Scientific paper

Heat Transfer in Citric Acid Production with Axial and Radial Flow Impellers

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Dedicated to the memory of the late Prof. Dr. Valentin Koloini

Abstract

In order to produce fermentation broth for downstream recovery, a total of 15 fermentations were done in a 15 m^3 and two 7.5 m^3 vessels. Apart from the evaluation of fermentation yield and productivity, information on the heat and mass transfer coefficients were required for design purposes. The focus of the fermentation study was therefore directed to obtain information on broth rheology, heat transfer aspects and considerations. Broth rheology was found to deviate from Newtonian behavior with increasing biomass concentration. Using axial flow impellers, rather than radial flow producing Rushton turbines, significantly improved heat transfer in this study.

Keywords: heat transfer coefficients, pilot scale, citric acid fermentation

1. Introduction

Effective temperature regulation and control in bioreactor systems is one of the most basic but very essential parameters for biosynthesis operation and optimization of cell growth and product formation. If all of the most sophisticated bioreactor equipment and instrumental control is perfectly functioning and temperature regulation and control in bioreactor is not operating the whole technology process is blocked.[1–3]

In bioreactor systems, temperature control is essential for optimisation of biomass production and/or

product formation. The optimum cultivation temperature is strain dependent and for sub-optimal temperatures the dependency of specific growth rate on temperature can be approximated by an Arhennius-type relationship[4].

Plant and animal cell systems are typically cultivated at temperature of 25 to 37 °C. Maintenance of the bioreactor at the desired operating temperature is achieved through either heat addition or, more usually, the removal of heat from the system, as required. In aerobic systems, heat generation and oxygen consumption are closely linked and, from a design perspective, almost impossible to separate.

While laboratory studies can be used to predict the oxygen demands of a given biological system under specified cultivation conditions, successful *scale-up* depends critically on the choice and appropriateness of models predicting heat and mass transfer performance in large-scale, three-phase systems, often exhibiting non-Newtonian characteristics.[4,5]

1. 1. Heat Balance

The overall heat balance in a bioreactor reads as:

$$\dot{Q}_r + \dot{Q}_g + \dot{Q}_{ag} = \dot{Q}_c + \dot{Q}_{ev} + \dot{Q}_{ac} \qquad (1)$$

The total rate of heat to be removed, $\dot{Q}_r$, is then:

$$\dot{Q}_r = \dot{Q}_g\ \rho_g\ Y\ r_i + Q_c\ \rho_{pc}\ c_{pc}(T_{c\ in} - T_{c\ out}) - (P_g / V)\ V\ Q_c - Q_g\ \rho_g\ c_{pg}\ (T_{g\ in} - T_{g\ out}) \quad (2)$$

The rate of heat generation is proportional to the rate of the substrate conversion, which is linked to oxygen consumption, and can be estimated with considerable accuracy on the basis of the laboratory data.

In aerobic fermentation it is possible to make a rough estimation of the specific heat generation rate if oxygen consumption rate is known:[5,6]

$$\dot{Q}_{R/V} = 5 \cdot 10^5\ (W_o^{\cdot} / V) \quad (3)$$

The oxygen uptake rate in an industrial fermentation may reach 1 to 3 · 10^{-2} mol/m^{3}s, with an estimated heat production rate of 5 to 15 kW/m^3. Additional sources of heat are due to agitation by stirrers and input of hot air and/or feeds.[3]

For effective heat exchange, bioreactors are provided with external cooling of the outside wall and/or with cooling coils immersed in the reactor. The necessary heat transfer surface, *A*, is related to the total heat exchange rate $Q^{\cdot}$ by:

$$\dot{Q} = U \cdot A \cdot \Delta T \quad (4)$$

ΔT is the log-mean temperature difference between the temperature $(T_{i\_} - < T_C >)$ of the broth and coolant or heating source, U is the overall heat transfer coefficient.

Overall heat transfer coefficient U could be calculated from Eq. 5

$$1/U = 1/h_{cw} + \Delta l / \lambda_w + 1/h_i \quad (5)$$

Terms on the right side of Eq. 6 can be considered as thermal resistances on the bioreactor broth side, the reactor or coil wall, and the coolant side, respectively. In well-designed cooling or heating systems with water as coolant, h_{cw} = 3000 W/m^{2}K is an achievable value. If stainless steel is used as constructional material for heat exchange surfaces, then $\lambda_w = 17$ W/mK and $\Delta l = 5 \cdot 10^3\ m$ are typical values for heat conductivity and wall thickness. The wall heat transfer coefficient, h_i in a well stirred reactor containing a low viscosity fermentation broth is 2000–3000 W/m^{2}K. Much lower values of h_i, in the range of 300– 1000 W/m^{2}K can be expected in broths which are non-Newtonian or which contain a considerable concentration of the polymeric compounds such as starch. Even lower values have been observed in systems with biopolymers such as xantane gum[7]. Sources of the overall heat transfer coefficient data in well-stirred tank reactors are presented in Table 1.

The heat transfer surface needed to keep the bioreactor content at constant temperature in the case of an intensive bioprocess, employing a filamentous micro-organism, at the usually available cooling water mean temperature of 20 °C, is rather large. Serious difficulties may be encountered if the optimal temperature of the bioprocess is low and the available ΔT *is* lower than 10 °C.

Table 1. The overall heat transfer coefficient across the wall in well-stirred pilot and industrial bioreactors[1]

type of ferm. broth	h_i (W/m^{2}K)	U (W/m^{2}K)	*Example*
low viscosity	2000–3000	800–1100	anaerobic water treatment
slightly non-Newtonian low viscosity	1100–1500	600–800	Bacitracin
low to medium viscosity non-Newtonian, with filamentous biomass	600–1000	300–700	salinomycin oxyteracyclin
viscous highly non-Newtonian	100–300	100–200	Xantane

1. 2. Heat Transfer Correlations in Stirred Tank Reactors

Methods used to estimate the individual heat transfer coefficient on the side of cooling or heating media in the coils are well known. The heat transfer resistance of the wall can also be easily estimated, although there might be some problems if the outer wall is only partially cooled or heated. Most uncertainties are related to estimation of the individual heat transfer coefficient on the side of fermentation broth, h_i, which has the most important effect on the overall heat transport coefficient U.

A Rushton turbine, often used as a stirrer in bioreactors, promotes radial flow and the heat transfer coils are usually placed opposite to the stirrer to be exposed to the most turbulent flow of the broth. Other types of stirrers as a propeller or a pitched blade turbine, which promote more pumping and axial flow may offer an advantage as far as is heat transfer is concerned.

For estimation of h_i in fermentation broths, there are several correlations available. The general form of correlating equations for Re > 200 proposed from Zlokarnik and Judat (1987)[8] is:

$$Nu = C\ Re^{0.67} Pr^{0.33} (\eta / \eta_w)^{0.14} = hD/\lambda = C[Nd^2 \rho / \eta]^{0.67} [c_p \eta / \lambda]^{0.33} (\eta / \eta_w)^{0.14} \quad (6)$$

Only a few studies in stirred tanks under aerated conditions are available.[9,10,11] Depending on the operation conditions, the heat transfer coefficient may decrease or increase by introduction of a gas into bioreactor.

A theoretical approach based on the penetration theory concept has been used to analyse the available experimental data,[11] relating power imput contributions due to stirrer and due to the gas to mean residence time of a liquid element at the wall. Qualitatively this approach appears sound.

At a fixed stirrer speed a decrease of the heat transfer coefficient is observed at low gassing flow rates, followed by an increase at higher values of gas velocity[11]. Under normal operation conditions in industrial scale Eq.7 could be also safely employed to estimate h_i. It is worthwhile mentioning that

$$h_i \propto (P/V)^{0.22} D^{-0.11} \tag{7}$$

At the same power input h_i decreases with bioreactor diameter, while the heat load per unit surface increases.

A comparison between heat transfer coefficients achieved under similar process conditions, but with different impeller configurations was of specific interest of this study. For this purpose the 15 m^3 vessel was equipped with standard radial flow Rushton impellers, while axial producing flow marine turbine type impellers (A315) were fitted in 7.5 m^3 vessel.

In this study heat transfer studies were focused in three main directions: the effect of different impeller configurations on heat transfer, Nusselt number dependence on system properties and comparing the predicted heat transfer coefficients when using a dimensional analysis approach, to values predicted from the application of Kolmogoroff's theory.

2. Material and Methods

2. 1. Substrate

Milled maize (corn) suspended in tap water to 20% [m/v] starch dry matter concentration. The starch slurry was hydrolysed enzymatically to dextrose syrup. Sterilisation was done in situ at 117 °C for 45 minutes.

2. 2. Microorganism

Aspergillus niger from the LIKO cultures collection was used in all experiments. All fermentations were carried out at 32 °C. Conidia taken from 7 day old cultures on worth agar slants incubated at T = 32 °C were used as inocula for the fermentation process. The initial spore concentration of the inoculum was $5 \cdot 10^7$ conidia mL^{-1} as determined spectroscopically.[12] The growth of mycelia was followed by optical microscopic observations (Wild M-20, Switzerland)

2. 3. Analysis

Total acidity was determined by titration method using 0.1 N NaOH, the amount citric acid was analyzed by capillary isotachophoresis, biomass by was determined as dry weight after filtering and drying at 105 °C for 24 hours. Glucose, maltose and higher sugars were determined by HPLC.

2. 4. On-line Data Capture

A Honeywell control system measured and recorded process parameters on a continuous cycle. Data points recorded every 15 minutes included: pH, exit pO_2 and pCO_2, fermentation broth temperature, cooling water inlet and outlet temperature and flow rate, dissolved oxygen concentration, over-pressure in vessel and air flow rate.

2. 5. Fermentation Broth Rheology

Viscosity measurements were done with a coaxial type viscometer (Rheotest 2, Germany), at the Slovak Technical University in Bratislava. Rheological measurements of flow behavior and fluid consistency index, were performed using a Rheotest 3 viscometer in a double cylinder configuration (2VEB MLW, Pruferate-Werk, Medingen, Germany).

2. 6. Engineering Parameters

Stirred tank reactor of standard configuration 15 m^3 was equipped with standard Rushton impellers, while marine turbine type impellers (A315) were fitted in one 7.5 m^3 vessel. Agitator power input was determined from electrical power measurements on agitator motor. Gas hold-up as a difference in liquid height under aerated and unaerated conditions. For calculating k_La values the steady state gas balance method was used.

A steady state heat balance was done over the reactor, by equating the cooling water load to the microbial energy input plus the mechanical energy input. Mechanical energy input was determined from the agitator power measurements and microbial heat load from the oxygen uptake rate, according to Cooney et al (1968).[13]

2. 7. Heat Transfer Coefficients

The jacket side transfer coefficient was calculated according to the correlation presented by Stein and Schmidt (1986)[14], which is based on heat transfer in pipes. For application to cooling jackets, the *thermal diameter* (4 X jacket width) was used. At the temperatures encountered, the viscosity ratio could be taken as unity. The overall coefficient, U, was calculated from Eq. 4, while the process side coefficient from Eq. 5.

Fouling on the reactor wall was neglected and the resistance through the vessel wall calculated as 0.00035 $(W/m^2/K)^{-1}$. Values determined with the above equation could thus be correlated to process variables such as viscosity, gas flow and biomass concentration.

3. Results and Discussion

Citric acid fermentation broth is reologically characterized by biomass concentration and its physiology state. The rheological properties of the fungal fermentation broth depend entirely on the concentration and morphology of the biomass[15]. While the first part from the inoculation up to 18 hours, represents a typical Newtonian fluid, in the second part from 18 to 168 hours parallel increasing of the biomass growth and pseudoplastic behavior of the fermentation broth was indicated.

The rheological behaviour of fermentation broth can be characterized over the narrow range of applicable shear rates by Ostwald-de Waale model:

$$\eta_a = K\,\gamma^{n-1} \qquad (8)$$

The average initial viscosity was 0.062 Pa s, while at a biomass concentration of 11 g/l, the average apparent viscosity increased to 0.16 Pa s.

During a citric acid production stage polysaccharides are produced by young hyphae tips as a sticky substance needed for fixation of fungal hyphae on solid matrix. In the last part of fermentation bioprocess rheology is also influenced by the lysis products that are released in fermentation substrate. This factors largely contribute the decreasing of flow behavior coefficient n. In the range of biomass concentration up to 3.0 gL^{-1}, flow behaviour index is n = 1.00–0.95, while in further growth of biomass from 3.0 to 17.5 gL^{-1} it decreases to n = 0.43 at increasing of fluid consistency index K up to 0.135 Pa s^n.

Since the viscosity of filtrate was low 0.044 Pa s and did not change significantly with time, the dependence of consistency index K on the biomass concentration was approximated by correlation

$$K = A\,X^{b} \qquad (9)$$

The values of A were from 6.0 to 8.1 · 10^{-3} and b 2.67–3.85. The value of consistency index K is also in good agreement with reported data.[16,17] Comparing the efficiency of axial and radial mixing in Stirred Tank Reactor according to their influence on heat transfer, it was found that in axial mixing system influences the development of more uniform short and thick peripheral hyphae pellets which are optimal morphological forms that enables high product yielding fermentations.[18] This results are well in line with published data.[12,15,19]

3. 1. Rate Controlling Resistance

In agreement with results presented by Stein and Schmidt (1986)[14], the cooling side coefficient (h_{cw}) was found to be in the range 1500 to 3000 W/m^2/K. A typical value of the process-side transfer coefficient, h_i, would be 300–700 W/m^2/K (Koloini[1]). This leads to the following general observations:

- At the lower value of h_i, the process side transfer is the controlling resistance

and the effect of h_{cw} on U is minimal.

- At an intermediate value of h_i = 500 W/m^2/K, U varies by 25% when h_{cw} doubles.
- When h_i reaches a value as high as 700 W/m^2/K, the transfer rate is no longer controlled by the process side only.

3. 2. Overall Heat Transfer Coefficient

In agreement with rheological measurements that characterize non-Newtonian fermentation media, U was found to be in the range $300 < U < 800$ W/m^2/K. However, when comparing Fig. 1 and 2, it is clear that there is a marked difference between the Rushton and A315 impeller performance.

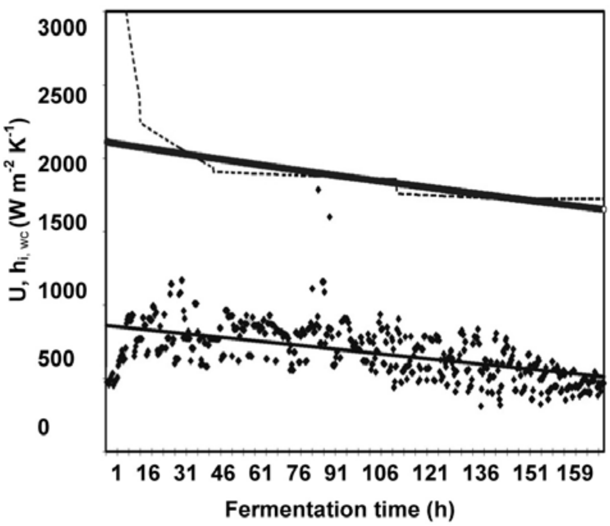

Fig 1: Heat transfer coefficients on 7.5 m^3 scale with A315 impellers
---- calculated heat transfer from the wall to fermentation media
═ approximated calculated results;
◆ overall heat transfer coefficient

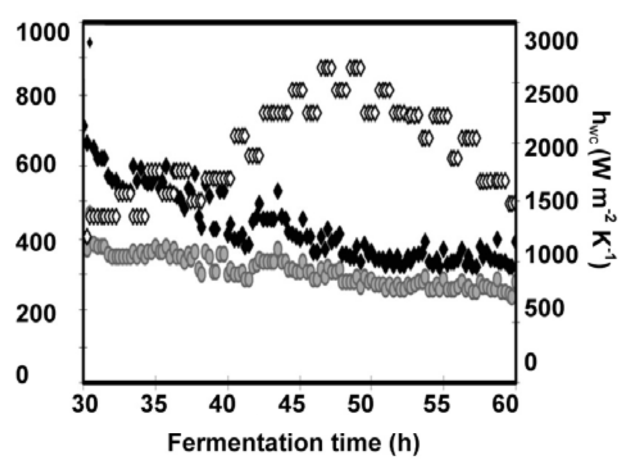

Fig 2: Heat transfer coefficients on 15 m^3 scale with Rushton impellers
◆ heat transfer from the wall to fermentation media (h_i)
◇ the cooling side coefficient (h_{cw})
● overall heat transfer coefficient

In spite of the scatter of data points, an approximate linear decrease of the overall coefficient can be observed during the course of the fermentation. With the Rushton impellers, the coefficient is considerably lower, in spite of a higher specific power input:

A definite decrease in U and h_i values are noticeable over the course of a fermentation cycle. With Rushton turbines, U is 40 to 50% lower than values observed with the A315 impellers. This is apparently not related to specific power input, as this trend was consistent at different values of P/V for the two vessels. The results achieved with both impeller configuration are due also to the different reactor sizes and possibly configuration.

3. 3. Process Side Heat Transfer Coefficient

Also included on Figs. 1 and 2 is the experimental value of h_i. Referring to the earlier comments on the rate controlling resistance, it can be stated that the process side transfer is rate controlling where values of $U \leq 400$ W/m^2/K are observed at the geometry in the present system. At values of $U \geq 600$ W/m^2/K, the cooling side transfer coefficient would also be important. From Fig.1 it can thus be deduced that h_i was not rate controlling: initial values of $U \cong 800$ W/m^2/K. This is confirmed when the experimental values of h_i are compared: for the vessel with A315 impellers, values as high as $h_i = 2000$ W/m^2/K are calculated, while for the other vessel $h_i < 600$ W/m^2/K was applied.

These values were compared to published data and values predicted by standard correlations in the literature. Although overall transfer coefficients are quoted in literature[1], published values of h_i are scarce.[20,21] The study of Mohan *et al.* [22] was used as a starting point, as they also worked with a non-Newtonian fluid. The conclusion from this comparison to previous work was that values of $300 < h_i < 500$ W/m^2/K would not be unreasonable.[23]

The Nusselt number was also calculated for each data set. Fig.3 represents the results from applying the standard correlation for heat transfer in a jacketed vessel:[8]

$$Nu = 0.74\, Re_i^{0.67}\, Pr^{0.33} (\eta / \eta_w)^{0.14} \qquad (10)$$

If the above equation is applied at constant impeller speed, with a given geometry, the viscosity becomes the only variable – as a function of biomass concentration.[24] The ratio of Newtonian to non-Newtonian viscosity was used in the last term. This is justified through the argument that the film coefficient viscosity remains Newtonian. Pseudo-plastic behaviour occurs only at higher biomass concentration.[17,18] As suggested by Edwards and Wilkinson,[25] the apparent viscosity was used in calculating the various dimensional groups. It is then possible to present the Nusselt number as a function of biomass concentration.

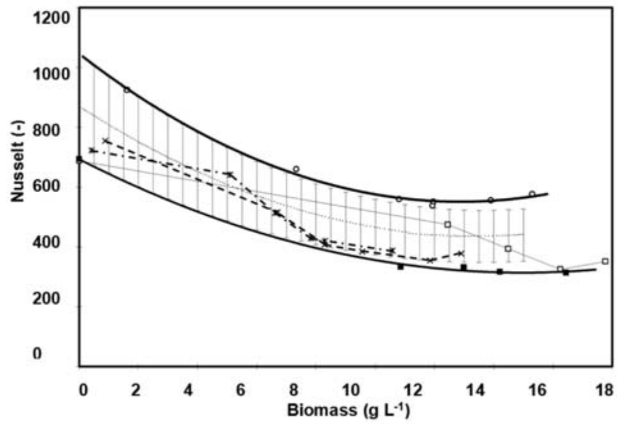

Fig 3: Nu as a function of X at constant impeller speed
Rushton impeller experiments: □ (SM-150); ∗ (SM-200 rpm)
A315 Marine turbine type impeller experiments:
■ (AE-100); × (AE-150); ○ (AE–200) correlation results ······

From the Fig. 3 it can be seen that the Nusselt number is predicted to decline as a parabola with increasing biomass. Within a ± 15% band the empirical relationship can be expressed as

$$Nu = 2.338X^2 - 63.764X + (600 + 2N) \qquad (11)$$

Taking point values of viscosity and biomass, the predicted dependence of h_i on these variables were examined. As could be expected ($\eta_w \cong 0.008X^{1.2}$, which reduces to η X at low X), the same trend of decreasing h_i–values were predicted as a function of viscosity (biomass).

3. 4. Application of Kolmogoroff's Theory of Isotropic Turbulence

As the standard heat transfer correlation did not offer an exact fit to the experimental data, the application of Kolmogoroff's theory of isotropic turbulence[26] was investigated. This predicted the heat transfer coefficient to be dependent on the specific energy dissipation rate, kinematic viscosity and Prandtl number:

$$h_i = 0.138\, (\varepsilon\nu)^{0.25}\, Pr^{-0.33} \qquad (12)$$

It should be pointed out that this equation is based on the condition of isotropic turbulence, which is only approximated in the system under investigation. The equation was applied in terms of the bulk flow conditions (average P/V) and using the apparent viscosity. Fig. 4 presents the results for the 15 m^3 scale. Also shown is the correlation from a dimensional analysis approach. Although it does seem that the equation from Kolmogoroff's theory provides a better correlation than the standard approach, a

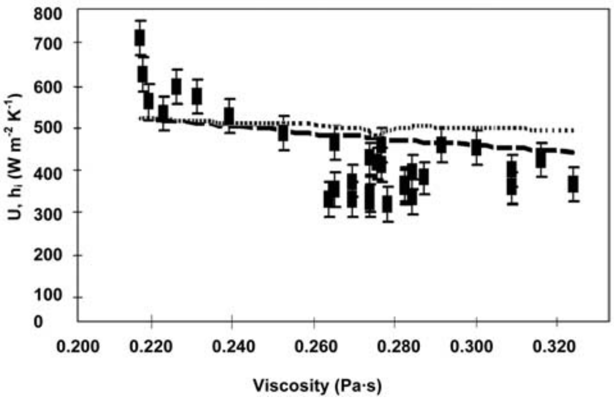

Fig.4 Correlation of h_i with Kolmogoroff's theory and dimensional analysis
■ experimental heat transfer from the wall to fermentation media
h($_{DA}$) heat transfer from the wall to fermentation media – from dimension analysis
Kolmogoroff
Note: Error bars in the above Fig.4 indicate a 25 W/m^2/K range

significant deviation from experimental data is still obvious. However, to some extent this could also be due to data scatter.

Judging from the data fit obtained in Fig. 4, it was concluded that it can not be stated that *statistically* the application of the theory of isotropic turbulence offers a better correlation. Returning to Figs. 1 and 2, the standard equation was therefore used with an appropriate proportionality constant, to fit experimental data. It was also noted that the data correlation improved if the exponent on the viscosity ratio was increased to 0.33. The final equation used to predict h_i on the above Fig.3 was thus

$$Nu = c\,(0.74)Re_i^{0.67}Pr^{0.33}(\eta/\eta_w)^{0.33} \qquad (13)$$

Apart from an initial deviation from experimental values in the region where the broth does not exhibit non-Newtonian behaviour, the correlation offers good agreement to data.

4. Conclusions

An axial flow impeller arrangement, where A315 impellers are used in combination with a Rushton turbine as the lower impeller, can improve the overall heat transfer coefficient by 40%. This improvement is not related to an increase in specific power input, but is ascribed to improved bulk blending.

The application of Kolmogoroff's theory of isotropic turbulence does not offer a statistical advantage in correctly predicting the process side heat transfer coefficient. The use of a standard correlation, based on a dimensional analysis approach is recommended. Such correlation should be corrected for non-Newtonian behaviour by appropriate use of the apparent viscosity. Probably due to the industrial interests presented data are rarely reported in the literature.

Symbols

A	surface area	(m^2)
C	proportionality constant	(/)
c_{pg}	specific heat capacity of the gas phase	(Jkg^{-1} K^{-1})
c_{pl}	specific heat capacity of cooling liquid	(Jkg^{-1} K^{-1})
c_{pg}	specific heat capacity of the gas phase	(Jkg^{-1} K^{-1})
c_{pl}	specific heat capacity of cooling liquid	(Jkg^{-1} K^{-1})
D	diameter	(m)
h	heat transfer coefficient	(Wm^{-2}K^{-1})
h_{cw}	coil side heat transfer coefficient	(Wm^{-2}K^{-1})
h_i	heat transfer from the wall to fermentation media	(Wm^{-2} K^{-1})
K	fluid consistency index	(Pas^n)
Δl	wall thickness	(m)
n	flow behaviour index	(/)
N	impeller speed	(s^{-1})
P	ungassed power	(W)
P/V	power imput	(Wm^{-3})
$\dot{Q}$	heat load	(W)
$\dot{Q}_{ag}$	rate of heat by agitation	(W)
$\dot{Q}_g$	rate of heat by aeration	(W)
$\dot{Q}_{ev}$	rate of heat by evaporation	(W)
$\dot{Q}_{R/V}$	heat of reaction rate	(W)
$\dot{Q}_c$	rate of heat by cooling	(W)
$\dot{Q}_{ac}$	rate of heat by accumulation	(W)
$\dot{Q}_r$	total rate of heat to be removed	(W)
T_g *in*	inlet gas temperature	(K)
T_g *out*	outlet gas temperature	(K)
T_c *in*	inlet cooling liquid temperature	(K)
T_c *out*	outlet cooling liquid temperature	(K)
U	overall heat transfer coefficient	(Wm^{-2}K^{-1})
$\dot{W}_o$	oxygen transfer rate	(kgm^{-3} s^{-1})
X	biomass	(gl^{-1})
Y	air saturation humidity	(kgm^{-3})

Dimensionless numbers

Nu	Nusselt number	(hD/λ) (/)
Pr	Prandtl number	($c_p\eta/\lambda$) (/)
Re	Reynolds number	(Nd^{2}ρ/η) (/)
Re_i	Reynolds number in fermentation media	(Nd^{2}ρ/η) (/)

Greek alphabeth

ε	specific energy dissipation	(kW/m^{3})
λ_w	termal conductivity of the wall	(Wm^{-1}K^{-1})
η	dynamic viscosity	(Pas)
η_w	viscosity of the liquid at the wall	(Pas)
ρ	medium density	(kgm^{-3})
ν	kinematic viscosity	(m^2 s^{-1})

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Povzetek

Prikazani rezultati se nanašajo na raziskave prenosa toplote v bioreaktorjih ob uporabi aksialnih mešal A315 in radijalnih mešal Rustonovih diskastih turbin v fermentaciji citronske kisline, na kompleksnem substratu melase sladkorne pese, v pilotnem 7.5 m^3 in 15 m^3 industrijskem bioreaktorju. Reologija fermentacijske brozge biosinteze citronske kisline z glivo *Aspergillus niger* je odvisna od glivine morfologije in se spreminja iz newtonijske v pseudoplastično. Prikazane meritve in izračuni kažejo, da aksialna mešala A315 v veliki meri izboljšajo prenos toplote med fermentacijo v obeh nivojih bioreaktorjev.