

PERFORMANCE ANALYSES OF MICROBIAL FUEL CELLS OPERATED IN SERIES

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Abstract: Microbial Fuel Cells (MFCs) are capable of producing electricity while cleaning wastewater. This novel technology can replace energy-consuming aerobic treatment even if low effluent concentrations are demanded. The goal of this work is to optimize a MFC-based wastewater treatment process. A MFC mathematical model, which accounts for co-existence of methanogenic and anodophilic microbial populations, is used to compare different operating modes and reactor configurations. The following observations are made based on the model analysis: (i) the ratio between the anodophilic and methanogenic populations can be controlled by the electrical load; (ii) co-existence of the two populations decreases reactor performance, and (iii) reactors connected in series always improves treatment efficiency.

Keywords: Microbial fuel cell; Cogeneration; Model-based design.

1. INTRODUCTION

The infrastructure and energy required to treat wastewater represents a major challenge for modern society. There are many technologies available to manage organically polluted waste streams and those that use organic waste as a source of renewable energy are among the most promising. However, only a few technologies can perform the task of using very diluted waste as substrate. Among these technologies, Microbial Fuel Cells (MFCs) are an interesting possibility.

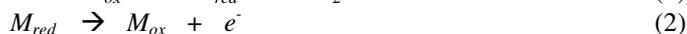
MFCs are bioreactors that have the potential to convert a large variety of highly diluted organic matter of various compositions into electricity (Logan and Regan 2006). They contain anodophilic bacteria, which oxidize organic matter and transfer electrons to an electrode during metabolism (Debabov 2008). The main restriction for MFC's application is its low power output (Logan et al. 2006). In addition, using wastewater as substrate implies the presence of mixed microbial populations, such as fermentative, methanogenic and anodophilic microorganisms (Arcand et al. 1994; Moletta et al. 1986; Quarmby and Forster 1995), which affect the performance of the MFC. One solution for understanding the complex problems posed by MFCs is to build a dynamic mathematical model that can describe the behavior of diverse microbial populations competing for the same substrate (Picioreanu et al. 2007).

This paper presents a steady state analysis of a dynamic model built to describe co-existence of two microbial populations in MFC (Pinto et al. 2010). The model takes into account the competition for substrate between anodophilic and methanogenic microorganisms. As has been shown before, anodophils consume more organic matter at low substrate concentrations, while methanogens perform well at high substrate concentrations (Torres et al. 2007). To maximize the

treatment capacity (flow rate for a given effluent concentration) it is shown here that it is better to connect MFCs in series, with the first stages converting the substrate at high rates, and the final stages polishing the effluent to a specific requirement demand, a technique often called staging (Van Lier et al. 2001).

2. A DYNAMIC MODEL FOR MICROBIAL FUEL CELLS

This section presents a model for the MFC, derived from Pinto et al. (2010), that will be used for the analysis and optimization purposes in this paper. The model represents competition between anodophilic and methanogenic microorganisms for the substrate, which in this case is acetate. Charge transfer at the anode is modeled using an intracellular mediator. The reactions at the anode are as follows:



where S is the substrate (acetate) concentration. M_{red} and M_{ox} are the reduced and oxidized forms of the intracellular mediator, respectively. The dynamic mass balance equations of the model are given below.

$$\frac{dS}{dt} = D(S_0 - S) - \frac{q_{max,a}S}{K_{S,a} + S} \frac{M_{ox}}{K_M + M_{ox}} x_a - \frac{q_{max,m}S}{K_{S,m} + S} x_m \quad (4)$$

$$\frac{dx_a}{dt} = \left(\frac{\mu_{max,a}S}{K_{S,a} + S} \frac{M_{ox}}{K_M + M_{ox}} - K_{d,a} - \alpha D \right) x_a \quad (5)$$

$$\frac{dx_m}{dt} = \left(\frac{\mu_{max,m}S}{K_{S,m} + S} - K_{d,m} - \alpha D \right) x_m \quad (6)$$

$$\frac{dM_{ox}}{dt} = \frac{\gamma I_{MFC}}{mFVx_a} - Y \frac{q_{max,a}S}{K_{S,a} + S} \frac{M_{ox}}{K_M + M_{ox}} \quad (7)$$

where S and S_0 are the substrate concentration and the influent substrate concentration, respectively (mg-S L^{-1}); x_a and x_m are the concentration of anodophilic and methanogenic microorganisms, respectively (mg-x L^{-1}); t is time (d); D is the dilution rate ($D=F_{in} V^{-1}$), F_{in} is the incoming flow (L d^{-1}), V is the anodic compartment volume (L); M_{ox} is the oxidized mediator fraction per anodophilic microorganism [mg-M mg-x^{-1}]; I_{MFC} is the MFC current [A]; α , is a dimensionless biofilm retention parameter. Biofilm formation and retention at $\mu_{max,a} < D$ and $\mu_{max,m} < D$ was simulated using a two-phase biofilm growth model as in Mu et al. (2008), where a biomass retention factor is introduced as:

$$\alpha = \left(\frac{1 + \tanh[K_x(x_a + x_m - X_{max})]}{2} \right) \quad (8)$$

The MFC current is calculated by an electrochemical balance neglecting activation losses. Open circuit potential and internal resistance values were chosen to be a function of the concentration of anodophilic microorganisms as (Pinto et al. 2010):

$$I_{MFC} = \left(E_{OCV} - \frac{RT}{F} \ln \left(\frac{M_{total}}{M_{total} - M_{ox}} \right) \right) (R_{ext} + R_{int})^{-1} \quad (9)$$

$$R_{int} = R_{min} + (R_{max} - R_{min}) e^{-K_R x_a} \quad (10)$$

$$E_{OCV} = E_{min} + (E_{max} - E_{min}) e^{\frac{-1}{K_R x_a}} \quad (11)$$

where E_{OCV} is the MFC open circuit voltage [V]; R_{ext} is the external resistance [Ω]; R_{int} is the internal resistance [Ω].

The parameters of the above model were either estimated using experimental results obtained in a 50 mL (anodic chamber volume) MFC or chosen from values reported in the literature. Parameters description, units and values are provided in Table 1. The model responses were validated with two independent data sets. A detailed description of the model and the experimental results can be found in Pinto et al. (2010).

Table 1. Model parameters

Param	Value	Description and dimension
F	96485	Faraday constant - A d mole ⁻¹
R	8.314	ideal gas constant - J K ⁻¹ mol ⁻¹
T	298.15	MFC temperature - K
Y^*	22.75	yield in (7) - mg-M mg-S ⁻¹
Y_{CH_4}	0.3	methane yield - mL-CH ₄ mg-S ⁻¹
$q_{max,a}^*$	8.48	max. anodophilic reaction rate mg-S mg-x ⁻¹ d ⁻¹
$q_{max,m}^*$	8.20	max. methanogenic reaction rate mg-S mg-x ⁻¹ d ⁻¹
$\mu_{max,a}^*$	1.97	max. anodophilic growth rate - d ⁻¹
$\mu_{max,m}^*$	0.1	max. methanogenic growth rate - d ⁻¹
$K_{d,a}$	0.04	decay rate of anodophils - d ⁻¹

$K_{d,m}$	0.002	decay rate of methanogens - d ⁻¹
X_{max}	525	maximal attainable biomass - mg-x L ⁻¹
$K_{S,a}$	20	half-rate constant of anodophils mg-S L ⁻¹
$K_{S,m}$	80	half-rate constant of methanogens mg-S L ⁻¹
M	2	e ⁻ transferred per mol of mediator mole ⁻¹ mol _{med} ⁻¹
γ	663400	mediator molar mass - mg-M mol _{med} ⁻¹
M_{total}	0.05	mediator fraction - mg-M mg-x ⁻¹
K_M	0.01	mediator half-rate constant - mg-M L ⁻¹
K_x	0.4	parameter in (8) - L mg-x ⁻¹
R_{min}^*	25	minimum internal resistance - Ω
R_{max}^*	2025	maximum internal resistance - Ω
E_{min}	0.01	minimum E_{OCV} - V
E_{max}^*	0.674	maximum E_{OCV} - V
K_R	0.024	parameter in (10) and (11) - L mg-x ⁻¹

*Estimated parameters in Pinto et al. (2010)

3. MODEL ANALYSIS: COEXISTENCE AND SUBSTRATE CONSUMPTION

3.1 Competitive Exclusion and Coexistence

One of the first questions to be answered is related to the coexistence of the two populations. The competitive exclusion principle (Hardin 1960) suggests the extinction of one of the species when there is a competition for the same substrate in the same ecological niche. This principle was mathematically characterized by Harmand et al. (2008), where it was shown that similar kinetics for growth rate are required to cause this type of exclusion. In the model considered, only the growth rate of anodophilic microorganisms is limited by the mediator concentration. The mediator concentration in turn is influenced by the external resistance; hence the external resistance plays a key role in the type of microorganisms that are present in the MFC.

The influence of the external resistance on the populations at steady state is presented in Figure 1. The R_{ext} varied between 10 to 5000 Ω , while the influent concentration was 1000 mg.L^{-1} . Three regions can be distinguished in this figure, i.e. (I) only anodophilic microorganisms, (II) coexistence, and (III) only methanogenic microorganisms.

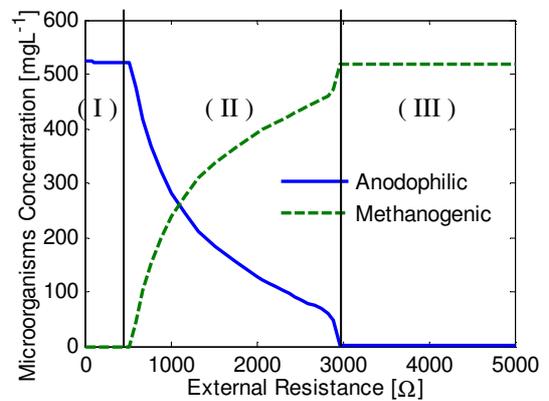


Figure 1. Predicted concentration of anodophilic and methanogenic populations as a function of R_{ext} .

Proposition 1: Let the decay rates be negligible and the half-rate constants of the Monod kinetics of the methanogens be greater than that of the anodophils, i.e. $K_{s,m} > K_{s,a}$. Then, the existence of different microorganisms is determined by the expression $\mu^* = \frac{\mu_{max,m} K_M + M_{ox}}{\mu_{max,a} M_{ox}}$. Three regions can be

distinguished based on the value of μ^* : (I) $\mu^* < 1$, only anadophils exist. (II) $1 < \mu^* < K_{s,m}/K_{s,a}$, both microorganisms coexist and (III) $\mu^* > K_{s,m}/K_{s,a}$, only methanogens exist.

Proof: The steady state solution of (5) and (6) present four possible solutions:

- (I) $A=0$ and $x_m = 0$ (only anodophilic microorganisms)
- (II) $A=0$ and $B=0$ (coexistence)
- (III) $B=0$ and $x_a = 0$ (only methanogenic microorganisms)
- (IV) $x_m = 0$ and $x_a = 0$ (wash-out solution)

$$\text{where } A = \frac{\mu_{max,a} S}{K_{s,a} + S} \frac{M_{ox}}{K_M + M_{ox}} - \alpha D, \text{ and } B = \frac{\mu_{max,m} S}{K_{s,m} + S} - \alpha D$$

For coexistence to occur, the following condition has to be satisfied: $A = B = 0$. Under this condition, A and B can be rearranged as:

$$\frac{\mu_{max,a} S}{K_{s,a} + S} \frac{M_{ox}}{K_M + M_{ox}} = \frac{\mu_{max,m} S}{K_{s,m} + S} = \alpha D \quad (12)$$

Equation (12) can be solved to give:

$$S = \frac{K_{s,m} - K_{s,a} \mu^*}{\mu^* - 1}, \text{ where } \mu^* = \frac{\mu_{max,m} K_M + M_{ox}}{\mu_{max,a} M_{ox}} \quad (13)$$

If the above expression gives a positive value of S , coexistence is possible. On the contrary, negative values of S indicate that one of the microorganisms would be extinct. Therefore, that coexistence is possible only between $1 < \mu^* < K_{s,m}/K_{s,a}$. Furthermore, by linearization of (5) and (6) and analysing sign of its eigenvalues, one can see that when $\mu^* < 1$, methanogens grow slower than the anodophils for any value of S , leading to methanogens extinction, i.e. only solution (I) is stable. The reverse case occurs for $K_{s,m}/K_{s,a} < \mu^*$ when by the eigenvalues analysis only solution (III) is stable. This stability analysis is omitted here for the sake of simplicity. ■

3.2 Variation of Substrate Consumption with External Load and Outlet Concentration

The coexistence depends on the difference between the growth rates of the methanogens and the anodophils, while the treatment capacity depends on the substrate consumption of these microorganisms. From (4), it can be seen that the consumption rate depends upon the desired effluent substrate concentration and the quantity of each of these species. As seen in the previous section, the later is determined by the external resistance (electric load). So, the effect of external

load and outlet concentration on the consumption rate is studied here.

The influence of the consumption rate on the effluent substrate concentration (S) is presented in Fig. 2. Three values of R_{ext} are chosen that correspond to (I) only anodophils, (II) coexistence, and (III) only methanogens. The influent concentration was varied from 2500 to 150 mg/L. It can be seen that the methanogens perform better for higher substrate concentrations while the anodophils do better at lower concentrations. The coexistence always results in a decreased substrate consumption rate.

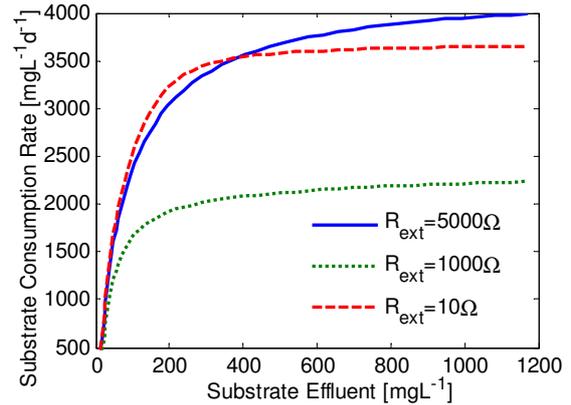


Figure 2. Steady state substrate consumption rate for MFC colonized by either anodophils ($R_{ext} = 10\Omega$), both populations ($R_{ext} = 1000\Omega$), or methanogens ($R_{ext} = 5000\Omega$).

The intersection point of the two curves in Fig. 2 can be expressed as follows,

$$S^* = \frac{K_{s,m} - K_{s,a} q^*}{q^* - 1}, \text{ where } q^* = \frac{q_{max,m} K_M + M_{ox} x_m}{q_{max,a} M_{ox} x_a} \quad (14)$$

If $x_m = x_a$ and $M_{ox} = M_{total}$, for a given set of model parameters S^* can be computed as 355 mg.L⁻¹. So, for $S < S^*$ the anodophils have higher substrate consumption rate than methanogens and vice versa. As R_{ext} increases, M_{ox} decreases and the value of S^* increases. Furthermore, the lower R_{ext} larger is the substrate consumption rate for anodophils.

The influence of external resistance on the substrate consumption rate is shown in Fig. 3. Two fixed substrate effluent concentrations, one smaller than S^* and the other higher were selected for this analysis. At low effluent substrate concentrations, an MFC with small external resistance (anodophils only) consumes the most organic matter, while for high concentrations the best treatment performance is reached at high resistance (methanogens only).

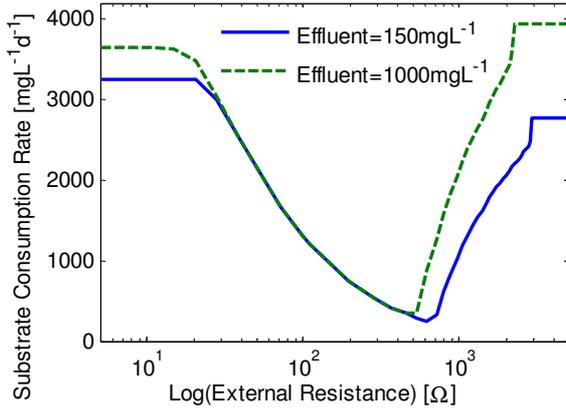


Figure 3. Steady state substrate consumption rate for MFC operated at low (150 mg.L⁻¹) and high (1200 mg.L⁻¹) effluent concentration.

Proposition 2: If $\frac{q_{max,a}\mu_{max,m}}{q_{max,m}\mu_{max,a}} \leq 1$, then coexistence always leads to lower substrate consumption.

Proof: This result can be proved by comparing the substrate consumption rates (r) for each region. For a given S , r_m = constant and r_a is maximized when $M_{ox} = M_{total}$, i.e., at low R_{ext}

$$r_{max,a} = \frac{q_{max,a}S}{K_{S,a} + S} \frac{M_{tot}}{K_M + M_{tot}} X_{max} \quad (15)$$

$$r_{max,m} = \frac{q_{max,m}S}{K_{S,m} + S} X_{max} \quad (16)$$

For the coexistence region the substrate consumption rate is (r_c):

$$r_{max,c} = \frac{q_{max,a}S}{K_{S,a} + S} \frac{M_{ox}x_a}{K_M + M_{ox}} + \frac{q_{max,m}S(X_{max} - x_a)}{K_{S,m} + S} \quad (17)$$

The coexistence only occurs when $A = B = 0$, then (12) is valid. Substituting the same gives,

$$r_{max,c} = \left[\frac{q_{max,a}\mu_{max,m}}{q_{max,m}\mu_{max,a}} \frac{x_a}{X_{max}} + 1 - \frac{x_a}{X_{max}} \right] \frac{q_{max,m}SX_{max}}{K_{S,m} + S} \quad (18)$$

$$\frac{r_{max,c}}{r_{max,m}} = [1 + (\beta - 1)\alpha] \leq 1 \quad (19)$$

Because $\alpha = x_a/X_{max}$ varies between 0 and 1, the value of the above function varies between 1 and $\beta = \frac{q_{max,a}\mu_{max,m}}{q_{max,m}\mu_{max,a}}$. If $\beta < 1$,

then $r_{max,c} \leq r_{max,m}$. ■

4. OPTIMIZATION OF SUBSTRATE CONSUMPTION BY STAGING

4.1 Staging

It is well known that when the substrate consumption is described by Monod kinetics, the reaction proceeds far more rapidly in a plug flow reactor than in a continuous stirred tank reactor (Eddy 2002; Shuler and Kargi 1992). This means that more substrate can be consumed with the plug flow reactor rather than using a CSTR reactor. In the case of MFCs, where the continuous mode is used, it is recommended to use reactors-in-series to approach the results from plug-flow operation (Shuler and Kargi 1992), a technique often called staging. First, it is shown mathematically that if two MFCs are present, staging would always lead to better performance than running the reactors in parallel.

Proposition 3: Given a fixed influent concentration and a specified effluent concentration, operating two MFCs in series leads to a higher performance than operating them in parallel.

Proof: Consider the two MFCs to have only methanogens and have the same volume. This analysis would be similar with anodophils. Let S_f be the specified effluent concentration and suppose that $x_m = X_{max}$. The flow rate of one reactor (F_1) at steady state is given by:

$$F_1 = \frac{V}{(S_0 - S_f)} \frac{q_{max,m}S_f}{K_{S,m} + S_f} X_{max} \quad (20)$$

For the MFCs in series, let S_{mid} be the effluent concentration of the first and the influent concentration of the second. Also assume that the two MFCs have same biomass concentration (X_{max}). Then, the flow rate (F_2) is given by:

$$F_2 = \frac{VX_{max}}{(S_{mid} - S_f)} \frac{q_{max,m}S_f}{K_{S,m} + S_f} = \frac{VX_{max}}{(S_0 - S_{mid})} \frac{q_{max,m}S_{mid}}{K_{S,m} + S_{mid}} \quad (21)$$

The above equation can be solved to give:

$$S_{mid} = \frac{S_f(S_f + S_0)}{2(K_{s,m} + 2S_f)} + \sqrt{\frac{S_f^2(S_f + S_0)^2}{4(K_{s,m} + 2S_f)^2} + \frac{S_0K_{s,m}S_f}{(K_{s,m} + 2S_f)}} \quad (22)$$

For the series configuration to be better than the parallel one, $F_2 > 2F_1$:

$$\frac{1}{(S_{mid} - S_f)} > \frac{2}{(S_0 - S_f)} \text{ i.e., } S_{mid} < \frac{(S_0 + S_f)}{2} \quad (23)$$

So, the proposition can be proved if it can be shown that

$$\frac{S_f(S_f + S_0)}{2(K_{s,m} + 2S_f)} + \sqrt{\frac{S_f^2(S_f + S_0)^2}{4(K_{s,m} + 2S_f)^2} + \frac{S_0K_{s,m}S_f}{(K_{s,m} + 2S_f)}} < \frac{(S_0 + S_f)}{2} \quad (24)$$

Moving S_f ($(S_f + S_0)/2(K_{s,m} + 2S_f)$) to the right hand side, removing the denominators and re-arranging gives:

$$S_f^2(S_f + S_0)^2 + 4S_0K_{s,m}S_f(K_{s,m} + 2S_f) < (S_0 + S_f)^2(K_{s,m} + S_f)^2 \quad (25)$$

(25) can be further simplified to:

$$4S_0S_f < (S_0 + S_f)^2 \quad (26)$$

Which is obviously true since $(S_f - S_0)^2 > 0$. Therefore, $S_{mid} < (S_f + S_0)/2$ and so, $F_2 > 2 F_1$. ■

4.2 Optimizing a Two-Stage Process

The optimization problem addressed is the following: Given (i) two MFCs of prefixed volume (ii) a fixed influent concentration, (iii) a constraint on the effluent concentration, choose (i) the interconnection structure between the MFCs, (ii) the external resistance of each MFCs and (iii) the substrate concentration between the two MFCs if they are in series, in order to maximize the flow rate, i.e. maximize MFC treatment capacity.

From a substrate consumption point of view, the choice of external resistance can be considered binary. This is in fact justified by Fig 3, where there are two plateaus for methanogens and anodophils. For operation with methanogens, which necessitates high external resistance, $R_{ext} = 5000\Omega$ was selected, while simulations with anodophils, the low external resistance was chosen as 10Ω . Thus, the interconnection structure and external resistance are binary variables, which in turn lead to the following six interconnection configurations:

- #1. MP - Two MFCs with high external resistance in parallel (Methanogens)
- #2. AP - Two MFCs with low external resistance in parallel (Anodophils)
- #3. MM - Two MFCs in series, both with high external resistance (Methanogens)
- #4. MA - Two MFCs in series, the first with high external resistance followed by the second with low external resistance (Methanogens and Anodophils)
- #5. AM - Two MFCs in series, the first with high external resistance followed by the second with low external resistance (Anodophils and Methanogens)
- #6. AA - Two MFCs in series, both with low external resistance (Anodophils)

Moreover, if the reactors are connected in series, the substrate concentration in between has to be determined so as to have the same flow in both reactors. Thus, the optimization problem is purely a combinatorial one. So, the flow rates for all the six configurations would be evaluated and best is selected. All calculations were done at steady state using the model presented in Section 2.

Figure 4 shows the steady state response for the treated flow as a function of the effluent substrate concentration (1st MFC)

or influent concentration (2nd MFC). From this figure all six configurations' treatment capacity can be computed for a fixed influent and effluent.

The operation of reactors in parallel can be analyzed using lines corresponding to 1st MFCs lines in Fig. 4. As expected, the treatment capacity was larger for an MFC occupied by anodophils when substrate effluent concentration was low. When the treatment requirements were less strict, an MFC with methanogens present a larger treatment capacity. For the two MFCs in parallel, with a specific substrate effluent concentration, the maximum treatment capacity could be found simply by multiplying the treated flow by the number of MFCs in parallel.

The treatment capacity of MFCs operating in series was also computed from Figure 4. For this, additional curves (2nd MFC) that link the substrate influent concentration with the flow for a given fixed effluent concentration were required. The crossing points in Figure 4 represent the treatment capacity of each configuration in series (MM, MA, AM, or AA).

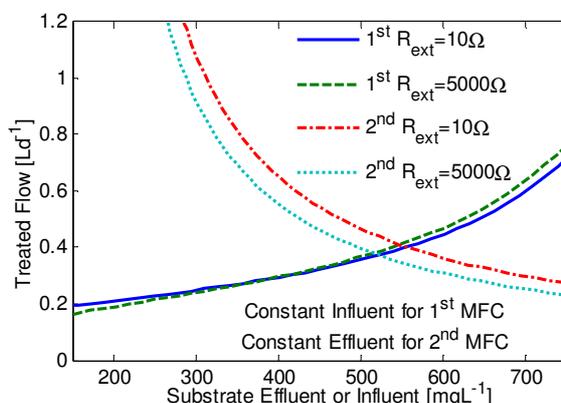


Figure 4. Crossing treatment capacities for two staged reactors with 1000 mg.L^{-1} of influent concentration and effluent concentration of 150 mg.L^{-1} .

For this influent and effluent, the best series solution was the first reactor with methanogens and the second with anodophils. These results are summarized in the following table:

Table 2. Comparison of diverse configurations for two identical MFCs treating a stream with organic load of 1000 mg.L^{-1} being reduced to 150 mg.L^{-1} .

Case	Treatment capacity [L/d]	Increase* [%]
MP	0.327	-
AP	0.384	17.4
MM	0.38	16.2
MA	0.41	25.4
AM	0.373	14.1
AA	0.40	22.3

*In relation to base case MP

The treatment capacity for all configurations was computed for several influent and effluent concentrations. The results are summarized in Figure 5, where the design that presents the largest treatment capacity is indicated in each region:

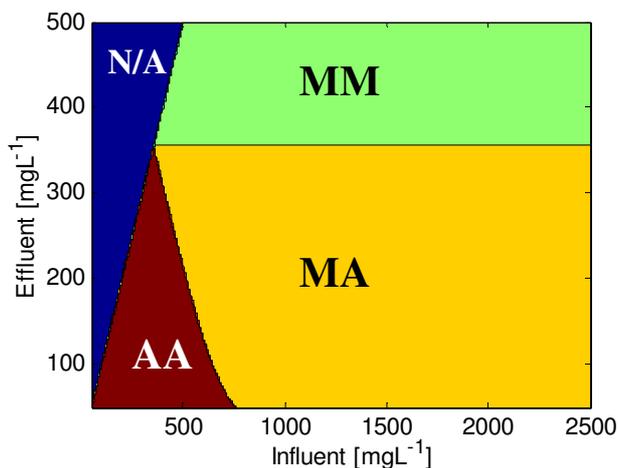


Figure 5. Regions with the largest treatment capacity. Area denoted by N/A represents a section where effluent is larger or the same as influent (unfeasible region).

The configuration in series with MFCs operating with methanogens first and anodophils second represent the best treatment capacity for most of the operating regions. This configuration has the methanogens consuming substrate at large substrate concentrations and anodophils polishing the effluent concentration to a specific requirement. When the effluent requirements are less strict, the configuration with two MFCs with methanogens in series is the best. For low concentrations of influent, two MFCs with anodophils in series present the best results.

5. CONCLUSIONS

This paper presents analysis of a two-population MFC model. The model predicts the concentration of anodophilic and methanogenic microorganisms, and shows that MFC external resistance (electric load) and organic load affect steady state distribution of microbial populations. Steady state analysis of the model showed that three possible scenarios could be obtained: (I) only anodophilic microorganisms, (II) coexistence, and (III) only methanogenic microorganisms. Furthermore, it was shown mathematically that methanogens have higher substrate consumption rate at higher substrate effluent concentrations than the anodophils, while the reverse occurs at lower substrate concentrations. The coexistence scenario always had lower substrate consumption rate.

In addition, staged MFCs were proven to always present better treatment capacity than parallel MFCs. Diverse designs for a co-generation/staging unit with two MFCs were simulated to compare the maximum organic load treating capacity. The best design will be a function of dominant microbial population in each MFC, selected according to its external load. Regions with the best design were drawn as a function of influent and effluent concentrations. For the largest and most common region of operation, two MFCs in series, the first with high external resistance followed by a second with low external resistance, presented the best result.

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