Aerobic Digester Design for the Biodegradation of Plant Tannins in Industrial Wastewater

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This paper describes aerobic digester design for the biodegradation of plant tannins in industrial wastewater. For optimal design, using the criterion of minimal total holding time, some experimental investigations into tannins' biodegradation rate in industrial wastewater were performed in the first part of this research. The chemical oxygen demand method (COD) was applied to follow the tannins degradation rate. The kinetic parameters of a supposed Aiba's inhibition kinetic model were determined using experimental data. In the second part of the study, equations for determining the optimal volumes of two in series connected aerobic digesters were established. Furthermore, a comparison is presented of volumes between one and two in series connected aerobic digester systems regarding wastewater volume flow rate, $q_V = 120\ m^3\ d^{-1}$ and wastewater treatment efficiency, η_{COD} = 98 %.

Key words:
Digester design, plant tannins, Aiba's inhibition kinetic model, aerobic digestion, industrial wastewater

Introduction

Tannins are defined as water-soluble polyphenolic compounds of varying molecular mass.[1] They have traditionally been used for converting animal hides into leather, due to their ability to interact and precipitate proteins found in animal skin.[2] Basic tannin production for tanneries is progressively being transmitted into a wide range of industrial applications – especially those aimed at the preservation and support of a healthier environment. Tannins are known to be toxic to many microorganisms.[3] Furthermore, their inhibition properties have a large impact on the antioxidant activities of many products.[4–6] On the other hand, tannins are also natural organic pollutants.[7] They are present in pulp, paper and wood processing wastewaters.[8,9] New requirements in the world global market with regard to sustainable development require a more efficient wastewater management. Therefore, the major concern is to treat wastewaters before they are released into the environment. There are many methods available for the removal of tannins from industrial wastewater.[7,10–12]

The activated sludge process (ASP) is the most generally applied biological wastewater treatment method.[13] The biomass in ASP derives its energy through bioconversion of organic matter (OM) in wastewater. The overall indicator of OM is the chemical oxygen demand (COD).[14,15] The main component of the ASP is a continuous-flow aerated biological reactor (aerobic digester).[16] Over recent years wastewater management has become a significant cost factor and an important aspect of sustainable development. Consequently, the optimal design and operation of an aerobic digester are very important. Although wastewater treatment plants are associated with large investments, the aerobic digesters they contain are usually designed using extremely simplified and idealized kinetic models. Owing to insufficient literature available in this field, our previous work[17] determined the degradation rate of plant tannins in industrial wastewater based on a biochemical experimental method using tannins mass concentration measurements. The COD method[15] is an alternative method of following tannins degradation rate indirectly.

The main intention of this study was to design an aerobic digester for the degradation of those plant tannins present in industrial wastewater. For this purpose, the parameters of the supposed inhibition kinetic model (using COD experimental measurements) were determined first. The tannins biodegradation rate versus concentration curve shows a similar shape (with characteristic maxima) as that for autocatalytic reactions. It is well known that, for the optimal reactor design in this case, the criterion of a minimal total holding time of two serially connected CSTRs should be applied. Therefore, optimal volumes regarding industrial wastewater capacity and wastewater treatment efficiency were established in the second part of the research.

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Materials and methods

Equipment

The laboratory W10 (Armfield) bench top aerobic digester consists of a 9 L reactor vessel mounted on a vacuum formed plastic base, with a liquid feed pump, air supply and instrumentation (temperature, pH and dissolved oxygen sensors) for monitoring and controlling the process. Their technical characteristics allow for the essential features of the aerobic treatment process to be studied, without the distractions of having to settle the solids adequately enough for external recycle – a well known laboratory problem.

Industrial wastewater

Industrial wastewater (liquid waste stream after the extraction of tannins from chestnut chips) used in the laboratory research was obtained from the existing industrial production plant. In addition to dissolved plant tannins, wastewater also contains other dissolved organic compounds. The OM (tannins and other organic compounds) mass concentration in industrial wastewater indicated as COD was, $\gamma_{COD,i} = (5.10 \pm 0.10)$ kg m^{-3}. The tannins chemical analysis using the UV spectrophotometric method[18] showed that their mass fraction regarding overall OM was, $w_{T,i} = (31 \pm 2)$ %.

Aerobic biomass sludge

The aerobic biomass sludge from an existing (municipal and industrial) full-scale aerobic wastewater treatment plant was used in this study. The analysis of initial biomass showed a mixed culture of micro-organisms, with *Vorticella sp.* prevailing. The mass concentration of biomass was, γ_X = 5.0 kg m^{-3}.

Analytical methods

The dichromate method[15] was introduced for COD determination. All reagents used, sulphuric acid (Merck), silver sulphate (Kemika), potassium dichromate (J. T. Baker), mercury(II) sulphate (Fluka), ammonium iron(II) sulphate (Sigma Aldrich) and 1,10-phenanthroline monohydrate (Fluka), are commercially available. The mass concentration of biomass was determined gravimetrically.[19]

Kinetic model development

The process scheme (Fig. 1) of two unequal (different volumes) in-series connected aerobic digesters can be described by a cascade of two CSTRs.

Typically, the aeration basin (digester) is treated as a perfectly mixed vessel, and aerobic sludge is viewed as a single pseudo-species whose growth rate follows supposed kinetics. If we assume equal inlet and outlet industrial wastewater volume flow rates ($q_{V,i} = q_{V,1} = q_{V,2} = q_V$) then the degradation rate of OM expressed as COD, using the CSTR model under steady state conditions, can be described separately for aerobic digester 1, by equation:

$$-r_{COD,1} = \frac{1}{Y_{X/COD}} \mu \gamma_X = \frac{q_V}{V_1}(\gamma_{COD,i} - \gamma_{COD,1}) \quad (1)$$

and for aerobic digester 2 as follows:

$$-r_{COD,2} = \frac{1}{Y_{X/COD}} \mu \gamma_X = \frac{q_V}{V_2}(\gamma_{COD,1} - \gamma_{COD,2}) \quad (2)$$

where:

$r_{COD,1}$ – degradation rate of OM in aerobic digester 1, kg m^{-3} d^{-1}
$r_{COD,2}$ – degradation rate of OM in aerobic digester 2, kg m^{-3} d^{-1}
V_1 – volume of aerobic digester 1, m^3
V_2 – volume of aerobic digester 2, m^3
$Y_{X/COD}$ – yield (biomass regarding OM), kg kg^{-1}
γ_X – mass concentration of biomass, kg m^{-3}
μ – specific growth rate of biomass, d^{-1}
$\gamma_{COD,i}$ – inlet mass concentration of OM, kg m^{-3}
$\gamma_{COD,1}$ – outlet mass concentration of OM from digester 1, kg m^{-3}
$\gamma_{COD,2}$ – outlet mass concentration of OM from digester 2, kg m^{-3}

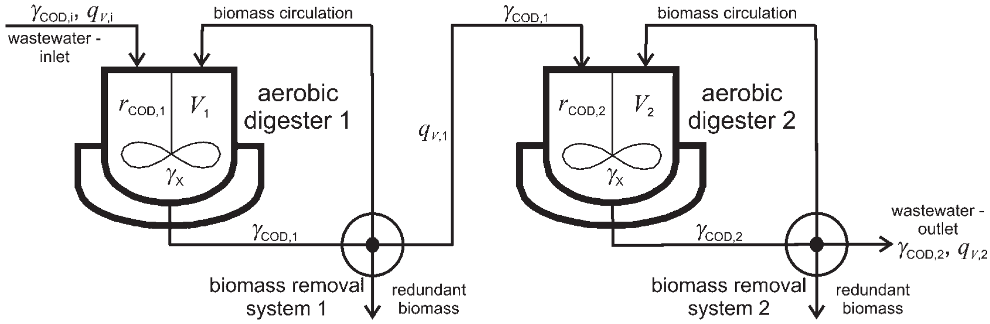

Fig. 1 – *Process scheme of two unequal in series connected aerobic digesters*

Traditionally, for wastewater treatment plants design and optimization purposes, the specific growth rate, μ, is described using simplified and idealized Monod kinetics.[16] However, the Monod equation is unsuitable in the presence of toxic compounds, which inhibit biomass growth. Therefore, some other general formulas considering inhibition were found in literature.[21–23]

It is well-known that inhibition appears during the biodegradation of phenolic wastewater.[24] Consequently, we assumed that biomass growth during OM degradation in industrial wastewater was based on Aiba's inhibition kinetic model:[25]

$$\mu = \frac{\mu_{\max}\gamma_{\mathrm{COD}}}{K_s + \gamma_{\mathrm{COD}}}\exp\left(-\frac{\gamma_{\mathrm{COD}}}{K_{\mathrm{IA}}}\right) \tag{3}$$

where:

$\mu_{\max}$ – maximum specific growth rate of biomass, $\mathrm{d^{-1}}$

K_s – saturation constant for OM, $\mathrm{kg\ m^{-3}}$

K_{IA} – Aiba's inhibition coefficient, $\mathrm{kg\ m^{-3}}$

Different assumptions, well-known from literature, were applied in our kinetic model. Firstly, the $Y_{\mathrm{X/COD}}$ is identical in the entire OM mass concentration range. Secondly, the microbial composition and viability of biomass is unchangeable. Thirdly, the γ_{X} in both digesters is constant, irrespective of q_V which is achieved with biomass circulation. By considering eq. (3) and the above assumptions, eqs. (1) and (2) can be defined as follows:

$$\begin{aligned} -r_{\mathrm{COD,1}} &= \frac{r_{\max}\gamma_{\mathrm{COD,1}}}{K_s + \gamma_{\mathrm{COD,1}}}\exp\left(-\frac{\gamma_{\mathrm{COD,1}}}{K_{\mathrm{IA}}}\right) \\ &= \frac{q_{\mathrm{V}}}{V_1}(\gamma_{\mathrm{COD,i}} - \gamma_{\mathrm{COD,1}}) \end{aligned} \tag{4}$$

$$\begin{aligned} -r_{\mathrm{COD,2}} &= \frac{r_{\max}\gamma_{\mathrm{COD,2}}}{K_s + \gamma_{\mathrm{COD,2}}}\exp\left(-\frac{\gamma_{\mathrm{COD,2}}}{K_{\mathrm{IA}}}\right) \\ &= \frac{q_{\mathrm{V}}}{V_2}(\gamma_{\mathrm{COD,1}} - \gamma_{\mathrm{COD,2}}) \end{aligned} \tag{5}$$

Thus, with further algebraic manipulation V_1 and V_2 can be calculated by:

$$\begin{aligned} V_1 = &\left(\frac{r_{\max}\gamma_{\mathrm{COD,1}}}{K_s + \gamma_{\mathrm{COD,1}}}\exp\left(-\frac{\gamma_{\mathrm{COD,1}}}{K_{\mathrm{IA}}}\right)\right)^{-1} \cdot \\ &\cdot q_V(\gamma_{\mathrm{COD,i}} - \gamma_{\mathrm{COD,1}}) \end{aligned} \tag{6}$$

$$\begin{aligned} V_2 = &\left(\frac{r_{\max}\gamma_{\mathrm{COD,2}}}{K_s + \gamma_{\mathrm{COD,2}}}\exp\left(-\frac{\gamma_{\mathrm{COD,2}}}{K_{\mathrm{IA}}}\right)\right)^{-1} \cdot \\ &\cdot q_V(\gamma_{\mathrm{COD,1}} - \gamma_{\mathrm{COD,2}}) \end{aligned} \tag{7}$$

where:

$r_{\max}$ – maximum degradation rate of OM, $\mathrm{kg\ m^{-3}\ d^{-1}}$

It is evident from eqs. (4) and (5) that only γ_{COD} is functional depending upon q_V, meanwhile other quantities are constant. Therefore, if the mass balance of OM in a steady state at various industrial wastewater dilution rates is known, parameters ($r_{\max}$, K_s and K_{IA}) can be determined using the non-linear least squares method. Dilution rate is defined as the quotient between wastewater volume flow rate and digester volume. Generally, in the case of OM inhibition presence, the curve $-r_{\mathrm{COD}} = f(\gamma_{\mathrm{COD}})$ has a specific convex-shape. Consequently, such a curve has a maximum which, when considering Aiba's expression, follows the equation:

$$\begin{aligned} \frac{\mathrm{d}r_{\mathrm{COD}}}{\mathrm{d}\gamma_{\mathrm{COD}}} = &\left(\frac{r_{\max}}{K_s + \gamma_{\mathrm{COD}}} - \frac{r_{\max}\gamma_{\mathrm{COD}}}{(K_s + \gamma_{\mathrm{COD}})^2} - \right. \\ &\left. - \frac{r_{\max}\gamma_{\mathrm{COD}}}{(K_s + \gamma_{\mathrm{COD}})K_{\mathrm{IA}}}\right)\exp\left(-\frac{\gamma_{\mathrm{COD}}}{K_{\mathrm{IA}}}\right) = 0 \end{aligned} \tag{8}$$

According to the criterion for minimal total holding time of two in series connected aerobic digesters,[20] the mathematical solution of eq. (8) presents the value $\gamma_{\mathrm{COD,1}}$ in eqs. (6) and (7). Therefore, the optimal aerobic digesters' volumes, V_1 and V_2, for desired q_V, $\gamma_{\mathrm{COD,i}}$ and $\gamma_{\mathrm{COD,2}}$, can be calculated.

Experimental

The experiments performed in the laboratory bench-top aerobic digester provided experimental data which were used for kinetic parameters determination. The outflow mass concentration of OM vs. industrial wastewater flow rate was measured. The wastewater volume flow rate was varied from 0.001 to 0.0035 $\mathrm{m^3\ d^{-1}}$. Seven experiments were carried out.

Initially, the digester was inoculated with 9 L of the aerobic biomass sludge (γ_{X} = 5.0 g $\mathrm{L^{-1}}$) and accustomed to its new environment by aeration at constant volume air flow rate ($q_{V,\mathrm{a}}$ = 1.6 L $\mathrm{min^{-1}}$) for a further 24 h. Thereupon, the aerobic digester was continuously started up using industrial wastewater at q_V = 0.001 $\mathrm{m^3\ d^{-1}}$. It was operated at constant temperature, ϑ = (25 ± 1) °C during the experimental period. The outflow mass concentrations of OM (expressed as COD) were determined daily. Furthermore, the mass concentration of biomass was also monitored daily during the operation and adjusted to the desired value (γ_{X} = 5.0 g $\mathrm{L^{-1}}$), if required. The experiment was conducted until steady-state conditions were established, i.e. until reaching constant outlet mass concentration of COD. At the same time, the mass concentration of tannins in steady state was determined using the

UV spectrophotometric method. To assure the same initial conditions and biomass sludge viability, only one experiment was performed with a single batch inoculum. Consequently, fresh biomass sludge was used for experiments at higher industrial wastewater volume flow rates.

Results and discussion

The measured outlet mass concentrations of OM, γ_{COD}, under steady-state conditions at different industrial wastewater volume flow rates, q_V, are collected in Table 1. Corresponding dilution rates, D, degradation rates of OM, $-r_{COD}$, confidence intervals, CF, and wastewater treatment efficiencies as COD, η_{COD}, were calculated. The $-r_{COD}$ is a product of D and the difference between $\gamma_{COD,i}$ = (5.10 ± 0.10) kg m^{-3} and γ_{COD}. Furthermore, the η_{COD} was calculated using the following equation:

$$\eta_{COD} = \frac{\gamma_{COD,i} - \gamma_{COD}}{\gamma_{COD,i}} \cdot 100 \tag{9}$$

The aerobic biomass sludge removed more than 97 % of the OM presented in the industrial wastewater at D = 0.11 d^{-1} (sixth column in Table 1). Moreover, the UV spectrophotometric method[18] for chemical determination of tannins showed again that mass fraction of plant tannins regarding overall OM in outflow remained almost the same as that regarding inflow (w_T = 30 %). Therefore, it can be concluded that biomass sludge from the existing wastewater treatment plant has a good degradation ability regarding plant tannins in industrial wastewater. However, the wastewater treatment efficiency decreases sharply with increasing dilution rate. The highest degradation rate for OM was achieved at D = 0.28 d^{-1} where the wastewater efficiency was only 76 %.

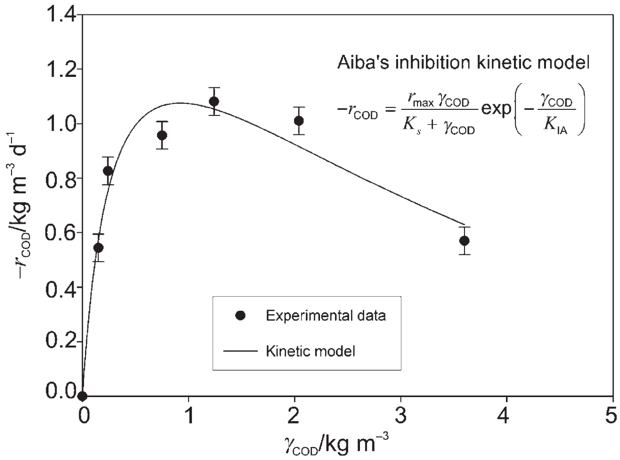

Fig. 2 – *Experimental and model-based values of degradation rate of organic matter, expressed as COD ($-r_{COD}$) in relation to the mass concentration (γ_{COD})*

Variation in the degradation rate of OM, $-r_{COD}$, with mass concentration of OM, γ_{COD}, obtained from batch experiments, is shown in Fig. 2. The Aiba's inhibition kinetic model was fitted to the experimental data of $-r_{COD}$, as a function of γ_{COD}, using the commercially available SigmaPlot® 9.0 software.[26] The biodegradation rate of OM increases with increasing OM concentration almost until γ_{COD} = 1 kg m^{-3} and then decreases with increasing OM concentration, suggesting Aiba's inhibition kinetic model. By using a non-linear least squares method, a multiple correlation coefficient, R = 0.9815 and a coefficient of determination, R^2 = 0.9633, were obtained. The R and R^2 are both measures of how well the kinetic model describes the experimental data. The R value near 1 indicates that the kinetic equation is a good description of the relationship between the independent (γ_{COD}) and dependent ($-r_{COD}$) variables. R equals 0 when the values of the independent variable does not allow any prediction of the dependent variables,

Table 1 – *Mass concentration of organic matter, expressed as COD (γ_{COD}), wastewater treatment efficiency (η_{COD}) and degradation rate of organic matter ($-r_{COD}$) with appurtenant confidence interval (CF) at steady state versus wastewater volume flow rate (q_V) and dilution rate (D) respectively*

q_V/m^3 d^{-1}	D/d^{-1}	γ_{COD}/kg m^{-3}	$-r_{COD}$/kg m^{-3} d^{-1}	CF/%	η_{COD}/%
0.0010	0.11 ± 0.01	0.150 ± 0.02	0.544 ± 0.051	9.3	97.1 ± 2.8
0.0015	0.17 ± 0.01	0.240 ± 0.02	0.826 ± 0.052	6.2	95.3 ± 2.7
0.0020	0.22 ± 0.01	0.750 ± 0.06	0.957 ± 0.050	5.3	85.3 ± 2.8
0.0025	0.28 ± 0.01	1.240 ± 0.06	1.081 ± 0.051	4.7	76.7 ± 2.7
0.0030	0.33 ± 0.01	2.040 ± 0.07	1.010 ± 0.051	5.0	60.0 ± 2.7
0.0035	0.38 ± 0.01	3.600 ± 0.08	0.570 ± 0.051	8.9	29.4 ± 2.6

and equals 1 when you can perfectly predict the dependent variables from the independent. The experimental data yielded the following Aiba's kinetic parameters' values: r_{max} = (1.87 ± 0.38) kg m^{-3} d^{-1}; K_s = (0.32 ± 0.12) kg m^{-3}; K_{IA} = (3.58 ± 0.96) kg m^{-3}, i.e.:

$$-r_{\mathrm{COD}} = \frac{1.87\,\gamma_{\mathrm{COD}}}{0.32+\gamma_{\mathrm{COD}}}\exp\left(-\frac{\gamma_{\mathrm{COD}}}{3.58}\right) \qquad (10)$$

In the second part of the research we calculated the optimal outlet mass concentration of OM in aerobic digester 1, $\gamma_{\mathrm{COD,1}}$, using the criterion of a minimal total holding time of two in-series connected aerobic digesters. When considering experimentally-defined Aiba's inhibition kinetic parameters, a numerical solution of eq. (8), $\gamma_{\mathrm{COD,1}}$ = 0.922 kg m^{-3} was obtained using '*fzero'* function in MATLAB® software (Version 7.0.0.19920 (R14)). Therefore, the minimal volumes of the two in-series connected aerobic digesters (eqs. (6) and (7)) for the degradation of wastewater from an industrial tannins production plant, can be calculated as follows:

$$V_1 = 0.932\, q_{\mathrm{V}}(\gamma_{\mathrm{COD,i}} - 0.922) \qquad (11)$$

$$V_2 = \left(\frac{1.87\,\gamma_{\mathrm{COD,2}}}{0.32+\gamma_{\mathrm{COD,2}}}\exp\left(-\frac{\gamma_{\mathrm{COD,2}}}{3.58}\right)\right)^{-1} \cdot$$

$$\cdot\, q_{\mathrm{V}}(0.922-\gamma_{\mathrm{COD,2}}) \qquad (12)$$

It is evident from eqs. (11) and (12) that V_1 and V_2 depended only on industrial wastewater volume flow rate and the inlet and outlet mass concentrations of OM. The cumulative influence of q_V and $\gamma_{\mathrm{COD,i}}$ on V_1 is plotted in Fig. 3. The effect of q_V and $\gamma_{\mathrm{COD,2}}$ on V_2 is also presented in Fig. 3. Apparently, increased $\gamma_{\mathrm{COD,i}}$ and q_V mutually increased the V_1. The same correlation is valid for q_V and V_2, meanwhile increased $\gamma_{\mathrm{COD,2}}$ or decreased wastewater efficiency reduces V_2.

Wastewater treatment plant construction beside existing tannins production plant with q_V = 120 m^3 d^{-1} and $\gamma_{\mathrm{COD,i}}$ = 5.1 kg m^{-3}, would require investments into two aerobic digesters with V_1 = 467 m^3 and V_2 = 228 m^3, respectively. An outlet mass concentration of OM, $\gamma_{\mathrm{COD,2}}$ = 0.1 kg m^{-3} is supposed. The total volume of both aerobic digesters is, V = 695 m^3. However, this is appreciably less than the volume for only one aerobic digester (V = 1386 m^3) which is calculated by eq. (6) when considering experimentally defined Aiba's inhibition kinetic parameters, q_V = 120 m^3 d^{-1}, $\gamma_{\mathrm{COD,i}}$ = 5.1 kg m^{-3}, and $\gamma_{\mathrm{COD,1}}$ = 0.1 kg m^{-3}.

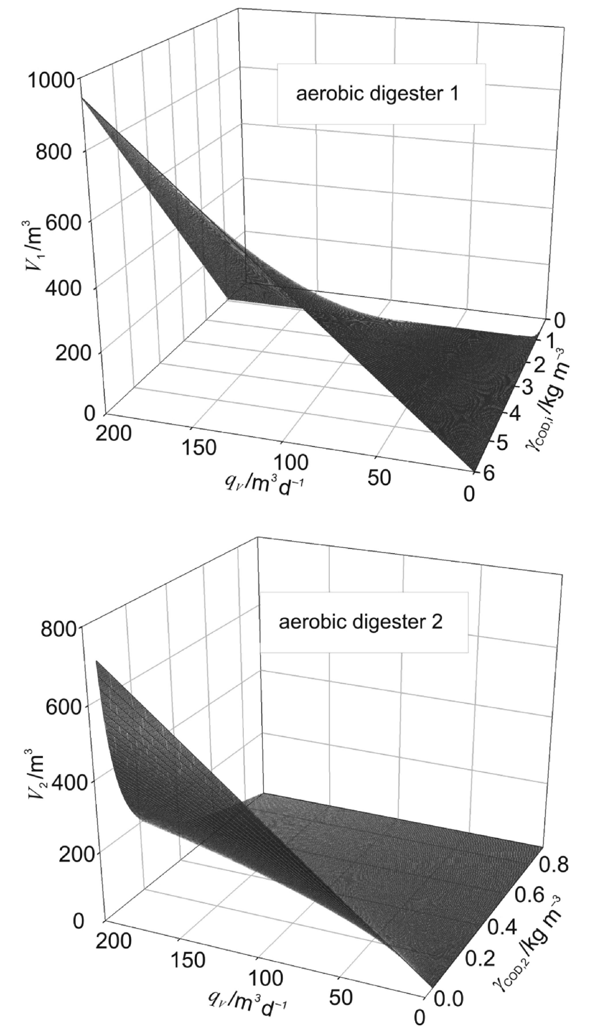

Fig. 3 – *The effect of wastewater volume flow rate (q_V), inlet mass concentration of organic matter, expressed as COD ($\gamma_{COD,i}$), and outlet mass concentration of organic matter from aerobic digester 2 ($\gamma_{COD,2}$) on volume of digester 1 (V_1) and 2 (V_2)*

Conclusion

The plant tannins presented in industrial wastewater after the extraction process cause inhibition of aerobic bacteria. However, numerous studies suggest biological methods for their degradation. Nonetheless, there is still a data deficiency regarding inhibition kinetic parameters and degradation capacity predictions for aerobic digester design. Therefore, we acquired experimental data which enabled us to determine the inhibition kinetic parameters, and the optimal volumes of two in-series connected aerobic digesters for industrial wastewater treatment.

The kinetic parameters of the Aiba's inhibition kinetic model were successfully determined using the mass balance of OM data obtained from experiments in the laboratory bench-top aerobic digester. Thereupon, we estimated the optimal volumes of two in series connected aerobic digesters using the criterion of a minimal total holding time. The assessment was based on industrial wastewater volume flow rate, q_V = 120 m^3 d^{-1} and wastewater treatment efficiency, η_{COD} = 98 %. Under these conditions two aerobic digesters with V_1 = 467 m^3 and V_2 = 228 m^3 resulted. Technological and economical authorization for a two-stage industrial wastewater treatment plant was mainly confirmed with a more than two times lower total digester volume compared to one-stage plant. Consequently, the importance of aerobic digester design using the criterion of a minimal total holding time of two in-series connected digesters was proved.

The experimentally based aerobic digester design procedure presented in this paper could be employed successfully for full-scale aerobic tannins wastewater treatment plant design.

Symbols

D – dilution rate, d^{-1}
CF – confidence interval, %
K_{IA} – Aiba's inhibition coefficient, kg m^{-3}
K_s – saturation constant for OM, kg m^{-3}
q_V – wastewater volume flow rate, m^3 d^{-1}
$q_{V,a}$ – air volume flow rate, L min^{-1}
$q_{V,i}$ – inlet wastewater volume flow rate, m^3 d^{-1}
$q_{V,1}$ – outlet wastewater volume flow rate in digester 1, m^3 d^{-1}
$q_{V,2}$ – outlet wastewater volume flow rate in digester 2, m^3 d^{-1}
R – multiple correlation coefficient
R^2 – coefficient of determination
r_{COD} – degradation rate of OM, kg m^{-3} d^{-1}
$r_{COD,1}$ – degradation rate of OM in aerobic digester 1, kg m^{-3} d^{-1}
$r_{COD,2}$ – degradation rate of OM in aerobic digester 2, kg m^{-3} d^{-1}
r_{max} – maximum degradation rate of OM, kg m^{-3} d^{-1}
V_1 – volume of aerobic digester 1, m^3
V_2 – volume of aerobic digester 2, m^3
$w_{T,i}$ – tannins mass fraction regarding overall OM in inflow, %
w_T – tannins mass fraction regarding overall OM in outflow, %
$Y_{X/COD}$ – yield (biomass regarding OM), kg kg^{-1}
γ_{COD} – mass concentration of OM, kg m^{-3}
$\gamma_{COD,1}$ – outlet mass concentration of OM from digester 1, kg m^{-3}
$\gamma_{COD,2}$ – outlet mass concentration of OM from digester 2, kg m^{-3}
$\gamma_{COD,i}$ – inlet mass concentration of OM, kg m^{-3}
γ_X – mass concentration of biomass, kg m^{-3}
η_{COD} – wastewater treatment efficiency, %
μ – specific growth rate of biomass, d^{-1}
μ_{max} – maximum specific growth rate of biomass, d^{-1}
ϑ – temperature, °C

Abbreviations

ASP – Activated Sludge Process
COD – Chemical Oxygen Demand
OM – Organic Matter
CSTR – Continuous Stirrer Tank Reactor
ISO – International Standardization Organization

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