 \times 10^{-3}\,A\,W^{-1}$.

ules will be necessary to generate the electric power that electrochemical reactors demand to operate at V_{min}. At low irradiances, the $(NA_{PV})\cdot A_a^{-1}$ ratio is very sensitive to G_{min} so slight changes in the latter variable lead to very significant variations in the ratio of areas. On the other hand, when relatively high solar irradiations are chosen so that the plant becomes operational, relevant changes in G_{min} are required to modify $(NA_{PV})\cdot A_a^{-1}$ notably.

Furthermore, the relationship between $(NA_{PV})\cdot A_a^{-1}$ and G_{min} is influenced by the effluent conductivity. When this conductivity is relatively low, the effluent resistance (described by the parameter c) increases and permits lower current densities than those applied with high conductivity wastewaters to establish the minimum cell voltage. Consequently, in Fig. 3 the line which corresponds to the effluent with high conductivity ($3.6\,mS\,cm^{-1}$) is above the line of low conductivity ($1.0\,mS\,cm^{-1}$), since in the first case a higher $(NA_{PV})\cdot A_a^{-1}$ ratio is needed to supply the current density which allows electrochemical cells to operate at V_{min}, for a given G_{min}. This fact is logical as a given voltage between electrodes can be obtained with different combinations of high ohmic resistance and low applied current. Attention should be given to the fact that the current density obtained leads to η_c below 100% so that the described model equations could be applied.

Combining Eqs. (1) and (3), the area of the PV modules, NA_{PV}, is calculated as a function of A_a, G and i:

$$NA_{PV} = \frac{i}{\alpha G} A_a \qquad (7)$$

The relationship between NA_{PV} and A_a is shown in Fig. 4 for an effluent with low conductivity ($\lambda = 1.0\,mS\,cm^{-1}$). This linear dependence can be explained considering that greater anodic areas demands more applied electric current whenever current density remains constant. This higher electric current consumption must be satisfied by means of a higher number of PV modules, if G is considered constant. On the other hand, for a given A_a imposed by the mass balance, the solar irradiance, G, determines NA_{PV} (Eq. (7)), because higher G makes it possible to provide the same electric current with lower NA_{PV}.

Even though G has a variation during the day, the stand-alone PV system sizing methodologies use daily mean irradiations as values of G, so this input to the design (Fig. 2) is therefore considered constant for each case. Its value is
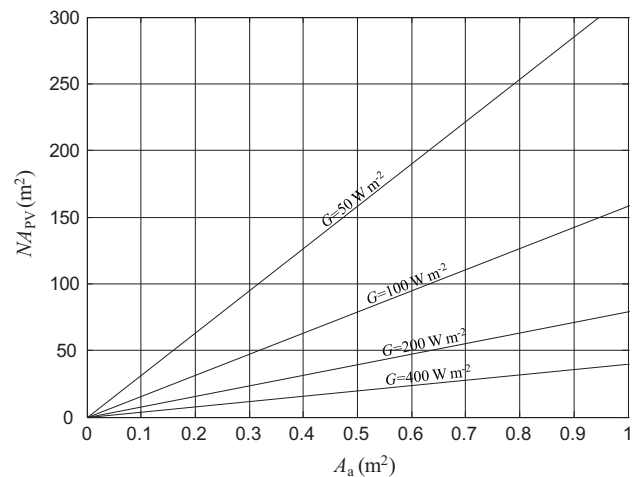

Fig. 4 – Relationship between NA_{PV} and A_a for different solar irradiances, G, for an effluent conductivity, $\lambda = 1.0\,mS\,cm^{-1}$, and $\alpha = 4.44 \times 10^{-3}\,A\,W^{-1}$.

represented by the daily mean irradiation, which depends on geographical location, the period of the year and the PV modules orientation (Lorenzo and Narvarte, 2000). For this reason, solar irradiation databases usually have a daily mean irradiation value for each month. For sizing purposes, the daily mean irradiation corresponding to the month with the lowest value is selected (Lorenzo, 2003) so the PSEO process could operate even in the most unfavorable conditions. This is the irradiation value that must be used to replace G in Eq. (7) in order to assess the PV module area required in a preliminary PSEO design process. However, in order to treat all the inlet effluents generated during the whole day, a storage tank is required to accumulate wastewaters during the periods of time in which the PSEO plant cannot operate (e.g. at night) or to assume fluctuations in water discharge. Sizing of the storage tank has not been included in the model because it is not considered a main item of the process (Cañizares et al., 2009).

The relationship between NA_{PV} and A_a may also vary with the effluent conductivity when G remains constant (Fig. 5). As previously stated, when the treated effluent presents low conductivity, a lower current density is needed to reach V_{min},
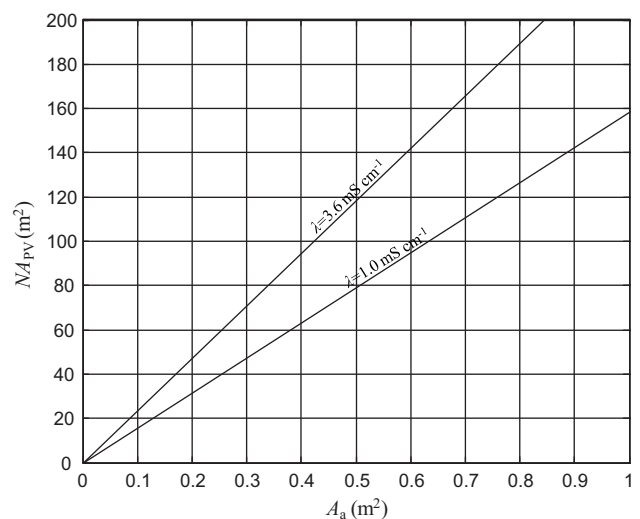

Fig. 5 – Influence of the effluent conductivity, λ, in the NA_{PV}–A_a relationship for $G = 100\,W\,m^{-2}$ and $\alpha = 4.44 \times 10^{-3}\,A\,W^{-1}$.

so the required PV module area to supply the electric current for a given A_a decreases.

4. Results and discussion

The urban wastewater treatment in isolated communities is one of the applications of the PSEO process. In this way, the sizing of several installations located at different geographically representative places in Spain has been carried out in order to determine the influence of the location.

For this purpose, and as a reference, an urban wastewater characterized by a $TOC_{in} = 80\,mg\,C\,L^{-1}$ and $\lambda = 1.0\,mS\,cm^{-1}$ is considered for the treatment. As the secondary treatment effluent of a conventional water reclamation facility has around $14\,mg\,C\,L^{-1}$ (Metcalf & Eddy, Inc., 2003), therefore an outlet effluent with the same TOC is established as a reference. As a result, 0.825 is the TOC conversion sought with this treatment. A wastewater generation of $154\,L\,inhab^{-1}\,d^{-1}$ is considered, which is the mean water consumption of the Spanish population in 2008 (INE, 2010). Then, F_{in} equals $154\,L\,d^{-1}$ so that the equipment sizes represent the values demanded by each inhabitant.

The PSEO process installations developed by Dominguez-Ramos (2010) are considered. The electrochemical cells are Diacell® filter-press reactors with BDD anodes manufactured by Adamant, whereas the monocrystalline PV modules were manufactured by SunTech (STP 160 model) for an effective area (A_{PV}) of $1.125\,m^2$ per module. The model parameter values have been determined for the case in which the process is carried out with this equipment and wastewaters described by Alvarez-Guerra et al. (2011): $\alpha = 4.44 \times 10^{-3}\,A\,W^{-1}$, $k = 1.80 \times 10^{-3}\,m\,min^{-1}$ (for urban wastewaters), $a = 2.28$, $b = 2.41$, $c = 0.0969$ for effluents with low conductivity, $\lambda = 1.0\,mS\,cm^{-1}$. It should be noted that these values of a, b and c are obtained whenever voltage is in V and current density in $mA\,cm^{-2}$ (Eq. (2)).

The mass balance is applied for the mentioned case in order to determine A_a. However, the consideration of a flow model is necessary for this purpose. The hydrodynamic behavior of filter-press reactors is generally modeled by means of a plug-flow reactor (PFR) and a continuously stirred tank reactor (CSTR) in series, the latter accounting for the inlet and outlet effects (Szpyrkowicz and Radaelli, 2006). However, in this case A_a is estimated considering both PFR and CSTR, because they lead to different A_a values that represent the extreme cases between which the anodic areas determined with other flow models are delimited. When a conversion equals 0.825, the $A_{CSTR} \cdot A_{PFR}^{-1}$ ratio is 2.70. This ratio increases with the conversion, so the influence of flow model is more important at high conversions. As a result, $0.104\,m^2$ is the anodic area for treating the effluent generated daily by each inhabitant assuming a PFR behavior.

The solar irradiation, G, associated with each location corresponds with the daily mean solar irradiation that falls in the most unfavorable month as was stated in Section 3. In Spain, the month with the lowest G is December. Moreover, this mean also varies with the orientation of PV modules. A south-facing and an optimally inclined plane that PV modules describe at each location, $G_{opt,Dec}$, is considered. Irradiation data have been obtained from the Photovoltaic Geographical Information System, PVGIS (JRC-EC, 2010). With these values of solar irradiation, $G_{opt,Dec}$, and the anodic areas for PFR flow model, NA_{PV} can be obtained according to Eq. (7).

Table 1 shows the PV module areas demanded by the A_a value calculated previously to treat the wastewaters produced by one inhabitant in different locations in Spain. The following cities are representative of the different irradiance areas where the PSEO process can be applied to isolated communities: North (Santander), Center (Madrid) and South (Malaga) of the Iberian Peninsula. In addition, Las Palmas de Gran Canaria has also been considered because it is the largest city in the ultraperipheral Spanish region (Canary Islands).

Table 1 states that the PV power supply sizing in the PSEO process is highly dependent on the geographical location, especially latitude. The obtained NA_{PV} values vary in the range $7.2–19.6\,m^2$ for PFR flow model. If the continental cities are compared, there is a significant difference in NA_{PV} between Santander (the location with the lowest $G_{opt,Dec}$) and Malaga (the highest $G_{opt,Dec}$), because in the former, a module area 66% higher is necessary to generate the same electric current. Considering the Canary Islands, this difference (between Santander and Las Palmas de Gran Canaria – the location with the highest $G_{opt,Dec}$) reaches a remarkable 172%.

The influence of the treatment efficiency, measured as TOC conversion, on the equipment size has been also analyzed. For this purpose, several characteristic TOC removals have been considered: 50%, 70% and 90%. The disinfection kinetics are notably faster than the TOC mineralization process, so a TOC conversion of 50% is enough to guarantee the disinfection of the wastewaters, which makes their reuse in applications that do not require high quality water possible (Pérez et al., 2010). 70% and 90% are the representative efficiency values of the trickling filters and activated sludge treatments, respectively (Spellman, 2009). Table 2 shows the PV module areas demanded at each location when wastewaters are treated with these TOC conversions, assuming a PFR behavior. The higher the treatment efficiency, the higher the $A_{CSTR} \cdot A_{PFR}^{-1}$ ratio: 1.44 ($X_{TOC} = 0.50$), 1.94 ($X_{TOC} = 0.70$) and 3.91 ($X_{TOC} = 0.90$). However $(NA_{PV}) \cdot A_a^{-1}$ is not affected by the TOC removal, as can be deduced from Eq. (7). The size of equipment increases with the treatment efficiency, this tendency being more evident when higher TOC conversions are considered. In this way, whereas $2.9–7.8\,m^2$ of PV module areas are only necessary for disinfection purposes, more than three times these values are

Table 1 – Area of the PV modules, NA_{PV}, required at different locations in Spain to treat the wastewaters produced daily by one inhabitant when PV modules are installed in the optimum inclined plane. An anodic area, A_a, of $0.104\,m^2$ is considered assuming a PFR model flow, so that the effluent generated daily by one inhabitant can be treated.

Location	Latitude	Optimum inclination angle (°)	$G_{opt,Dec}$ (W m^{-2})	$(NA_{PV}) \cdot A_a^{-1}$ (m^2 m^{-2})	$NA_{PV,PFR}$ (m^2)
Las Palmas de Gran Canaria	28°06′	28	228	69.4	7.18
Madrid	40°25′	34	111	142	14.7
Malaga	36°46′	32	139	114	11.8
Santander	43°28′	35	84	189	19.6

Location	$(NA_{PV})\cdot A_a^{-1}$ ($m^2\,m^{-2}$)	$NA_{PV,PFR}$ (m^2)		
		$X_{TOC} = 0.5$	$X_{TOC} = 0.7$	$X_{TOC} = 0.9$
Las Palmas de Gran Canaria	69.4	2.86	4.96	9.49
Madrid	142	5.85	10.2	19.4
Malaga	114	4.68	8.13	15.5
Santander	189	7.78	13.5	25.8

Table 2 – Influence of the TOC conversion achieved by the PSEO process over the required area of the PV modules when a PFR model flow is considered, $NA_{PV,PFR}$, and the $(NA_{PV})\cdot A_a^{-1}$ relationship.

required to obtain the same efficiency of a sludge activated treatment: 9.5–26 m^2.

5. Conclusions

The PSEO process design allows the sizing of the electrochemical reactor anodic area and the effective PV module area. The anodic area depends on the wastewater flow rate (which is a function of the number of inhabitants for urban wastewaters), the characteristics of the effluent (inlet TOC) and the treatment efficiency.

Once the electrochemical reactor is sized, the effective photovoltaic module area is a function of the effluent conductivity and the solar irradiation (measured as the daily mean irradiation for the month with the lowest value). The conductivity determines the current necessary to achieve a minimum voltage (established in 5 V) that guarantees that the desired electrochemical oxidation reactions take place. However, solar irradiation is the most important variable affecting the required PV area, since lower irradiances imply higher areas to generate the same electric current. The solar irradiation is a function of the location, which leads to effective PV module area differences as large as 170% among selected Spanish irradiation areas. For example, an installation for one inhabitant with the solar irradiation of Santander will require an anodic area of 0.104 m^2 and a PV module area of 19.6 m^2. The area for the PV system decreases to 7.19 m^2 if the process is installed in Las Palmas de Gran Canaria. Consequently, the PSEO process feasibility is a function of its geographical location, since wastewaters that are treated by means of this technology in a sunny place demand a higher photovoltaic module area in other locations with lower irradiations.

Acknowledgements

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