SENSITIVITY ANALYSIS OF A MODEL WHICH DESCRIBES THE BIOFILTRATION OF VRSC COMPOUNDS

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***Abstract*— A sensitivity analysis of a model which describes the biooxidation of volatile reduced sulfur compounds (VRSC) using a biotrickling filter is developed. Consistent results are obtained using three methodologies; Standard Regression Coefficients (SRC), Fourier amplitude sensitivity test (FAST) and the Morris method (MOAT). The model presents a highly linear behavior of the uncertain parameters. It is shown how the properties of the biofilm and the parameters related with the kinetics have the highest influence on the model behavior; while on the other hand the properties related with mass transfer have low influence.**

***Keywords*— Biofiltration; sensitivity analysis; VRSC; modeling biofilter; biotrickling filter**

I. INTRODUCTION

The biotrickling filters (BTF) are an interesting alternative for removing contaminants from gaseous effluents, especially when operating at very low concentrations and large amounts of gas must be treated. In these systems, the contaminant diffuses from the gas phase to a biofilm, where microorganisms biooxidize the pollutant, usually using it as an energy source. One of the most common applications of this technology is the removal of odors caused by gaseous effluents which contain low levels of H_2S and other volatile reduced sulfur compounds (VRSC) concentration. These compounds are frequently found in processes where heating or anaerobic decomposition of organic matter is present (Wani *et al.*, 1999).

For the modeling and simulation of these systems, it is very important to determine unknown parameters such as microbial kinetic constants or physicochemical properties values. While in some cases these values can be estimated, in others they must be determined from experimental data using nonlinear fitting techniques (Alonso *et al.*, 2000). Simulations show that the effect of these parameters on them is different, and many authors estimate these values by fitting experimental results (Devinny and Ramesh, 2005). The sensitivity of these parameters is not entirely clear. Bath *et al*. (2006) indicate that the thickness of the biofilm has an important influence on the biooxidation; whereas Chmiel *et al.* (2005) show that the kinetics parameters have high influence in the model behavior.

To solve these uncertainties, sensitivity analysis is an useful tool which allows to determine the behavior of a model due to changes on the input parameters (Viotti *et al.*, 2002). Basically, this methodology allows determining the significance of one input over an uncertain output (Saltelli, 2000).

Several techniques are available for sensitivity analysis. For example: the one-factor-at-a-time (OAT) (Saltelli, 1999), Fourier amplitude sensitivity test (FAST) (McRae *et al.*, 1982), fractional factorial design method (Henderson-Sellers *et al.*, 1996), Morris method (Morris, 1991), sampling-based methods (Helton *et al*., 2006), Sobol's method (Sobol, 1993) and McKay's method based on a one-way ANOVA (McKay, 1997).

The aim of this work is developing a sensitivity analysis of a biotrickling filter model which biooxidizes VRSC, in order to analyze the effect that each parameter has on the simulation.

II. MATHEMATICAL MODEL

The biooxidation is described by a model which considers the mass transfer from the gas phase to the liquid phase and subsequent biochemical degradation by the microorganism attached in the support. When air contaminated with VRSC flows throughout the column, VRSC are transferred from the gas phase to the liquid phase where they diffuse to the biofilm; there they are oxidized by the microbial activity.

A. Assumptions

The following assumptions for the model development were considered:

- Steady-state operation; therefore the absorption of VRSC on the packing material is in equilibrium and should not be considered in the mass balance. Temperature and pH are constant.
- The biomass accumulation rate in the reactor is small compared to the VRSC's biodegradation rate; therefore, mass balance for biomass will not be performed.
- Oxygen is present in excess in relation to the VRSC; therefore the microorganism growth is not limited by this element.
- The biofilm coating is formed on the surface of the packing. Due to the very small thickness of the coating, mass transfer is assumed perpendicular to the gas flow.
- The concentration of VRSCs in the interface is calculated using Henry's law. It is assumed that the distribution coefficient is similar than water's.
- The effective diffusivity of the compounds in the biofilm is similar to the diffusivity of the compounds in water, thus the effective diffusivity can be calculated by applying a correction factor in the

water diffusivity.

- The biofilm's thickness is relatively small compared to the curvature of the media; therefore modelling can be performed by using planar geometry.
- The mixture of gases in the biotrickling filter can be described using a dispersion model.
- The kinetics of biooxidation is described by a Monod model type.

B. Equations

The model is based on the work of Spigno *et al*. (2004).

Equation 1 shows the dimensionless general mass balance in the gas phase:

$$\frac{1}{Pe}\cdot\frac{\partial^2 C_g}{\partial\zeta^2}-\frac{\partial C_g}{\partial\zeta}+v_b\cdot Ti\cdot\left.\frac{\partial C_b}{\partial\psi}\right|_{\psi=O}=0 \qquad (1)$$

where C_g is the dimensionless concentration in the gas phase, C_b is the dimensionless concentration in the biofilm; Pe is the Peclet Number, ζ is the dimensionless axial co-ordinate along the bed's height, ψ is the dimensionless spatial co-ordinate in the biofilm, v_b is the specific volume in the biofilm and *Ti* is the residence to diffusion time ratio.

Equation 2 shows the dimensionless general mass balance in the liquid phase:

$$\frac{\partial^2 C_b}{\partial\psi^2}-Th^2\cdot\phi\cdot\kappa=0 \qquad (2)$$

where *Th* is the Thiele number, ϕ is the dimensionless inlet biomass concentration and κ is the dimensionless specific degradation rate.

Equation 3 shows the dimensionless kinetic equation for biooxidation rate:

$$\kappa=\frac{1}{Y_{x/S}}\cdot\frac{C_b}{\sigma+C_b} \qquad (3)$$

where $Y_{x/S}$ is the yield coefficient of biomass (g biomass g^{-1} substrate) and σ is the dimensionless Monod constant.

The dimensionless concentrations in the gas, in the biofilm and in the biomass are defined as:

$$C_g=\frac{c_g}{c_g{}^{in}}\qquad C_b=\frac{c_b}{c_b{}^{in}}\qquad \phi=\frac{X_b{}^{in}}{c_g{}^{in}} \qquad (4)$$

where c_g is the concentration in the gas phase (g m^{-3}), $c_g{}^{in}$ is the inlet concentration in the gas phase (g m^{-3}), c_b is the concentration in the biofilm (g m^{-3}), $c_b{}^{in}$ is the inlet concentration in the biofilm (g m^{-3}) and $X_b{}^{in}$ is the inlet biomass concentration (g m^{-3}).

The dimensionless axial and radial axes systems along the biotrickling filter are defined as:

$$\psi=\frac{x}{\delta},\qquad \zeta=\frac{z}{H} \qquad (5)$$

where *x* is the spatial co-ordinate in the biofilm (m), δ is the biofilm thickness (m), *Z* is the axial co-ordinate (m) and *H* is the biotrickling filter's height (m).

The Péclet number (*Pe*), modified Thiele module (*Th*), the distribution of residence time (*Ti*) and specific volume of the biofilm (*vb*) are defined as:

$$Pe=\frac{V_z\cdot H}{W^{ef}}\qquad v_b=\delta\cdot\alpha\cdot a_s$$

$$Ti=\frac{H/V_z}{\delta^2/D_i}=\frac{\tau_R}{\tau_D}\qquad Th_i=\sqrt{\frac{\delta^2\cdot\mu_{\max}}{D}} \qquad (6)$$

$$W^{ef}=\varepsilon W$$

where a_s is the specific surface area per reactor unit volume (m^{-1}), *D* is the diffusion coefficient (m^2 s^{-1}), V_z is the gas velocity (m s^{-1}), *W* is the dispersion coefficient (m^2 s^{-1}), α is the fraction of support surface area covered with biofilm, τ_R is the gas residence time (s), τ_D is the characteristic time of diffusion in the biofilm (s) and μ_{max} is the maximum specific growth rate (s^{-1}).

The dimensionless Monod constant is expressed as:

$$\sigma_i=\frac{K_s}{c_b^{in}} \qquad (7)$$

where K_S is the Monod constant (g m^{-3}).

The boundary conditions are:

$$\zeta=0\qquad 0\le\psi\le1\quad -\frac{\partial C_g}{\partial\zeta}+v_b\cdot Ti\cdot\left.\frac{\partial C_b}{\partial\psi}\right|_{\psi=O}=0$$

$$\zeta=1\quad 0\le\psi\le1\quad -\frac{\partial C_g}{\partial\zeta}+v_b\cdot Ti\cdot\left.\frac{\partial C_b}{\partial\psi}\right|_{\psi=O}=0 \qquad (8)$$

$$\psi=0\qquad 1\le\zeta\le0:\quad c_b=\frac{c_g}{m}$$

$$\psi=1\qquad 1\le\zeta\le0:\quad \frac{\partial C_b}{\partial\psi}=0$$

where *m is the* air–water distribution coefficient.

Fan *et al.* (1990) indicate that the diffusion in a biofilm depends on the concentration of biomass; they developed an empirical model which corrects the diffusion coefficient of water as a function of the biomass concentration (Eq. 9),

$$D_b=D_w\left[1-\frac{0.47\left(X_b\right)^{0.92}}{11.19+0.27\left(X_b\right)^{0.99}}\right] \qquad (9)$$

where D_b is the diffusion coefficient in the biofilm, D_w is the diffusion coefficient in the water and X_b is the biomass concentration.

Biofilm thickness was determined as shown in Eq. (10).

$$\delta=\frac{X_r}{\alpha\cdot a_s\cdot X_b} \qquad (10)$$

where X_r is the biomass concentration in the recirculating medium.

III. SENSITIVITY ANALYSIS

The sensitivity analysis is based on the work done by Saltelli *et al.* (2005) where a Monte Carlo simulation using Latin hypercubic sampling method (LHS) with standardized variables is performed. LHS is a so-called stratified sampling without replacement technique, where the random parameter distributions are divided into *N* equal probability intervals, which are then sampled. *N* represents the sample size. *N*'s value should be

at least k+1, where k is the number of parameters varied, but it should usually be much larger to ensure accuracy (Marino *et al.*, 2008).

In order to solve the differential equation system, finite differences are developed and simultaneously Newton–Raphson method for multiple variables is used after the discretization of the differential equations. Applying this discretization to the liquid phase and using the boundary conditions presented above, a nonlinear equation system is obtained. This system has a number of variables which depend on the number of partitions assigned. The solution is obtained using an algorithm written in MatLab 7.0. This resolution is described by Deshusses *et al*. (1995). In this model, the biotrickling filter was divided into n layers along the column and the non-lineal system equations developed were solved simultaneously using the Newton-Raphson method for multiple variables. The input variables were sampled using LHS method and a Monte Carlo simulation was performed and used for FAST and SRC methods. The same algorism was used for MOAT sampling. Ten evaluations equally spaced on the same data range were used in the previous case.

The gas phase, the liquid film (water), and the biofilm are considered as an ideally mixed layer. The contaminated air stream passes through the biotrickling filter in co-current mode. On the gas–liquid film interface equilibrium is assumed for the VRSC, also air/water partition coefficients are used. The parameters tested are diffusion coefficient (D), partition coefficient (m), maximum specific growth rate (μ_{max}), Monod constant (Ks), Yield ($Y_{x/s}$), specific surface (a_s), porosity (ε), fraction covered by biofilm (α), biofilm thickness (δ) and dispersion coefficient (W). The range of sampling was determined considering each parameter's mean and standard deviation and considering lineal distribution. Such means and deviations are calculated for four compounds (Hydrogen sulfide (H_2S), dimethyl sulfide (DMS), Methyl mercaptan (MM) and dimethyl mercaptan (DMDS)). 6000 evaluations were performed for each study parameter. The reference microorganism was *Thiobacillus thioparus*. This microorganism has been widely used for odor removal and it has shown high elimination capacities and good performance (Rattanapan and Ounsaneha, 2012).Table 1 shows the mean and standard deviation calculated. These values were determined using experimental data from different authors or estimated. A linear data range equally spaced was used in this work either for LSH sampling method (FAST and SRC) or MOAT. This input data range goes from $\bar{x}-\sigma$ to $\bar{x}+\sigma$ where $\bar{x}$ is the mean deviation and σ is the standard deviation considering a lineal distribution. The operating conditions considered were taken from Silva *et al.* (2010).

The sensitivity analysis was performed by the standard regression coefficient method (SRC), FAST and MOAT.

The SRC method considers a scatter plot (obtained from the Monte Carlo simulation) (Kleijnen and Helton, 1999) and a lineal regression in order to develop a linear model of the form

$$y = b_0 + \sum_i x_i b_i \tag{11}$$

where y is the output of the model, x_i are the parameters of the model and b_i are the regression coefficients. Those coefficients are dimensioned, and standardizing them provides a better practical use

$$\tilde{y} = \sum_i x_i \beta_i \tag{12}$$

where $\tilde{y}$ is the dimensionless output of the model and β_i are the dimensionless regression coefficients. It is possible to obtain the standard variables according to Eq. (136).

$$\tilde{y} = \frac{y - \bar{y}}{\sigma_y} \tag{13}$$

where $\bar{y}$ is the mean deviation and σ_y is the standard deviation. It is a known result of linear regression analyses that if the factors are independent and the model is linear (Saltelli *et al.*, 2005), the Eq. (14) can be assumed as valid

$$\sum_i \beta_i^2 = 1 \tag{14}$$

Table 1. List of input factors (parameters) of the bio-filtration model

Parameter	mean	Standard deviation	Units	Reference
Transport properties				
Diffusion coefficient in water (D)	1.4×10^{-9}	4.087×10^{-10}	m^2/s	Tamimi *et al.*, 1994 Chiang *et al.*, 2000 Saltzman *et al*., 1993
Partition coefficient air - water(m)	0.198	0.187	Dimensionless	Dobryakov and Vitenberg, 2006
Dispersion coefficient (W)	1.55×10^{-8}	7.7×10^{-9}	m^2/s	Perry and Green, 2007
Packing				
Specific surface (a_s)	300	50	m^{-1}	Estimated
Porosity (ε)	0.3	0.2	Dimensionless	Estimated
Surface covered by biofilm (α)	0.4	0.2	Dimensionless	Zaarok *et al*., 1998
Biofilm properties				
Thickness (δ)	30	20	μm	Spigno *et al.*, 2004
Kinetics parameters				
Specific grow rate (μ_{max})	0.044	0.039	h^{-1}	Chung *et al.*, 1996
Monod constant	50.1	42.8	g/m^3	De Zwart *et al.*, 1997
Yield	0.029	0.015	Dimensionless	Li *et al.*, 2003

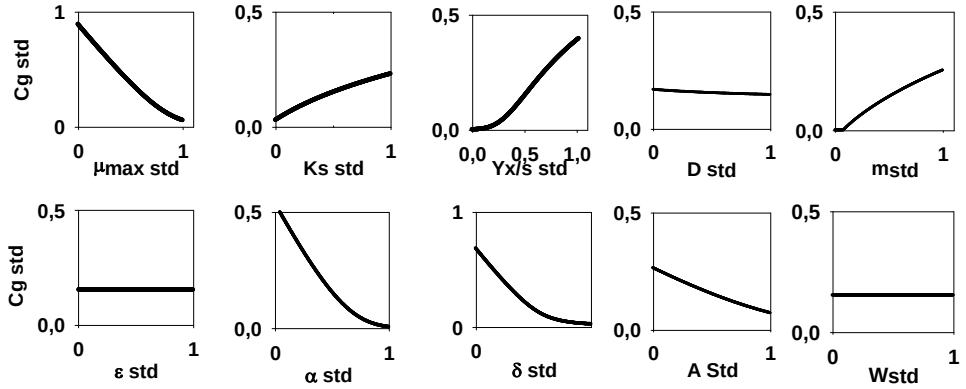

Fig. 1: Model behavior in Monte Carlo simulation.

If the model, as in our case, deviates from linearity, then the sum of the squared β's will quantify the deviation (more than 0.7) (Draper y Smith, 1998). In that case, the regression coefficients will represent the sensitivity of such parameter.

The FAST method considers the variances ratio of the input and the output of the model as shown in equation 15 and it is able to estimate directly the conditional variances characterizing the sensitivity indices (Ratto *et al.*, 2007).

$$S_i = \frac{V_{x_i}\left(E_x\left(y|x_i\right)\right)}{V\left(y\right)} \tag{15}$$

where $V(y)$ is y's variance and $E_x(y|x_i)$ is the remainder which represents the mathematical expected values of the output due to variations of x, and $V_{xi}(E_x(y|x_i))$ is called the main effect of x_i on y and represents the variance of such possible values (Saltelli *et al.*, 2004). Given that $V_{xi}(E_x(y|x_i))$ is large if x_i is influential, its ratio to the unconditional variance $V(Y)$ is used as a measure of sensitivity (Saltelli *et al.*, 2006). The Morris method operates at a lower sample size than the variance based measures, since in experimental design, when using Morris method each factor is sampled at a small number (e.g. 2, 3, 4) of selected values, called levels. For each factor, a number of one-step differences are estimated (Δ) along the various axes and the function is evaluated in these points. The differences are estimated as:

$$EE^i_{x_j} = \frac{y\left(x^i + \Delta\right) - y\left(x^i\right)}{\Delta} \tag{16}$$

where EE is the expected value. Excessive values of $EE^i_{x_j}$ are averaged and their related standard deviation determined to produce a Morris coefficient. In our study case, 8 levels were used since no changes were observed when more levels were tested.

IV. RESULTS

Figure 1 shows the results of the Monte Carlo Simulation for each parameter. In this figure, it is possible to see the parameters that do not have appreciable effect on the output.

Table 2 shows the standard regression coefficients for the SRC method. It is possible to observe that the regression performed is able to capture 94% of the outputs with a coefficient of determination of 0.84, so there is a high linearity in the effect of each parameter on the outputs. When this number is high, e.g., 0.7 or higher, we can use the standardized regression coefficients for sensitivity analysis. The diffusivity, maximum specific growth rate, specific surface, fraction covered by biofilm and biofilm thickness are negative due to an increase on the value results in the increased substrate consumption, reducing the concentration at the outlet of BTF.

Table 3 shows the results of sensitivity analysis using FAST and MOAT (where Si are the sensitivity indices for each method). It is observed that in most parameters present, low standard deviation values with a high sensitivity are found. Such deviations confirm that the system has an important linearity on uncertain parameters and that the values obtained by the MOAT are a good approximation for the relations between uncertainty and the output obtained. Moreover, it is possible to observe that the biofilm parameters have the highest influence in the model behavior while the support parameters have the lowest influence.

Figure 2 shows a comparative graph of the sensitivity for every parameter through the three methods. It is possible to observe that there is no contradiction between the magnitudes of the sensitivities of each parameter for all the used methods. The sensitivity indices obtained by MOAT method have a high correspondence with the ones obtained by SRC method. Moreover, it is possible to observe that the kinetic parameters (μ_{max}, K_S, $Y_{x/s}$) have the highest influence on the model behavior,

Table 2: Regression coefficients for SRC method

Parameter	β_i	β_i^2
D	-0,0118	0,0001
m	0,1742	0,0304
μ_{max}	-0,7213	0,5203
Ks	0,1216	0,0148
Yx/s	0,2816	0,0793
As	-0,1198	0,0144
ε	0	0
α	-0,3382	0,1144
δ	-0,41	0,1681
W	0,0003	0
Square sum		0.9418

Table 3. Sensitivity for each parameter by FAST and MOAT

Parameter	S_i^{FAST}	S_i^{MOAT}	σ_i^{MOAT}
Transport parameters			
D	0,0001	0,0136	0,0054
W	0	0	0
Sum	0,0001	0,0136	0,0136
Kinetics and biofilm parameters			
μ_{max}	0,5234	0,7054	0,1962
Ks	0,0152	0,1244	0,0317
$Y_{x/s}$	0,0810	0,2453	0,0284
α	0,1144	0,3262	0,1653
δ	0,1681	0,4038	0,2668
Sum	0,9021	1,8051	0,6884
Support parameters			
a_s	0,0144	0,1189	0,0205
ε	0	0	0
Sum	0,0144	0,1189	0,0205

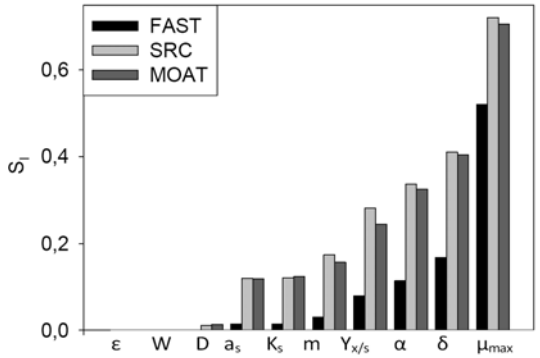

Fig. 2: Comparison of each parameter's sensitivity by different methods of sensitivity analysis

especially in the specific growth rate. Figure 3 shows the effect of the saturation constant and specific growth rate in the VRSC removal capacity, where a high slope is observed.

A lower influence is observed for the transport properties (W, m, D), especially the dispersion where the sensitivity is zero, therefore, despite of flow regime, the Péclet number would not be relevant in the BTF performance. Diffusion is another important parameter that has a low influence on the model. The average value of Thiele module was 2.8×10^{-3}. It was calculated according to Eq. (6) and using the average values of D, δ and μ_{max}. These values indicate that degradation is limited by mass transfer. Diffusion has a low sensitivity therefore can be concluded that high variations does not change the controlling pass in the biofiltration. Figure 4 shows the effect of the diffusion and distribution coefficients in the biooxidation capacity in a BTF.

The fraction covered by biofilm (α) and biofilm thickness (δ) are variables which also have an important influence on the biofiltration performance. Both are characteristics of the biofilm which even though they can be determined by experimental techniques, their control is hard, so the efforts are aimed for monitoring and evaluation. Figure 5 shows the effect of the fraction covered by biofilm (α) and biofilm thickness (δ) in the biooxidation capacity. It can be observed that these qualities play an important role in the operation and in the simulation.

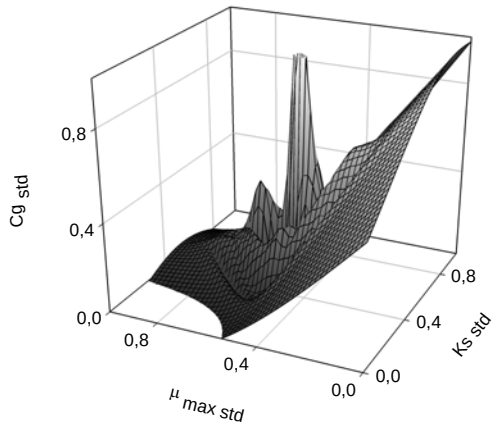

Fig. 3: Effect of specific growth rate (μ_{max}) and Monod constant (K_S) in the removal of VRCS

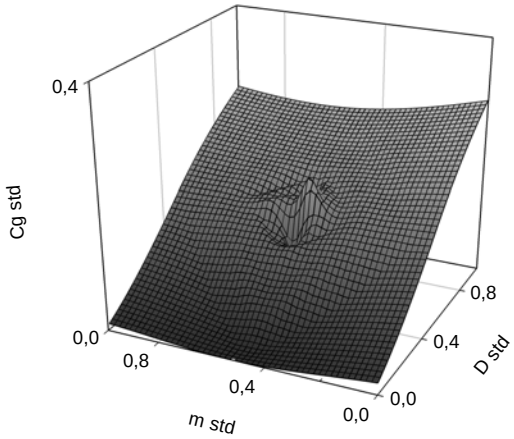

Fig. 4: Effect of diffusion (D) and partition coefficient (m) in the removal of VRSC

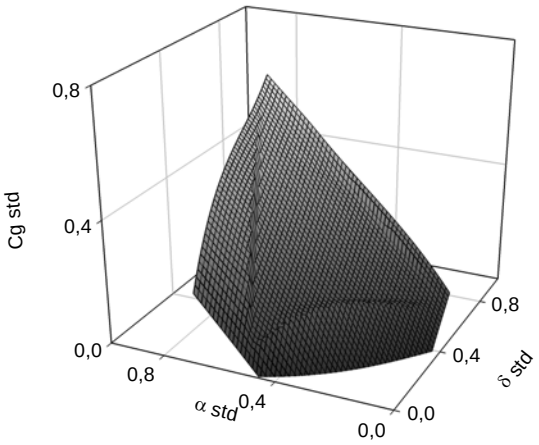

Fig. 5: Effect of the fraction covered by biofilm (α) and biofilm thickness (δ) in the removal of VRSC

V. CONCLUSIONS

There are kinetic, physicochemical and biological characteristics that determine the behavior of a BTF. The biofilm establishes the performance of the BTF, while the transport properties are less important. The gas flow

regime, despite that describes the turbulence in the gas phase, has a low influence in the model behavior, therefore the dispersion described in the column does not generate important changes in the removal capacity. The specific surface has a high influence in packed columns, however, in the case of the BTF biooxidizing VRCS the analysis shows that this parameter does not produce wide modifications in the removal capacity, therefore, effort aimed at improving the global mass transfer coefficient will not produce important changes in the removal capacity, therefore the features of the packing are not decisive in the column behavior.

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