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Influence of Viscosity on Mass Transfer Performance of Unbaffled Stirred Vessels

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Unbaffled stirred tanks are seldom employed in the process industry as they are considered poorer mixers than baffled vessels. However they may be expected to provide significant advantages in a wide range of applications (e.g. crystallization, food and pharmaceutical processes, etc) where the presence of baffles is often undesirable. Moreover, in plants or animal cell cultivation bioreactors, where cell damage is often caused by bursting bubbles at the air –medium interface (Barret *et al*., 2010), they can provide sufficient mass transfer through the free surface vortex, so bubble formation and subsequent bursting inside the reactor can be conveniently avoided (Scargiali *et al*., 2012).
In this work the influence of viscosity on oxygen transfer performance of an unbaffled stirred vessel is investigated in view of its use as a biochemical reactor for animal cell growth.
Liquid viscosity was increased by adding weighted amounts of polyvinylpyrrolidone (PVP) to distilled water. Experimental results show that at rotational speeds lower than the critical one (N_{crit} , at which the free surface vortex reaches the impeller), despite the absence of gas dispersion inside the reactor and relevant cell damage due to bubble bursting, gas-liquid mass transfer is not adversely affected by viscosity and the systems remains able to provide sufficient oxygen for typical animal cell cultures. At rotational speeds higher than N_{crit} air entrapment and dispersion occurs inside the reactor and an increase of mass transfer performance is observed while increasing viscosity, probably due to smaller bubble coalescence rates due in turn to the viscosity increase itself as well as to gas-liquid interface modifications by PVP.

1. Experimental

The experimental apparatus employed is depicted in Figure 1. It involved a cylindrical stirred reactor (D_T = 190mm) with a total height of 300mm. A “Rushton turbine” (six flat blade disk mounted), 65 mm in diameter was mounted on the 17 mm dia. shaft, leaving a clearance of T/3 from vessel bottom. The vessel was filled up to an height of 190 mm (H=T) under no agitation conditions.
A static frictionless turntable and a precision scale were employed for measuring the mechanical power dissipated by the impeller at various agitation speeds for each liquid viscosity investigated. Details of this apparatus may be found in Brucato *et al*. 2010.
The volumetric mass transfer coefficient, k_La, was assessed by unsteady state experiments by means of the simplified dynamic pressure method (SDPM) (Scargiali *et al*., 2010 a and b).
The reactor was equipped with a stainless steel top to allow for reactor evacuation. Oxygen concentration was measured by means of an electrode sensor (WTW CellOx 325) and control unit (WTW Oxi 340i). The electrode time constant was experimentally assessed to be about 3.0 s, hence a value small enough for not affecting in any way the k_La values here measured (Scargiali *et al*., 2010a). In all runs temperature inside the reactor was maintained at 16°C ±0.1 by means of a refrigeration system situated downstream the oxygen probe as depicted in Fig. 1.
Liquid viscosity was increased by adding weighted amounts of polyvinylpyrrolidone (PVP) to distilled water in order to obtain liquid viscosities ranging from 1 to 9 cP. PVP amounts used to obtain relevant liquid viscosities at 16 °C are reported in Table 1.

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Rotational speed ranged from 100 to 1300 rpm in order to explore different fluid-dynamics regimes occurring inside the unbaffled stirred reactor.

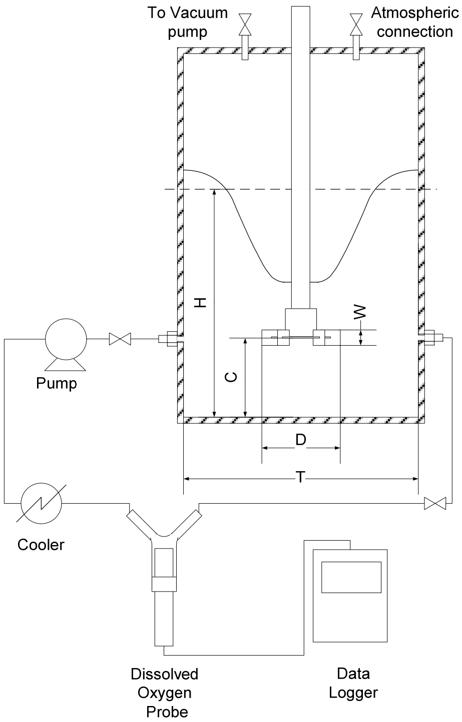

Fig.1: Schematic diagram of the experimental apparatus

2. Results and discussions

2.1 Power consumption

The specific power dissipations obtained at the various agitation speeds by the static frictionless turntable are reported in Figure 2 for each liquid viscosity investigated. As it can be seen a steep increase of power dissipation with rotational speed is observed for the three liquid viscosities investigated and, for each rotational speed, power dissipation increases as liquid viscosity increases, as it could have been expected. In the same figure black symbols indicate rotational speed (N_{crit}) and relevant specific power dissipation at which the free surface vortex reaches the impeller. Visual observation of the reactor shows that, for all three liquid viscosities investigated, at rotational speeds lower than the N_{crit} no bubbles are observed inside the liquid phase, while for $N > N_{crit}$ air is ingested from the free surface vortex and vivid bubble dispersion is observable in the liquid phase. Notably, a weak increase of N_{crit} with liquid viscosity was also observed. The relevant total power dissipation was translated into power number (N_p) values, defined as

$$N_p = \frac{P}{\rho_L \, N^3 \, D^5} \tag{1}$$

where P is agitation power, ρ_L is liquid density, N is agitation speed (s^{-1}) and D is impeller diameter.
Power Number results are reported in Figure 3 for the three liquid viscosities here investigated. As it can be observed power number is almost-constant/slightly-decreasing as long as the free surface vortex does not reaches the impeller (non-aerated regime, $N<N_{crit}$), consistently with previous observations by Ruston (Rushton *et al*., 1950), while a neatly decreasing trend is observed after the vortex reaches the impeller and air bubbles begin to be dispersed inside the reactor ($N>N_{crit}$). Clearly in this regime the growing air dispersion rate inside the reactor gives rise to increasingly larger cavities behind stirrer blades that bring

about an important reduction of power consumption, as already observed in sparged gas-liquid reactors by many authors (Paul *et al.*, 2004).

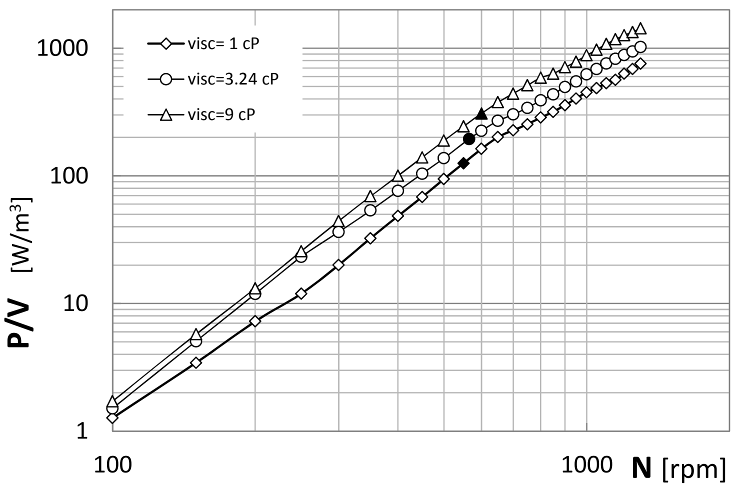

Fig.2: Experimental specific power dissipation versus rotational speed for the three liquid viscosities investigated. Black symbols refer to the critical rotational speed (N_{crit}) at which the free surface vortex reaches the impeller.

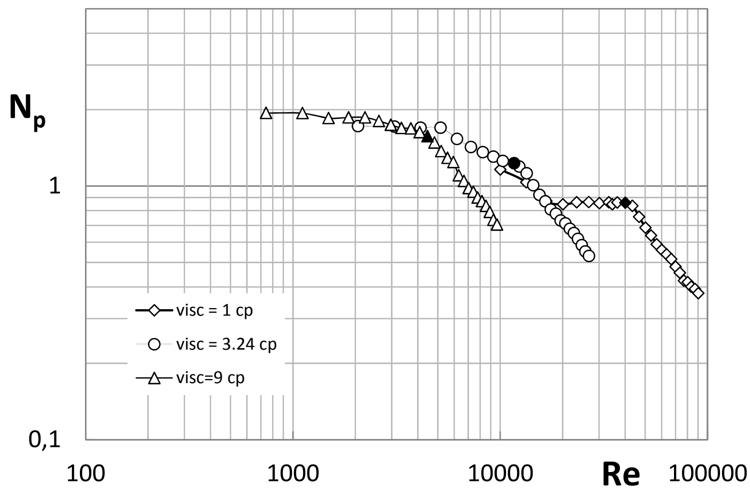

Fig.3: Experimental power number N_p versus Reynolds number for the three liquid viscosities investigated. Black symbols refer to the critical Reynolds number at which the free surface vortex reaches the impeller.

2.2 *Gas-liquid mass transfer*

The k_La values obtained by means of the SDPM technique (Scargiali et al., 2010b) for the three liquid viscosities investigated are reported in Figure 4 as a function of rotational speed. In the same figure black symbols mark critical agitation speeds (N_{crit}) and relevant mass transfer coefficients (k_La_{crit}) at which the free surface vortex reaches the impeller. As it is possible to see, for all three liquid viscosities here investigated, the mass transfer coefficient increases as rotational speed is increased, showing three different regions corresponding to different fluid dynamics regimes.

In particular, at the lowest rotational speeds, below about 200 rpm for all the three liquid viscosities investigated, the liquid free surface is almost flat, no bubbles are present in the liquid phase and gas-liquid mass transfer occurs only through the flat surface. In this range the k_La varies only due to k_L increase, being the inter-phase surface area almost constant. The relevant k_La values are very low and adversely affected by viscosity, as it could have been expected, Notably k_La is found to increase with N with an exponent slightly larger than 0,5, the expected result on the basis of penetration theory for constant interfacial area.

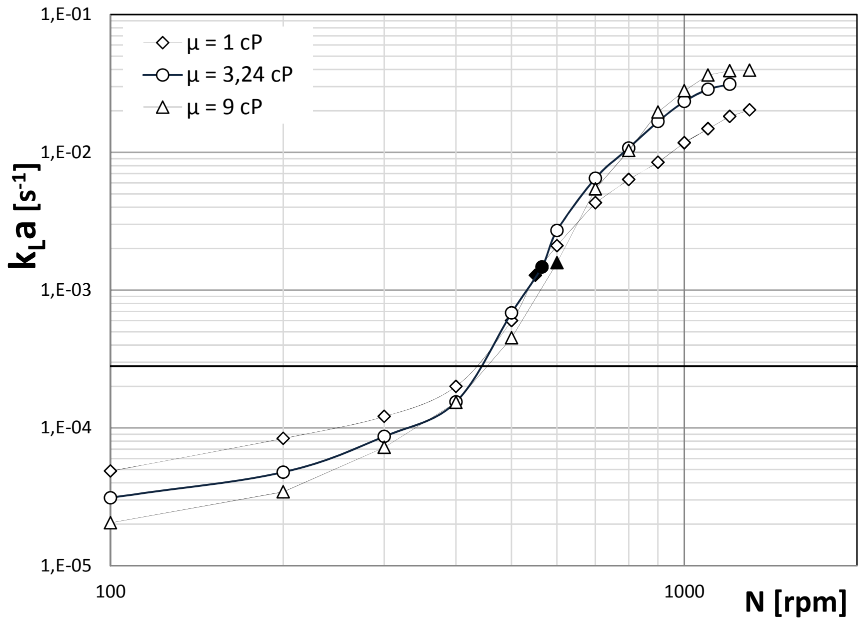

Fig.4: Experimental mass transfer coefficient vs rotational speed for the three liquid viscosities investigated. Solid symbols mark the critical rotational speed at which the free surface vortex reaches the impeller (N_{crit}) and relevant mass transfer coefficients. The solid horizontal line represents the minimum k_La requirements for animal cell culture according to Barrett et al (2010).

At rotational speeds higher than 300 rpm the free surface vortex becomes progressively more pronounced up to reaching the impeller at a rotational speed of about 550, 565 and 600 rpm for the 1, 3.24 and 9 cp liquid viscosities respectively. In this range, no gas dispersion is present inside the reactor while interfacial area significantly increases due to vortex formation, making k_La versus N slope of the order of 5, as it can be observed in Figure 4. This is a steeper dependence than expected on the basis of vortex shape change, possibly related to the fact that the resulting vortex surface is rippling rather than smooth, and therefore the relevant area increase is larger than estimated on the basis of vortex shape. At rotational speeds higher than 550-600 rpm air starts being ingested in the liquid phase and the consequent gas dispersion counterbalances the lack of further increase of central vortex interfacial area. As a consequence k_La keeps growing with an exponent smaller than 5 but still significantly larger than 0.5.

It is worth noting that while at the lowest rotational speeds (100 and 200 rpm) mass transfer coefficient quite strongly decreases while increasing viscosity (k_La approximately proportional to $\mu^{-0.5}$), at rotational speeds closer to N_{crit} the influence of viscosity becomes much smaller, and beyond the critical point it becomes even direct (higher mass transfer the higher the viscosity), probably due to smaller and smaller bubble coalescence rates due in turn to the viscosity increase itself as well as to gas-liquid inter-phase modifications by PVP.

For the sake of clarity, the critical values for rotational speed (N_{crit}), mass transfer coefficient (k_La_{crit}), specific power input (P/V_{crit}) and power number (Np_{crit}), for the various liquid viscosities investigated, are reported in Table 1 together with relevant PVP concentrations (wt%). It is worth noting that when liquid viscosity increases critical conditions are achieved at increasing rotational speeds and consequently increasing specific power dissipations as it might have been expected. Also, at higher liquid viscosities increasing critical power numbers and slightly increasing critical mass transfer coefficients are observed.

Table 1: Liquid viscosities investigated and relevant PVP amounts, (wt%) critical rotational speeds (N_{crit}) at which the free surface vortex reaches the impeller, specific power inputs (P/V_{crit}), power numbers (N_p) and mass transfer coefficients k_La. (T= 16°C for all experiments).

Liquid viscosity [cP]	*PVP wt %*	N_{crit} [rpm]	P/V_{crit} [W/m^3]	Np_{crit}	k_La_{crit} [s^{-1}]
1	0	400	125	0,86	1,30*10^{-3}
3,24	1	550	195	1,23	1,47*10^{-3}
9	2,36	650	308	1,57	1,58*10^{-3}

It is worth noting that in order to avoid gas dispersion inside the reactor, so preserving cells from bubble burst damage, an operating agitation speed smaller than the critical one (N_{crit}) should be used. In the present system this implies a maximum k_La value which ranges from 1.3*10^{-3} to 1.58*10^{-3} s^{-1} for the 1 cP and 9 cP solutions respectively, as highlighted in Table 1 or by the black symbols in Figure 4. Both values are much higher than the minimum value of about 2.8*10^{-4} s^{-1}, reported in Fig.4 as a solid horizontal line, required to support animal cell cultures at typical culture concentrations of about 10^6 – 10^7 cells/ml (Barret et al., 2010). It can be concluded that an unbaffled vessel equipped with a Rushton turbine and operated at sub-critical agitation speeds, is fully able to satisfy the Oxygen transfer rate demand of current animal cell cultures. In practice, the range of values above the solid line and below the solid black symbols may be regarded as the operating range of each liquid viscosity for shear sensitive cultures, and as it can be seen, at all viscosities it is wider than 100 rpm, a fully viable operating range. This depends on the circumstance that the highest mass transfer coefficient achieved in sub-critical conditions is always well above the black solid line, and actually slightly increases with viscosity. As a consequence this kind of bioreactor is found to provide sufficient oxygen mass transfer for animal cell growth independently of liquid viscosity, so confirming itself as a valid alternative to more common sparged reactors also for cultures involving high viscosity media.

2.3 Comparison with sparged baffled vessels

For the sake of comparison with baffled vessel performance, In Figure 5 the same data of Figure 4 are reported versus *specific power input*, together with baffled vessel data obtained with deionized water (Scargiali *et al*., 2010) and Xantham-gum solutions (Garcia-Ochoa & Gomez, 2004). As in previous figures, critical conditions are marked by a black symbol. Notably, at the highest agitation powers (i.e. speeds) the present data remarkably gather together regardless of viscosity as if P/V were the main (if not the sole) correlating parameter in such conditions. This finding marks a different behaviour with respect to literature data on baffled sparged tanks. For instance Garcia Ochoa and Gomez (2004) found a k_La dependence on liquid viscosity raised to an exponent ranging from -0.5 to -0.9 while Nocentini et al. (1993) found an even steeper dependence. Reasons behind the different behaviour observed are not clear and surely deserve further investigation. In any case, if confirmed by further experimental evidence, this finding would mark a further and notable advantage of unbaffled over baffled tank bioreactors. As a matter of fact, a closer observation of Fig.5 shows that at the highest agitation powers unsparged-unbaffled tanks are at least as efficient as (if not more efficient than) sparged baffled tanks. This is a noticeable result in view of the fact that self-ingesting unbaffled tanks, by avoiding the need of a gas-sparger, also skip the relevant power demand and operating difficulties (e.g. sparger clogging). It may be concluded that unbaffled self-ingesting tanks may well be a viable alternative to baffled sparged vessels also for non shear sensitive cultures.

3. Conclusions

Influence of viscosity on oxygen transfer performance of an unbaffled stirred vessel has been presented in view of its use as a biochemical reactor for animal cell growth.

Experimental results may be summarized as follows: (i) at low rotational speeds and flat free surface the mass transfer coefficient decreases while increasing viscosity with an exponent of about -0.5; (ii) for N closer to, yet smaller than, N_{crit} the dependence on viscosity becomes much smaller and the maximum k_La at no-aeration conditions is even increased, confirming the suitability of these bioreactors for animal cell growth, also with viscous culture broths; (iii) at rotational speeds higher than N_{crit} air entrapment and

dispersion occurs inside the reactor and an increase of mass transfer performance is observed while increasing viscosity, probably due to smaller bubble coalescence rates; k_La values in these conditions are found to be as large as, if not larger than, relevant values obtained in sparged baffled tanks.
These results suggest that unbaffled stirred vessels should be regarded as an interesting option for shear-sensitive as well as for non-shear-sensitive cultures.

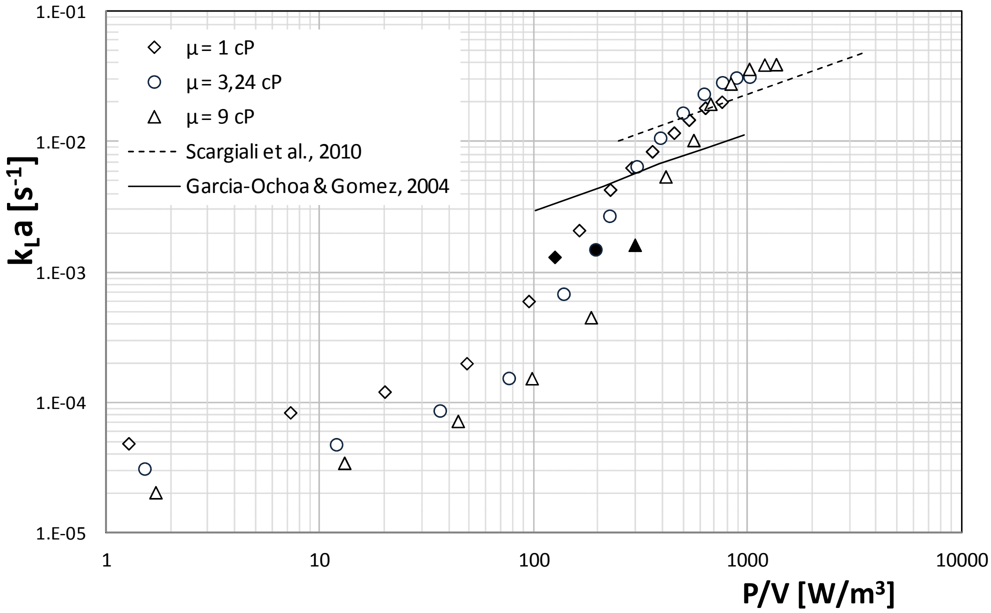

Fig.5: Comparison between present unbaffled data and literature data obtained in baffled sparged tanks. Dashed line: Scargiali et al., 2010, 1 cP, 0.5 vvm, Rushton turbine. Solid line: Garcia-Ochoa & Gomez, 2004, 2.7 cP, 0.5 vvm, stirrer 4FBT.

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