Rotor-stator spinning disc reactor

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Rotor-Stator Spinning Disc Reactor

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Summary

**Rotor-Stator Spinning Disc Reactor**

The chemical industry is continuously working to make the production more efficient and safer. Process intensification is the trend in which new equipment and processing methods are developed, which require less energy, are safer and produce less waste products. The main improvements can be made by increasing the mass and heat transfer rates significantly. The mass transfer from the gas phase to the liquid phase, and from the liquid phase to the solid phase are often the rate limiting steps in multiphase reactors. Increasing the mass transfer rates therefore leads to higher reaction rates, and thus more productivity or a lower reactor volume, thus leading to more efficient and safer reactors. This thesis describes the hydrodynamics and mass transfer in a novel type of multiphase reactor, the rotor-stator spinning disc reactor, which shows high mass transfer rates compared to conventional reactor equipment.

The rotor-stator spinning disc reactor consists of a rotating disc in a cylindrical housing. The distance between the rotor and the reactor wall, the stator, is small, in the order of 1 mm. Two reactor configurations are studied. In the first configuration, the liquid, which is the continuous phase, is injected to the reactor from the top. The gas phase is injected through a small gas inlet in the bottom stator, near the rim of the rotor. Gas bubbles are sheared off at the gas inlet, due to the high velocity gradient, and thus shear force, between the rotor and the stator. The gas bubble size decreases with increasing rotational disc speed. The centrifugal force causes the gas bubbles to move radially inward; the gas holdup in the rotor-stator spinning disc reactor with a single gas inlet in the bottom stator is only a few percent. The inward radial velocity decreases with decreasing bubble size; the residence time, and thus the gas holdup, increases with increasing rotational disc speed. The decrease in gas bubble size, combined with the increase in gas holdup, leads to an increase in gas-liquid interfacial area ($a_{GL}$) with increasing rotational disc speed.
Two types of gas bubbles are distinguished in the rotor-stator spinning disc reactor. At low rotational disc speeds, the gas bubbles are larger than the rotor-stator distance. The main gas-liquid interfacial area, and thus the main part of the mass transfer occurring, is between the gas bubble and the liquid films on the rotor and the stator. At higher rotational disc speeds, the gas bubbles are smaller than the rotor-stator distance. The mass transfer coefficient \( k_{GL} \) in the latter case is determined by the size and the velocity of the turbulent eddies in the liquid, and is therefore a function of the energy dissipation rate in the reactor.

The volumetric gas-liquid mass transfer coefficient \( k_{GL}a_{GL} \), is the product of the interfacial area and the mass transfer coefficient, and it therefore also increases with increasing rotational disc speed. It increases with increasing gas flow rate, increasing rotor radius and decreasing rotor-stator distance. The maximum value obtained in this study, measured using the desorption of oxygen from water, is \( 2.5 \text{ m}^3 \text{L}^{-1} \text{m}^{-3} \text{R}^{-1} \), at a gas flow rate of \( 1.5 \cdot 10^{-5} \text{ m}^3 \text{s}^{-1} \) and a rotational disc speed of \( 209 \text{ rad s}^{-1} \), using a rotor with 0.135 m radius and 1 mm rotor-stator distance. This is one order of magnitude higher than in conventional reactor systems, such as stirred tank reactors or bubble columns.

The energy dissipation rate in the rotor-stator spinning disc reactor is up to 3 orders of magnitude higher than in conventional reactors, such as bubble columns or stirred tank reactors. The increase in gas-liquid mass transfer is only a factor of 20. The rate of gas-liquid mass transfer per unit of energy dissipation \( \left( \frac{k_{GL}a_{GL}}{E_d} \right) \) is only \( 1.1 \text{ m}^3 \text{L}^{-1} \text{MJ}^{-1} \), while it is \( 80 \text{ m}^3 \text{L}^{-1} \text{MJ}^{-1} \) for stirred tank reactors. Increasing the rotor radius from 0.066 m to 0.135 m increases the volumetric mass transfer coefficient by a factor 3, while the energy input increases with a factor of 15. It is therefore, from energetic point of view, preferred to scale up the reactor by using multiple rotor-stator stages in series, instead of scaling up by increasing the rotor size. For the scale up by numbering up, of the reactor configuration with a single gas inlet, however, a gas redistribution system may be needed, which may prove to be inconvenient in practice.

In an alternative reactor configuration gas and liquid are fed together to the top of the reactor. The liquid forms a thin film, which flows outwards, on the rotor. The gas phase fills up the space between this liquid film and the top stator. Near the rim of the rotor, small gas bubbles are sheared off from the gas phase. The region surrounding the rim of the disc, and the region between the rotor and the bottom stator, is filled with gas bubbles dispersed in the liquid, which is the continuous phase. The gas-liquid mass transfer rate in the latter part, the dispersed flow region, is higher than in the film flow region. The mass transfer performance \( k_{GL}a_{GL}VR \), i.e. the mass transfer rate divided by the driving force, of the configuration where
gas and liquid are fed together to the reactor, is twice as high as in the case of the single gas inlet in the bottom stator.

The scale up of the configuration where gas and liquid are fed together is relatively easy. Multiple rotor-stator units can be added in series, preferably with the rotors on a common axis. It is shown that the mass transfer per stage is equal to the mass transfer in the single stage reactor, it is therefore highly probable that the same flow behaviour (film flow and dispersed flow) is obtained in the multistage reactor as in the single stage reactor. The pressure drop in the reactor is mainly caused by the centrifugal pressure; an increase in liquid flow rate leads to an increase in pressure drop. Additionally, in the case of gas and liquid flow, the pressure drop is up to a factor 2.5 higher than in the case of liquid flow only; the highest value measured is 0.64 bar pressure drop per stage, at a rotational disc speed of 459 rad s$^{-1}$.

A heterogeneously catalyzed reaction, the oxidation of glucose on platinum, is performed in the rotor-stator spinning disc reactor. The catalyst is supported on the rotor using a Nafion coating, the reaction is performed with a liquid phase only. The reaction rate is, under the conditions used, influenced by the kinetics of the reaction as well as by the mass transfer from the bulk of the liquid to the rotor. The liquid-solid mass transfer coefficient increases with increasing rotational disc speed. At low rotational disc speeds the flow is laminar, with an increasing rotational disc speed a transition towards turbulent flow will take place. The maximum liquid-solid mass transfer coefficient ($k_{LS}$) obtained is $8 \cdot 10^{-4} \text{m}_L^3 \text{m}_L^{-2} \text{s}^{-1}$ at 157 rad s$^{-1}$, which is one order of magnitude higher than in conventional reactors like a fixed bed reactor. The liquid-solid interfacial area ($a_{LS}$) that can be obtained, if the stators and the rotor are used to deposit catalyst, is $2000 \text{m}_L^2 \text{m}_R^{-3}$, which is comparable to a fixed bed reactor; the volumetric liquid-solid mass transfer coefficient ($k_{LSa_{LS}}$) is thus an order of magnitude higher.

The high gas-liquid and liquid-solid mass transfer rates give the rotor-stator spinning disc reactor a large potential for multiphase reactions. The energy input is relatively high, but the mass transfer coefficients are an order of magnitude higher than in conventional reactor systems. The rotor-stator spinning disc reactor is therefore mainly suitable for reactions where the energy costs play a minor role, and conversion and selectivity are more important. Also reactions with dangerous reactants or products and reactions at high pressure can benefit from the spinning disc reactor, since a large decrease in volume can be achieved. Additionally, the high heat transfer coefficients expected make the spinning disc reactor a promising alternative for exothermic reactions. The high mass transfer coefficients can also have a significant effect on the selectivity of a reaction, thereby giving scope for improved processes.
Samenvatting

Rotor-Stator Spinning Disc Reactor

De chemische industrie streeft ernaar de productie veiliger en efficiënter te maken. Proces-intensificatie is de trend waarbij nieuwe apparaten en processen worden ontwikkeld die significanet minder energie gebruiken, veiliger zijn en minder afval produceren dan conventionele processen. De belangrijkste verbeteringen kunnen behaald worden door de stofoverdracht en warmteoverdracht te versnellen. De stofoverdracht van de gasfase naar de vloeistoffase, en van de vloeistoffase naar de vaste fase beperken in veel gevallen de reactiesnelheid in meerfasereactoren. Hogere stofoverdracht leidt dus tot snellere reacties, en dus een hogere productie of een kleiner reactorvolume, en dus tot efficiëntere en veiligere reactoren. In dit proefschrift worden de hydrodynamica en stofoverdracht in een nieuw type meerfase reactor beschreven. Deze reactor, de rotor-stator spinning disc reactor, heeft een significant hogere stofoverdracht dan conventionele reactoren.

De rotor-stator spinning disc reactor bestaat uit een roterende schijf (rotor), in een cilindrische kamer. De afstand tussen de rotor en de reactorwand (stator) is klein, ordegrootte 1 mm. Twee verschillende reactorconfiguraties zijn gebruikt in dit onderzoek. In de eerste configuratie wordt de vloeistof, de continue fase, aan de bovenkant van de reactor gevoed. Gas wordt geïnjecteerd door een klein gaatje in de bodemstator, vlakbij de rand van de schijf. Gasbelletjes worden afgesnoerd van de gasinlaat, door de hoge snelheidsgradiënt tussen de rotor en de stator. De gasbellen worden dus kleiner bij hogere rotatiesnelheden. De gasbellen worden naar binnen geduwd vanwege de centrifugaalkracht. De gasfractie in de reactor is slechts een paar volumeprocent. Hoe kleiner de gasbellen zijn, hoe lager de snelheid naar het midden toe is. De verblijftijd van de gasbellen, en dus de gasfractie in de reactor, gaat omhoog als de rotatiesnelheid toeneemt. Gecombineerd met de kleiner wordende gasbellen geeft dit dus een verhoging van het gas-vloeistofoppervlak ($a_{GL}$) bij een stijgende rotatiesnelheid.
Bij lage rotatiesnelheden worden gasbellen gevormd die groter zijn dan de afstand tussen de rotor en de stator. Het meeste gas-vloeistofoppervlak, en dus de meeste stofoverdracht, vindt dan plaats tussen de gasbel en de vloeistoffilms op de rotor en de stator. Bij hogere rotatiesnelheden zijn de gasbellen kleiner dan de afstand tussen de rotor en de stator. De stofoverdrachtscœfficiënt wordt dan bepaald door de grootte en de snelheid van de turbulente wervelingen in de vloeistof, en is daarom een functie van de energiedissipatie in de reactor.

De volumetrische gas-vloeistofstofoverdrachtscoëfficiënt \( k_{GLaGL} \) is het product van de stofoverdrachtscœfficiënt en het gas-vloeistofoppervlak, en stijgt daarom ook met toenemende rotatiesnelheid. Ook een groter gasdebiet, een grotere straal van de rotor en een kleinere afstand tussen de rotor en de stator hebben een positieve invloed op de stofoverdracht. De hoogste waarde van \( k_{GLaGL} \), gemeten met de desorptie van zuurstof uit water, is 2.5 m\(^3\)L m\(^{-3}\)s\(^{-1}\), bij een gasdebiet van 1.5 \( \cdot \) 10\(^{-5}\) m\(^3\) s\(^{-1}\) en een rotatiesnelheid van 209 rad s\(^{-1}\), een rotorstraal van 0.135 m en een rotor-statorafstand van 1 mm. Dit is een ordegrootte groter dan in conventionele reactoren, zoals geroerde-tankreactoren of bellenkolommen.

De energiedissipatie in de spinning disc reactor is drie ordes groter dan in conventionele reactoren. De verbetering in stofoverdracht is echter maar een factor twintig. De gas-vloeistofstofoverdracht per eenheid energiedissipatie \( \left( \frac{k_{GLaGL}}{E_d} \right) \) is daarom maar 1.1 m\(^3\)L MJ\(^{-1}\), terwijl het in geroerde tanks 80 m\(^3\)L MJ\(^{-1}\) is. Het vergroten van de straal van de rotor van 0.066 m naar 0.135 m levert een verdrievoudiging van de stofoverdracht op, terwijl de benodigde energie met een factor vijftien stijgt. Vanuit het oogpunt van energieconsumptie wordt de reactor dus bij voorkeur opgeschaald door meerdere reactoreenheden achter elkaar te zetten, in plaats van op te schalen door een grotere rotor te gebruiken. Het opschalen van de reactorconfiguratie met de gasinlaat in de bodemstator door meerdere eenheden achter elkaar te plaatsen kan in de praktijk echter lastig blijken, omdat het gas geherdistribueerd moet worden na elke rotor-statoreenheid.

Een alternatief voor de voorgaande reactorconfiguratie is het gezamenlijk toevoeren van gas en vloeistof aan de bovenkant van de reactor. De vloeistof stroomt in dit geval in een dunne film over de rotor. Het gebied boven de rotor wordt gevuld door de gasfase. Kleine gasbellen worden afgesnoerd aan de rand van dit gebied. De rest van de reactor is dan gevuld met vloeistof met kleine gasbellen daarin gedispergeerd, het grootste gedeelte van de stofoverdracht vindt hier plaats. De totale stofoverdracht gecorrigeerd voor de drijvende kracht in concentratie \( k_{GLaGLVR} \) is tweemaal zo groot in de configuratie waar gas en vloeistof gezamenlijk worden toegevoerd dan wanneer het gas toegevoerd wordt via een inlaat in de bodemstator.
Het opschalen van de reactorconfiguratie waar gas en vloeistof gezamenlijk worden toegevoerd is relatief eenvoudig. Meerdere rotor-statoreenheden kunnen achter elkaar geplaatst worden, bij voorkeur met de rotors op een gezamenlijke as. De stofoverdracht per rotor-statoreenheid is hetzelfde als in het geval van een enkele rotor-statoreenheid. Het is daarom waarschijnlijk dat het stromingsgedrag hetzelfde is, dus met een vloeistoffilm op de rotor en de rest van de reactor gevuld met vloeistof met kleine bellen daarin. De drukval in de spinning disc reactor wordt voornamelijk veroorzaakt door de centrifugaaldruk, en neemt toe met een toenemend vloeistofdebiet. De drukval in het geval van gas-vloeistofstroming is 2.5 maal zo hoog als in het geval met alleen vloeistof, tot een maximum van 0.64 bar drukval per rotor-statoreenheid, bij een rotatiesnelheid van 459 rad s⁻¹.

Een heterogeen gekatalyseerde vloeistoffasereactie, de oxidatie van glucose, is uitgevoerd in de spinning disc reactor, waarbij de katalysator is aangebracht op één zijde van de rotor. De reactiesnelheid is afhankelijk van zowel de kinetiek van de reactie als van de stofoverdracht van de vloeistof naar de vaste katalysator. De vloeistof-vaststofoverdrachtscoëfficiënt neemt toe met toenemende rotatiesnelheid, waarbij het stromingprofiel van laminair naar turbulent overgaat. De hoogste gemeten stofoverdrachtscoëfficiënt \( k_{LS} \) is \( 8 \cdot 10^{-4} \text{ m}^3\text{L m}^{-2} \text{s}^{-1} \), bij een rotatiesnelheid van 157 rad s⁻¹. Dit is een orde groter dan in conventionele reactoren, zoals een gepakt bed reactor. Een vloeistof-vastoppervlak van 2000 m² m⁻³ kan bereikt worden, wat vergelijkbaar is met conventionele reactoren. De volumetrische vloeistof-vaststofoverdrachtscoëfficiënt \( k_{LSaLS} \) is dus ook een orde groter.

De hoge gas-vloeistof- en vloeistof-vaststofoverdrachtssnelheden maken de rotor-stator spinning disc reactor een veelbelovende meerfasereactor. De energiedissipatie is relatief hoog, maar dat levert ook stofoverdrachtscoëfficiënten op die een ordegrootte groter zijn dan in conventionele reactoren. De rotor-stator spinning disc reactor is daarom vooral geschikt voor processen waarbij conversie en selectiviteit belangrijker zijn dan de energieconsumptie. De spinning disc reactor is ook zeer geschikt voor reacties met gevaarlijke stoffen en/of hoge drukken, vanwege het kleinere reactorvolume. Daarnaast zijn hoge warmteoverdrachtscoëfficiënten te verwachten, wat vooral van pas komt bij (zeer) exotherme reacties. De hoge stofoverdrachtscoëfficiënten kunnen ook een significant effect hebben op de selectiviteit, wat mogelijkheden opent voor processen die voorheen niet mogelijk of economisch rendabel waren.
1.1 Process intensification

The chemical industry is continuously striving towards smaller and more efficient equipment and safer production. This is driven by economical motivations, but also by consciousness about the responsibility for the environment and by governmental regulations. Process intensification is the trend in which new equipment and processing methods are developed, which are substantially smaller, more energy efficient and/or produce less waste (Stankiewicz and Moulijn, 2000). This is done by improving or developing equipment, such as reactors, and heat transfer and separation equipment. Otherwise, improved process methods are developed, e.g. the integration of multiple process steps, such as reaction and separation, into one device. The key towards process intensification is often the improvement of mass transfer and heat transfer steps, which can both lead to higher production rates, but also to more control over the conditions in the process. This can open opportunities to perform processes under conditions where higher conversions or selectivities are reached. This thesis describes the rotor-stator spinning disc(s) reactor, a new reactor for multiphase reactions, and an example of an intensified reactor.

1.2 Multiphase reactors

The overall rate of multiphase reactions, such as heterogeneously catalyzed reactions, is often limited by the mass transfer from the gas phase to the liquid phase and/or from the liquid phase towards the solid catalyst. Increasing the gas-liquid and liquid-solid mass transfer rates can therefore lead to higher productivity or lower reactor volumes. Examples of mass transfer limited reactions performed in industry are oxidations and hydrogenations, which are conven-
tionally performed in bubble columns, stirred tank reactors, and packed bed reactors.

The overall reaction rate of a multiphase reaction, can be written as a sum of resistances in series, towards mass transfer and reaction. The overall reaction rate of a first order heterogeneously catalyzed reaction, for example, can be written as:

\[-r_A = \left( \frac{RT}{Hk_Ga_{GL}} + \frac{1}{k_{GL}a_{GL}} + \frac{1}{k_{LS}a_{LS}} + \frac{1}{\eta k_rC_{cat}} \right)^{-1}RTCG \] (1.1)

The first three terms describe the mass transfer resistances. These are, in consecutive order, the mass transfer from the bulk of the gas phase to the gas-liquid interface, from the gas-liquid interface to the bulk of the liquid, and from the bulk of the liquid towards the solid interface. The mass transfer from the bulk of the gas phase towards the gas-liquid interface is rarely limiting the overall reaction rate, due to the diffusion coefficient in the gas phase which is three to four orders of magnitude higher than in the liquid phase. This mass transfer resistance is often incorporated in the mass transfer from the gas-liquid interface, since it is difficult to determine separately and seldom plays an important role. The mass transfer from the gas-liquid interface to the bulk of the liquid is often rate limiting; both in homogeneously and heterogeneously catalyzed reactions it plays an important role. The mass transfer from the bulk of the liquid to the solid interface, is often rate limiting in heterogeneously catalyzed reactions. The last term in Equation 1.1 describes the reaction at the solid catalyst; the decrease in reaction rate due to internal diffusion limitation in the porous catalyst support is incorporated by the effectiveness factor.

The rate of mass transfer between two phases is determined by the concentration difference (driving force), the mass transfer coefficient \((k_G, k_{GL} \text{ or } k_{LS})\) and the interfacial area between the phases \((a_{GL} \text{ or } a_{LS})\). The volumetric mass transfer coefficient, \(k_{GL}a_{GL} \text{ or } k_{LS}a_{LS}\), is the product of the mass transfer coefficient and the interfacial area. This lumped parameter is often used, since it is difficult, or sometimes even impossible, to measure the mass transfer coefficient and the interfacial area separately. The mass transfer coefficient is determined by the diffusion coefficient, and by the contact time of a fluid element near the surface. This contact time can be the time it takes for a gas bubble to pass a liquid element, or a liquid element to flow past a solid. However, it can also be determined by the rate of surface renewal, e.g. by the velocity and size of turbulent eddies. In conventional reactor systems, the contact time is determined by gravity, or by the power input in the system, e.g. by stirring, or pushing the fluid through the system.

The liquid-solid interfacial area is in most reactors determined by the size of the solid
Spinning disc reactors

(catalyst) particle and the fraction of reactor volume occupied by these. The use of smaller particles increases the interfacial area, and thus the mass transfer rate, but it can also increase the pressure drop in the system, e.g. in the case of a packed bed reactor. The gas-liquid interfacial area is directly determined by the geometry of the reactor, e.g. in falling film reactors or trickle bed reactors, where the gas-liquid interfacial area is of the same order of magnitude as the interfacial area of the solids. However, in the case where the gas phase is present as gas bubbles, e.g. in bubble columns or stirred tank reactors, the interfacial area is determined by the size of the gas bubbles and the gas holdup in the reactor. The size of the gas bubbles depends on the hydrodynamics in the reactor, e.g. by the shear force at the gas injection point.

The use of higher velocities and accelerations leads to higher surface renewal rates, and thus to a higher mass transfer coefficient, and to higher shear forces, which can lead to a higher gas-liquid interfacial area. Rotating packed beds (Ramshaw, 1993) and spinning disc reactors (Aoune and Ramshaw, 1999, White and Litt, 1975) are examples of multiphase reactors where centrifugal forces are used to increase the rate of mass transfer.

1.3 Spinning disc reactors

Two types of reactors which use a rotating disc are present in literature, and have been researched already since the 1960's. The simplest configuration uses a rotating disc in a vessel filled with liquid. The rotation of the disc induces a flow towards the disc (Schlichting, 1955), thereby increasing the liquid-solid mass transfer coefficient. The liquid-solid mass transfer coefficient is constant over the whole disc in the laminar flow regime, due to the nature of the induced flow (Levich, 1962). With increasing rotational disc speed the laminar flow becomes unstable and, after a transition regime, the flow is turbulent (Mohr and Newman, 1976). The turbulent flow has increased mass transfer coefficients compared to the laminar flow case (Daguenet, 1968, Ellison and Cornet, 1971). The "free rotating disc" is used extensively for electrochemical processes (Selman and Tobias, 1978), but is also used for heterogeneously catalyzed reactions (e.g. hydrogenations), where the catalyst is present on the rotating disc (White and Litt, 1975).

Another type of spinning disc reactor is mainly used for gas-liquid systems. The liquid is fed near the middle of the rotating disc, and flows as a thin film over the disc, as shown in Figure 1.2a. This type of spinning disc reactor has been investigated since the 1960’s, e.g. for heat transfer from the rotor to the liquid film (Espig and Hoyle, 1965, Jachuck and Ramshaw, 1994). In the succeeding decades the interest was focused on the mass transfer from the gas
phase to the liquid film (Brauner and Maron, 1982, Rahman and Faghri, 1993, Ramshaw, 1993, Sisoev et al., 2005, Wood and Watts, 1973). The liquid-solid mass transfer, thus from the liquid film towards the rotor, was investigated in this configuration as well (Burns and Jachuck, 2005, Peev et al., 2007a, Rahman and Faghri, 1993).

A large part of the research focused on the hydrodynamics of the liquid film, i.e. the liquid film thickness, the radial velocity and the waviness of the film. The average liquid film thickness decreases with increasing disc radius and rotational disc speed, and increases with increasing liquid flow rate and increasing viscosity (Burns et al., 2003, Wood and Watts, 1973). The radial velocity increases with increasing rotational disc speed and increasing liquid flow rate, and decreases with increasing viscosity (Wood and Watts, 1973). At low flow rates, the liquid flows in rivulets over the surface (Woods, 1995); at increased flow rates and rotational disc speeds, a smooth liquid film is obtained. At higher flow rates and rotational disc speeds, surface waves appear on the liquid film (Aoune and Ramshaw, 1999, Brauner and Maron, 1982, Sisoev et al., 2005), see also Figure 1.2d; the growth of the disturbances is lower at a higher liquid viscosity (Charwat et al., 1972, Woods, 1995). Different types of waves, e.g. concentric waves or spiral waves, are present, depending on the conditions used, as described in detail by Charwat et al. (1972). The surface waves are known to increase the mass transfer rate (Aoune and Ramshaw, 1999, Peev et al., 2007b, Sisoev et al., 2005); rotating discs with surface modifications are designed, to increase the waviness and thus the mass transfer (Jachuck and Ramshaw, 1994, Matar et al., 2008).

The liquid film spinning disc has been used for various applications; mainly associated with the high rates of mass transfer, heat transfer and the plug flow behaviour of the liquid. Examples of applications are the manufacturing of pharmaceuticals (Oxley et al., 2000), polymerization reactions (Boodhoo et al., 1997) and crystallizations (Brechtelsbauer et al., 2001).

The liquid film spinning disc reactor has high mass and heat transfer rates, which makes it a promising concept, but there are also some major drawbacks. Scale-up can be done by increasing the rotor size, which can only be done up to a certain size, or by collecting the liquid and feeding it to the next stage. The apparatus which results from this will be relatively large, while only the top surface of the rotor will be used, which is thus directly the gas-liquid interfacial area. The rotor-stator spinning disc reactor, as described in this thesis, uses both sides of the disc, in a rotor-stator housing. This is thus an expansion of the conventional concept, which has several advantages, as described in this thesis.
1.4 Rotor-stator spinning disc reactor

Figure 1.1: Schematic representation of the concept of the rotor-stator spinning disc reactor with a gas inlet in the bottom stator. The reactor consists of a rotating disc in a cylindrical housing. The distance between the rotor and the stator is small, commonly 1 mm. Liquid is fed to the reactor from the top inlet, near the rotating axis. Gas is injected through an orifice in the bottom stator; small gas bubbles are sheared off, due to the high velocity gradient in the gap between the rotor and the stator. The surface renewal rate near the gas bubbles is increased due to the turbulence induced by the rotation of the disc, which leads to a high gas-liquid mass transfer coefficient; the same mechanism increases the mass transfer from the bulk of the liquid to the rotor and the stator.

This thesis describes the hydrodynamics and mass transfer in a novel type of multiphase reactor, the rotor-stator spinning disc reactor. It consists of a rotating disc in a cylindrical housing, as shown schematically in Figure 1.1. The distance between the rotor and the reactor wall, the stator, is small, in the order of 1 mm. The liquid, which is the continuous phase, is injected to the reactor from the top. The gas phase is injected through a small gas inlet in the bottom stator, near the rim of the disc. Gas bubbles are sheared off at the gas inlet, due to the high velocity gradient, and thus shear force, between the rotor and the stator. The gas bubble size decreases with increasing rotational disc speed, leading to an increased gas-liquid interfacial area \( a_{GL} \), and thus in an increase in gas-liquid mass transfer. The gas bubbles will move radially inward, due to the centrifugal force. The flow in the rotor-stator spinning disc
reactor is, under most conditions, turbulent. The small size and high velocity of the turbulent eddies increase the surface renewal rate, and therefore the mass transfer coefficient \( k_{GL} \), thus leading to a high volumetric mass transfer coefficient, \( k_{GLaGL} \). Chapter 2 describes gas holdup, gas-liquid interfacial area and gas-liquid mass transfer in the rotor-stator spinning disc reactor with a single gas inlet in the bottom stator.

The concept of a spinning disc reactor with a liquid film on the rotor, and the rotor-stator spinning disc reactor can be combined into an alternative configuration of the spinning disc reactor, which utilizes the whole reactor volume, as shown in Figure 1.2c. Gas and liquid are fed to the reactor through the top stator, near the rotating axis. The liquid will flow in a thin film over the rotor surface (Figure 1.2d), the gas phase is present between the liquid film and the top stator. Near the rim of the rotor small gas bubbles are sheared off; the region surrounding the rim of the rotor and the region between the rotor and the bottom stator are filled with small gas bubbles dispersed in liquid (Figure 1.2e). This reactor concept has advantages in scale up of the reactor, since the outlet stream can be fed directly to the next rotor-stator unit, thereby creating the possibility of the use of a multiple rotating disc reactor, where all the rotors are mounted on a common axis. Chapter 3 describes the gas-liquid mass transfer in the co-fed rotor-stator spinning disc reactor in detail, and it is compared with the mass transfer in the configuration with a single gas inlet.

The gas bubble size in the spinning disc reactor, and thus the gas-liquid interfacial area, depends on the shear force at bubble formation. This shear force depends on the rotational disc speed, but also on the size of the rotor and the rotor-stator distance. The gas-liquid mass transfer coefficient depends either on the relative velocity of the gas bubbles and the liquid, or on the size and velocity of the turbulent eddies. The former depends mainly on the tangential velocity, and thus the size, of the rotor; the latter depends on the energy dissipation rate, which is a strong function of rotor size and rotor-stator distance. Chapter 4 deals with the influence of the rotor radius and the rotor-stator distance on the volumetric gas-liquid mass transfer coefficient in the rotor-stator spinning disc reactor with a single gas inlet in the bottom stator. Additionally, the influence of the rotational disc speed and liquid flow rate on the volumetric gas-liquid mass transfer coefficient in the spinning disc reactor with a 0.135 m radius rotor is discussed.

The rate of a gas-liquid solid reaction, e.g. a heterogeneously catalyzed reaction, depends on both the gas-liquid mass transfer as well as the liquid-solid mass transfer. The oxidation of glucose on a Platinum catalyst is performed in the rotor-stator spinning disc reactor with liquid phase only. The rate of the reaction depends on the kinetics, as well as the mass transfer
Figure 1.2: (a) "Conventional" spinning disc reactor with liquid film on the rotor. (b) Rotor-stator spinning disc reactor with a single gas inlet in the bottom stator (Meeuwse et al., 2010). (c) Schematical drawing of the gas and liquid flow in the co-fed spinning disc reactor (this work). (d) Film flow region: Liquid film on the rotor with gas bubble formation at the rim of the rotor. (e) Dispersed flow region: Small gas bubbles surrounding the rim of the rotor and between the rotor and the bottom stator.
from the bulk of the liquid to the catalyst on the rotor. The kinetics and mass transfer are de-
coupled, using experiments at various temperatures and rotational disc speeds. The results of
this, including the liquid-solid mass transfer coefficient as function of rotational disc speed, is presented in Chapter 5.

The scale up of the spinning disc reactor can either be done by increasing the rotor radius,
or by putting multiple rotor-stator units in series. Chapter 6 describes the gas-liquid mass
transfer, energy dissipation and pressure drop in a 2-stage and a 3-stage rotor-stator spin-
n ing disc reactor. Additionally, the advantages, disadvantages and potential applications of
the rotor-stator spinning disc reactor are discussed, and compared with conventional reactor
equipment.

Chapter 7 describes the most important research results and conclusions presented in this
thesis.

**Bibliography**

of liquid films on rotating discs. International Journal of Heat and Mass Transfer 42 (14),
2543-2556.

Boodhoo, K. V. K., Jachuck, R. J. J., Ramshaw, C., 1997. Spinning disk reactor for the intensi-
fication of styrene polymerization. BHR Group Conference Series Publication 28 (Process
Intensification in Practice), 125–133.


spinning disc reactor for continuous processing. Organic Process Research & Development
5 (1), 65–68.

for a spinning disc reactor using a limiting current technique. International Journal of Heat
and Mass Transfer 48 (12), 2540–2547.

and the determination of spin-up radius on a rotating disc using an electrical resistance
 technique. Chemical Engineering Science 58 (11), 2245–2253.


Daguenet, M., 1968. Etude du transport de matiere en solution, a l’aide des electrodes a
1596.

ical Society 118 (1), 68–72.


Gas-liquid mass transfer in a rotor-stator spinning disc reactor

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Abstract

This chapter describes a new multiphase reactor, the rotor-stator spinning disc reactor, which shows high rates of gas-liquid mass transfer in comparison to conventional multiphase reactors. The volumetric gas-liquid mass transfer coefficient $k_{GLaGL}$ in the rotor-stator spinning disc reactor increases with increasing rotational disc speed, due to the higher surface renewal rate caused by the increasing turbulence, and with increasing gas flow rate. Measured $k_{GLaGL}$ values are as high as $0.43 \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$ at $7.3 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$ gas flow and a rotational disc speed of 179 rad s$^{-1}$, and are expected to increase even further at increasing rotational disc speed. This is twice as high as for conventional reactors as bubble columns, in spite of the low gas holdup of $0.021 \text{ m}^3 \text{ m}^{-3}$ with only one gas inlet. The volumetric mass transfer per unit volume of gas, $k_{GLaGL}/\varepsilon_G$, of $20.5 \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$ is 40 times higher than $0.5 \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$ for a bubble column.
2.1 Background and motivation of work

This chapter describes a new multiphase reactor, the rotor-stator spinning disc reactor, which shows high rates of gas-liquid mass transfer in comparison to conventional multiphase reactors. The spinning disc reactor consists of a rotating disc, the rotor, and a static disc, the stator. A large velocity gradient, and thus a large shear force, is created in the fluid in the gap between the discs, caused by the rotation of the rotor. Gas is supplied to the reactor through one or more gas inlets in the stator. The shear force tears the gas bubble away from the inlet. The size of the gas bubble is therefore much smaller than in a stagnant liquid, resulting in a high gas-liquid interfacial area, \( a_{GL} \). The high degree of turbulence, which is caused by the high liquid velocities in the small gap between the rotor and the stator, very rapidly renews the gas-liquid surface. This results in a high gas-liquid mass transfer coefficient, \( k_{GL} \). This leads to the high values of the volumetric gas-liquid mass transfer coefficient, \( k_{GL}a_{GL} \), shown in this paper. The maximum value presented in this paper is \( 0.43 \, \text{m}^3 \, \text{L}^{-1} \, \text{m}^{-3} \, \text{R}^{-1} \) at \( 7.3 \cdot 10^{-6} \, \text{m}^3 \, \text{s}^{-1} \) gas flow and a rotational disc speed of 179 rad s\(^{-1}\).

The overall rate of gas-liquid reactions with a solid catalyst is often limited by the mass transfer of the gaseous component to the catalytic site. Increasing the gas-liquid and liquid-solid mass transfer can therefore increase the reaction rate. Conventional gas-liquid-solid reactors that are used for reactions such as hydrogenations and oxidations are stirred slurry batch reactors, slurry bubble columns, and trickle bed reactors.

The rate of gas-liquid mass transfer is determined by the mass transfer coefficient, the gas-liquid interfacial area, and the concentration gradient. The mass transfer coefficient in the liquid phase is dependent on the rate at which fresh liquid packages reach the gas-liquid interface, the surface renewal rate. The size of the gas bubbles, and thus the gas-liquid interfacial area, is dependent on the shear stress on the bubbles, either at the moment they are formed or when they break up due to these shear stresses. In conventional reactors the shear stress and surface renewal rate are determined by gravitational forces and by the power input, e.g. by stirring or pushing the fluids through the reactor at high pressure. Use of higher accelerations can significantly increase the shear stress and the surface renewal rate, thereby improving heat and mass transfer. Rotating equipment, with centrifugal accelerations, can be used for this purpose. The use of rotating packed beds (Ramshaw, 1993) and rotating discs (Burns et al., 2003, White and Litt, 1975) are examples of these processes. This paper focuses on the use of a rotating disc to increase gas-liquid mass transfer.

Two types of reactors using a rotating disc can be found in literature. A rotating disc in a
tank, with a large gap between the disc surface and the reactor bottom, has been researched extensively in the 1960’s and 1970’s. For example liquid-solid mass transfer limited hydrogenation reactions are performed at rotational disc speeds up to 42 rad s$^{-1}$ of the catalytically active disc (White and Litt, 1975). If liquid is fed on top of a rotating disc, the liquid will flow outwards as a thin film with a high gas-liquid mass transfer and liquid-solid mass transfer (Burns et al., 2003).

In the rotor-stator spinning disc reactor, as used in this study, a gas-liquid dispersion with small bubbles is created, leading to a high gas-liquid interfacial area. The high degree of turbulence between the rotor and the stator results in high gas-liquid and liquid-solid mass transfer coefficients. This reactor can improve the efficiency of mass transfer limited multiphase reactions. Scale-up can be done by stacking multiple rotor-stator cells on a common axis.

This paper presents the gas holdup, the gas-liquid interfacial area, and the gas-liquid mass transfer coefficient in a rotor-stator spinning disc reactor, as a function of the rotational disc speed of the rotor and the gas flow rate. The rotor-stator spinning disc reactor has a reactor volume of $50 \times 10^{-6}$ m$^3$ and a maximum rotational disc speed of 179 rad s$^{-1}$. The gas holdup and gas-liquid interfacial area are determined from photographs made of the gas-liquid flow between the rotor and the stator. The rate of gas-liquid mass transfer is determined by the rate of desorption of oxygen from water.

2.2 Experimental

2.2.1 Experimental setup

The spinning disc reactor consists of a cylindrical housing (0.152 m inner diameter, $5.0 \times 10^{-3}$ m height), with a rotating disc in the middle between the top and bottom plate of the cylinder (see Figure 2.1). The distance between the rotating disc (rotor) and the bottom wall of the reactor (stator), the disc spacing, is $1.0 \times 10^{-3}$ m. The total reactor volume (including the space above the rotor) is $50 \times 10^{-6}$ m$^3$. The maximum rotational disc speed of the rotor is 179 rad s$^{-1}$ (1710 rotations per minute). The stainless steel disc, with radius of 0.066 m and thickness of $3 \times 10^{-3}$ m is sandblasted to prevent a sharp reflection of light, which would influence the image analysis. The housing of the reactor is made of transparent PMMA for visualisation purposes.
Liquid enters the reactor through the top wall, near the rotor axis. It flows outwards, to the rim of the reactor, and flows under the disc to the center of the reactor. Gas enters the reactor through an orifice in the stator, near the rim of the disc.

The spinning disc reactor is operated at room temperature.

**2.2.2 Image analysis**

A Canon EOS digital 400D camera is used to analyse the size of the gas bubbles. The image is collected via a mirror under the reactor, which is adjusted at an angle of 45°, to be able to record the bottom of the reactor with the camera placed horizontally and to prevent liquid spilling on the camera. One pixel represents a distance of $6.5 \times 10^{-5}$ m. The shutter time of the camera is set at 0.1 s. A Philips PR 9113 stroboscope is used to produce a flash, and thus an exposure time of the image, of less than $1 \times 10^{-5}$ s, in which the bubbles move less than 1 pixel, to get a sharp image.

The photographs are converted to a black and white image where only the bubbles are visible. This is done using a Matlab script. The script determines the position and size of the rotating disc in the image based on the large difference in reflection of the disc and the
Figure 2.2: Images of the gas-liquid flow between the rotor and the stator at 179 rad s$^{-1}$, 7.3 $10^{-6}$ m$^3$ s$^{-1}$ gas flow, with the gas inlet in the upper part of the picture. The original photograph (a) is converted to a black and white image (b) by a Matlab script, to determine the gas holdup and the gas-liquid interfacial area.

surroundings. The outer 4 $\cdot$ 10$^{-3}$ m radius of the disc and the inner 1.9 $\cdot$ 10$^{-2}$ m are excluded from the analysis, because the gas inlets and the outlet interfere with the recognition of the bubbles.

The contrast between the bubbles and the disc is improved by several functions of Matlab. Pixels above a certain threshold value are converted into white, pixels below this threshold to black, resulting in a black and white image, as is shown in Figure 2.2b. This threshold value is determined using Otsu’s method (Otsu, 1979). A group of white pixels adjacent to each other is defined as one bubble. The area of the pixels in this group is determined, and from this the equivalent bubble diameter is calculated, which is the diameter of a circle with the same surface area. If a pixel group has an opening in the middle, but is a closed contour, the gap is filled up. The position of the bubble is taken as the center of mass of the pixel group.

If the equivalent bubble diameter is smaller than the disc spacing, the volume and interfacial area of the bubble are calculated assuming a spherical bubble shape. If the equivalent bubble diameter is larger than the disc spacing, the bubble is assumed to have a shape which is the combination of the outer half ring of a torus, and a cylinder in the middle. A side view of this, with the appropriate equations for area and volume, is shown in Figure 2.3.
A group of less than 7 pixels, which corresponds to an equivalent bubble diameter smaller than $1.8 \cdot 10^{-4}$ m, is not taken into account as a bubble as this is found to be noise from the image analysis.

A list is created with all the bubbles present, with their positions, sizes and surface areas. This is used to determine the total gas holdup and the total gas-liquid interfacial area in the reactor. The gas holdup and the gas-liquid interfacial area determined by the image analysis are based on 20 images per measurement. The spread of the data is indicated in the graphs of the gas holdup and gas-liquid interfacial area in the next sections with error bars, which represent the standard deviation of the data points.

The conversion of the original photograph to a black and white image is based on the contrast between the bubbles and the background of the picture (the rotor). The stroboscope which is used enlightens the bubbles from one side, as can be seen in Figure 2.4, therefore one side of a bubble has a higher light intensity. This part will not be detected as part of the bubble. However, on the other side of a bubble a shadow of that bubble is present, which is detected as part of the bubble. Visual inspection of a number of bubbles indicates that these effects approximately counterbalance each other.

Bubbles that are too close to each other are detected as one pixel group, thus as one bub-
Figure 2.4: Detail of Figure 2.2, image at 179 rad s\(^{-1}\), 7.3 \(\cdot\) 10\(^{-6}\) m\(^3\) s\(^{-1}\) gas flow. The light intensity is higher on the bottom left of each bubble. A shadow from the bubble is visible at the top right. In the image analysis, which is based on contrast, these effects approximately counterbalance each other.

This will have only a small effect on the holdup, but the gas-liquid interfacial area will be lower than the actual value.

Figure 2.5: Cut of 0.012 m x 0.012 m of a photograph (left) and converted image (middle) at 56 rad s\(^{-1}\) and 7.3 10\(^{-6}\) m\(^3\) s\(^{-1}\) gas flow. In the top bubble the contour is closed, in the bottom bubble the contour is not closed, thus the gap in the middle will not be filled up (right). The gas holdup and the gas-liquid interfacial area are therefore underestimated in case of the bottom bubble.

The image analysis is based on the contrast between the edge of a bubble, and the liquid outside the bubble or the gas inside a bubble. The middle of a bubble with an equivalent bub-
ble diameter larger than $3 \cdot 10^{-3}$ m is therefore not detected (see Figure 2.5). However, if the contour of the bubble converted into black and white is closed, the gap will be filled in. If this contour is not closed only the edge of the bubble is detected as being a bubble, which means that the gas holdup and the gas-liquid interfacial area are underestimated. At 56 rad s$^{-1}$ and $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$ gas flow rate, the gas holdup determined by the image analysis is estimated to be approximately 50% of the actual value, while the value at 84 rad s$^{-1}$ is approximately 80%. The effect is negligible at rotational disc speeds higher than 100 rad s$^{-1}$. At $1.8 \cdot 10^{-6}$ m$^3$ s$^{-1}$ gas flow rate and 56 rad s$^{-1}$ rotational disc speed, the gas holdup is approximately 90% of the actual holdup, at higher rotational disc speeds the effect is again negligible.

### 2.2.3 Mass transfer measurement

The gas-liquid mass transfer is measured by desorption of oxygen from oxygen saturated water. The pressure in a single phase liquid would be around $1.5 \cdot 10^4$ Pa higher at the rim of the disc than in the center at the highest rotational velocity (Daily and Nece, 1960, Poncet et al., 2005a). Therefore desorption measurements are carried out, because these are less influenced by the pressure, and because the residence time distribution of the gas bubbles does not play a significant role with desorption experiments. The oxygen concentrations in the liquid in the inlet and in the outlet of the reactor are measured with fiber optic oxygen sensors (AVS-OXYPROBE, Avantes). All the measurements are performed at steady state. The liquid from the outlet is fed into an aerated $2.0 \cdot 10^{-3}$ m$^3$ vessel where air is flown through to increase the oxygen concentration. The liquid taken from this vessel is fed to the reactor. The oxygen concentration in the entering liquid is normally around 85% of saturation. From the difference between the oxygen concentrations in the inlet and in the outlet, the $k_{GLaGL}$ value is calculated with:

$$k_{GLaGL} = \frac{\phi_G}{V_R RT} \ln \left[ \frac{RT_C in - C out}{RT_C out - C out} \right]$$  \hspace{1cm} (2.3)

The oxygen concentration in the gas feed, $C_{in}^G$, is zero. The gas concentration in the outlet is calculated from the mole balance:

$$C_{out}^G = C_{in}^G + \frac{\phi_L}{\phi_G} \left( C_{in}^L - C_{out}^L \right)$$  \hspace{1cm} (2.4)

Equation 2.3 is based on a gas phase which behaves as plug flow and a liquid phase which is assumed to be ideally mixed. The plug flow of gas bubbles follows from visual observation that indicate that most gas bubbles have approximately the same residence time, therefore the
gas phase is considered to have plug flow behaviour. In this way the experimental $k_{GL}a_{GL}$ value is always underestimated.

The ideal mixing of the liquid follows from the complex flow patterns in this system. At the rotor of a rotor-stator system an outward radial velocity develops and an inward radial velocity develops at the stator (Daily and Nece, 1960, Dijkstra and van Heijst, 1983, Poncet et al., 2005b). The volumetric flow rates of these flows in the experimental conditions of the rotor-stator system used in this paper are up to a factor 50 higher than the net flow rate of liquid (Owen and Rogers, 1989). A large recycle is thus present in the reactor; the liquid phase can therefore be regarded as ideally mixed. This was confirmed by an ink tracer experiment. An ink tracer was injected from the gas inlet into the reactor; the flow path of this ink was recorded with a high speed camera. In less than a half rotation the ink was mixed with the rest of the liquid in the reactor, which corresponds to approximately 0.08 s, at a rotational disc speed of 100 rad s$^{-1}$. This time scale is much smaller than the average residence time of the liquid, which was 8 s.

### 2.3 Results and discussion

#### 2.3.1 Gas holdup

![Gas holdup as a function of rotational disc speed](image)

Figure 2.6: Gas holdup as a function of the rotational disc speed. The gas holdup increases with increasing gas flow rate and increasing rotational disc speed.

The gas holdup is plotted in Figure 2.6 as a function of the rotational disc speed. The gas holdup at 56 rad s$^{-1}$ and $7.3 \times 10^{-6}$ m$^3$ s$^{-1}$ gas flow rate is underestimated, approximately
by a factor 2, as described in Section 2.2.2. The gas holdup thus decreases with increasing rotational disc speed at disc speeds below 100 rad s\(^{-1}\) at 7.3 \(\cdot\) 10\(^{-6}\) m\(^3\) s\(^{-1}\) gas flow rate. An increase in gas holdup is observed at rotational disc speeds larger than 100 rad s\(^{-1}\). A bubble size distribution is determined to be able to explain this increase.

The gas holdup is shown in Figure 2.7 for various bubbles sizes at 7.3 \(\cdot\) 10\(^{-6}\) m\(^3\) s\(^{-1}\) gas flow rate. Most gas bubbles have an equivalent bubble diameter larger than 2 \(\cdot\) 10\(^{-3}\) m at a rotational disc speed below 100 rad s\(^{-1}\). The gas holdup of these large bubbles at rotational disc speeds below 100 rad s\(^{-1}\) is underestimated, the actual holdup is higher than determined by the image analysis, as described in Section 2.2.2. The holdup of gas bubbles larger than 5 \(\cdot\) 10\(^{-3}\) m decreases with increasing rotational disc speed, while the holdup of bubbles smaller than 2 \(\cdot\) 10\(^{-3}\) m increases.

The gas bubbles grow at the gas inlet, until the shear force is large enough to pull the bubble away from the gas inlet. The shear force is larger at a higher rotational disc speed, therefore the bubbles are sheared off faster. The number of small bubbles thus increases with increasing rotational disc speed. This is also observed for 1.8 \(\cdot\) 10\(^{-6}\) m\(^3\) s\(^{-1}\) gas flow rate, however with a lower number of large gas bubbles and a thus lower total gas holdup.

The highest gas holdup found is 0.021 m\(^3\) \(G\) m\(^{-3}\) \(R\), with one gas inlet as used in this study. This gas holdup is low in comparison to, for example, bubble columns, due to several reasons.
The gas can only be present in the bottom part of the reactor, under the rotating disc. If all this space would be occupied by gas, the gas holdup, which is defined per volume of reactor, would be $0.27 \, m^3/G \, m^{-3}_R$ at maximum. The centrifugal force, which is directed outwards, is present due to the rotation of the fluid. The liquid has a higher density than the gas, resulting in a net force on the gas bubbles directed inwards. The gas bubbles will therefore have a higher radial velocity, and thus a smaller residence time, than the liquid. This explains the low gas holdup compared to the ratio of the gas and liquid flow rates, which is near unity at $7.3 \cdot 10^{-6} \, m^3 \, s^{-1}$ gas flow rate.

The centrifugal force is proportional to the square of the rotational speed and is dependent on the bubble diameter to the power three. The gas bubbles have a higher radial velocity than the liquid, so a drag force is present on the bubble, which is directed outwards. This drag force is proportional to the square of the bubble diameter (Lohse and Prosperetti, 2003). The drag force is therefore of more importance for smaller bubbles than for larger bubbles. This means that the radial velocity of the smaller bubbles is smaller than that of the larger bubbles. A larger number of small bubbles is formed at increasing rotational speed, which means that the gas holdup can increase in spite of the increase in the centrifugal force.

### 2.3.2 Gas-liquid interfacial area

![Graph showing gas-liquid interfacial area as a function of the rotational disc speed. The gas-liquid interfacial area increases with increasing gas flow rate.](image)

Figure 2.8: Gas-liquid interfacial area as a function of the rotational disc speed. The gas-liquid interfacial increases with increasing rotational disc speeds above 100 rad s$^{-1}$, and increases with increasing gas flow rate.

The gas-liquid interfacial area, $a_{GL}$, increases with increasing gas flow rate (see Figure...
2.2 Gas-liquid mass transfer in a rotor-stator spinning disc reactor

2.8), because the gas holdup increases, despite the fact that the average bubble size increases. The gas holdup increases with increasing rotational disc speed, and the average bubble size decreases. The gas-liquid interfacial area will therefore increase even more with increasing rotational disc speed, because smaller bubbles have a higher interfacial area per unit volume. The maximum value of the gas-liquid interfacial area measured is 99 m$^2$ m$^{-3}$ at a gas flow rate of $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$ and a rotational disc speed of 179 rad s$^{-1}$.

2.3.3 Gas-liquid mass transfer coefficient

![Graph](image)

Figure 2.9: (a) Volumetric gas-liquid mass transfer coefficient as a function of the rotational disc speed. The gas-liquid mass transfer increases with increasing rotational disc speeds above 100 rad s$^{-1}$, and increases with increasing gas flow rate. (b) $k_{GLaGL}/\varepsilon_G$ as a function of the rotational disc speed. $k_{GLaGL}/\varepsilon_G$ increases slightly with rotational disc speeds above 100 rad s$^{-1}$.

The volumetric gas-liquid mass transfer coefficient, $k_{GLaGL}$ as a function of the rotational disc speed is shown in Figure 2.9a. Two mass transfer mechanisms can be distinguished:

- Mass transfer due to surface renewal of the liquid film between a large bubble and the rotor or the stator.
- Mass transfer due to turbulence.

At low rotational disc speeds large bubbles are present, and thus a relatively large area of the liquid film is obtained. A small decrease in mass transfer can be observed at rotational disc speeds lower than 80 rad s$^{-1}$ and at $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$ gas flow rate. At increasing rotational
disc speed the area of the liquid film decreases, because smaller bubbles are formed, resulting in a lower volumetric mass transfer coefficient. At increasing rotational disc speed the mass transfer due to turbulence increases rapidly. The minimum in the volumetric mass transfer coefficient in Figure 2.9a is caused by the combination of these two effects.

The gas-liquid mass transfer coefficient increases with increasing gas flow rate. The maximum value of the mass transfer coefficient measured is \(0.43 \text{ m}^3 \text{L}^{-3} \text{m}^{-3} \text{s}^{-1}\) at a gas flow rate of \(7.3 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}\) and a rotational disc speed of 179 rad s\(^{-1}\). This value is about two times higher than for conventional reactors as bubble columns (Van der Schaaf et al., 2007) and three times higher than for gas-inducing stirred reactors (Cents et al., 2005). The volumetric gas-liquid mass transfer coefficient is expected to increase even further at increasing rotational disc speeds larger than 179 rad s\(^{-1}\).

The value of \(k_{GLaGL}/\varepsilon_G\) as a function of the rotational disc speed is plotted in Figure 2.9b. This quantity represents the rate of volumetric mass transfer per unit volume of gas in the reactor. In bubble columns this quantity has a value of about \(0.5 \text{ m}^3 \text{L}^{-3} \text{m}^{-3} \text{s}^{-1}\) for air-water systems in the heterogeneous regime (Van der Schaaf et al., 2007). The spinning disc reactor shows values up to \(20.5 \text{ m}^3 \text{L}^{-3} \text{m}^{-3} \text{s}^{-1}\). An increase in the gas holdup can result in an even higher value of the volumetric gas-liquid mass transfer coefficient, \(k_{GLaGL}\). The gas holdup can be further increased by using more than one gas inlet in the stator at different tangential positions and by using higher gas flow rates.

### 2.4 Concluding remarks

The volumetric gas-liquid mass transfer coefficient \(k_{GLaGL}\) in the rotor-stator spinning disc reactor increases with increasing rotational disc speed and gas flow rate. Measured \(k_{GLaGL}\) values are as high as \(0.43 \text{ m}^3 \text{L}^{-3} \text{m}^{-3} \text{s}^{-1}\) at \(7.3 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}\) gas flow and a rotational disc speed of 179 rad s\(^{-1}\) and are expected to increase even further at increasing rotational disc speed. This is twice as high as for conventional reactors as bubble columns, despite the low gas holdup of \(0.021 \text{ m}^3 \text{G}^{-3} \text{m}^{-3}\). The volumetric mass transfer per unit volume of gas, \(k_{GLaGL}/\varepsilon_G\), is much higher than for conventional reactors, \(20.5 \text{ m}^3 \text{L}^{-3} \text{m}^{-3} \text{s}^{-1}\) instead of \(0.5 \text{ m}^3 \text{L}^{-3} \text{m}^{-3} \text{s}^{-1}\) for bubble columns. The volumetric mass transfer coefficient can be further increased by increasing the gas holdup by using more than one gas inlet, by adding gas to the top side of the reactor through gas inlets in the top stator, and by using larger gas flow rates.

The gas holdup in the spinning disc reactor increases with increasing rotational disc speed
and gas flow rate. The average diameter of the gas bubble decreases with increasing rotational disc speed. The gas-liquid interfacial area therefore increases with increasing rotational disc speed. The largest value of the gas-liquid interfacial area measured in this study is $99 \text{ m}^2 \text{ m}^{-3} R$ at a gas flow rate of $7.3 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$ and a rotational disc speed of 179 rad s$^{-1}$.

**Bibliography**


Mass transfer in a rotor-stator spinning disc reactor with co-feeding of gas and liquid

This chapter has been published as:

Abstract

This chapter presents a new type of spinning disc reactor configuration for gas-liquid operations. It combines the features of a classical spinning disc with a liquid film on the rotor (e.g., Aoune and Ramshaw (1999)) and those of a rotor-stator spinning disc unit with a single gas inlet in the bottom stator (Meeuwse et al., 2010). In this new configuration, gas and liquid are co-fed through an inlet in the top stator. It is shown that gas-liquid mass transfer mainly takes place in the dispersed region between the rotor and the bottom stator. \( k_{GL} a_{GLV_R} \) in this region is up to a factor of 6 larger than in the region with the liquid film on the rotor. Simulation of gas desorption from a saturated liquid shows that the gas-liquid mass transfer in this co-fed configuration is considerably improved in comparison to the separate reactors, at similar operating conditions. The new reactor has also higher potential for scaling-up: gas and liquid can be co-fed from one rotor-stator unit to another without the need for redistribution of the gas.
3.1 Introduction

Gas-liquid and/or liquid-solid mass transfer are often the rate limiting steps in multiphase catalytic reactors. Improving these mass transfer steps may therefore increase the overall reaction rate, and thus the production capacity, or it can decrease the reactor size at a given production rate. Smaller, more efficient reactors decrease the production costs, and increase the safety, due to the lower inventory of chemicals. The reduction in energy consumption of an industrial process can also be the driving force for intensifying the process.

Generally, the rate of gas-liquid mass transfer is a function of the gas-liquid mass transfer coefficient, \( k_{GL} \), the gas-liquid interfacial area, \( a_{GL} \), and the concentration difference between the gas and the liquid phase. The mass transfer coefficient is most often determined by the rate of diffusion of the gas component, or by the rate at which the liquid near the gas-liquid surface is refreshed. A large number of multiphase reactors has the gas phase as the dispersed phase, in the form of gas bubbles, to increase the gas-liquid interfacial area. The size of the gas bubbles, and thus the magnitude of the gas-liquid interfacial area, is determined by the shear stresses during bubble formation or bubble break up. In conventional reactors the surface renewal rate and the shear stresses are caused by gravity or by the power input, e.g. by stirring or by pushing the fluids through the reactor using pressure. Centrifugal forces increase the shear stresses and surface renewal rate, therefore increasing the volumetric mass transfer coefficient \( k_{GL}a_{GL} \). Examples of the use of centrifugal forces in multiphase reactors are spinning discs (Aoune and Ramshaw, 1999, Meeuwse et al., 2010) and rotating packed beds (Ramshaw, 1993).

Conventionally, a spinning disc reactor consists of one single rotating disc with a liquid film flowing over it (see Figure 3.1a). This system has been studied since the 1960’s, at first for heat transfer and evaporation (Espig and Hoyle, 1965), later also for gas absorption into the liquid film (Jachuck and Ramshaw, 1994, Wood and Watts, 1973). Several types of processes were performed in spinning disc reactors, e.g. manufacturing of pharmaceuticals (Oxley et al., 2000), polymerisation (Boodhoo et al., 1997) and crystallisation (Brechtmubauer et al., 2001). The type of liquid flow over the rotating disc and the liquid film thickness depend on the liquid flow rate and on the rotational speed of the rotor (Burns et al., 2003, Espig and Hoyle, 1965, Thomas et al., 1991, Wood and Watts, 1973). The type of liquid flow over the disc changes with increasing rotational disc speed and liquid flow rate, from rivulets, via a uniform liquid film, to a liquid film with surface waves. These surface waves are expected to increase the gas-to-liquid mass transfer. For example, a grooved disc increases the waviness (Matar et al., 2008), and therefore the mass transfer rate (Jachuck and Ramshaw,
Figure 3.1: (a) "Conventional" spinning disc reactor with liquid film on the rotor (Aoune and Ramshaw, 1999). (b) Rotor-stator spinning disc reactor with a single gas inlet in the bottom stator (Meeuwse et al., 2010). (c) Schematical drawing of the gas and liquid flow in the co-fed spinning disc reactor (this work). (d) Region F: Liquid film on the rotor with gas bubble formation at the rim of the rotor. (e) Region D: Small bubbles surrounding the rim of the rotor and between the rotor and the bottom stator.
In our previous work a so-called rotor-stator spinning disc reactor was presented (Meeuwse et al., 2010). In this particular rotor-stator configuration, the spinning disc is enclosed by a cylindrical housing (see Figure 3.1b). The distance between the rotor and the stators on top and bottom of the rotor is typically 1 mm. The liquid is the continuous phase. The gas is injected via a single gas inlet in the bottom stator. Small bubbles of 1 mm diameter and smaller are formed due to the high shear force between the rotor and the stator. The gas bubbles move to the centre of the reactor, as a result of the centrifugal force, leading to a low gas holdup of approximately $0.02 \, \text{m}^3 \, \text{m}^{-3} \, \text{R}$. Still, volumetric gas-liquid mass transfer rates up to $0.43 \, \text{m}^3 \, \text{L}^{-1} \, \text{m}^{-3} \, \text{R}^{-1}$ were measured. The gas holdup, and thus the gas-liquid mass transfer rate, can be increased by the use of multiple gas inlets in the stator instead of one. A disadvantage is that a gas distribution system is then needed for the gas supply. Another disadvantage is that the region between the rotor and the top stator and the region surrounding the rim of the rotor are not taking part in the gas-liquid mass transfer.

Here, in the present paper, we present an improved design of this rotor-stator spinning disc reactor wherein the gas and the liquid are now fed together through the inlet in the centre of the top stator, close to the rotating axis, as shown schematically in Figure 3.1c. A liquid film is present on top of the rotor and a gas-liquid bubbly dispersion is located between the rotor and the bottom stator. This new configuration is thus a combination of a conventional spinning disc (Aoune and Ramshaw, 1999) and the rotor-stator spinning disc reactor (Meeuwse et al., 2010). It will be shown that the combination of the film flow on the rotor and the dispersed flow in the remainder part of this rotor-stator reactor leads to higher gas-liquid mass transfer rates than in the case of the separate configurations.

This reactor system has a high potential for scaling up, by using several rotor-stator units in series, possibly with the rotors on a common axis. The gas-liquid outlet flow from one unit can directly be fed into the next one, thus avoiding the cumbersome step of redistribution of the gas. One spinning disc unit consists of a part in which the liquid is in plug flow and a part in which the liquid is ideally mixed, as will be shown later. For most reactions plug flow behaviour may be preferred, since this gives often higher conversions and selectivities. A number of spinning disc reactors in series will then behave as a plug flow system.

This paper presents experimental gas-liquid mass transfer data in this new rotor-stator spinning disc reactor. The reactor is split up in two regions, the liquid film on the rotor
and the dispersed flow in the rest of the reactor. The contribution of the two regions to the
gas-liquid mass transfer is quantified.

3.2 Gas-liquid mass transfer

The co-fed rotor-stator spinning disc reactor can be divided in two regions, the film flow
region F and the dispersed region D, as depicted in Figure 3.1(c). In the following paragraphs
these regions are discussed in detail.

3.2.1 Region F: Liquid film on the rotor

The liquid and the gas are injected through the inlet of the reactor in the top stator, near the
centre of the disc. The liquid flows as a thin film over the disc, with surface waves at higher
rotational disc speeds (see Figure 3.1d). The rotor surface is completely wetted at a radial po-
sition exceeding the inlet radial position, resembling the situation in a conventional spinning
disc reactor (Burns et al., 2003). The gas phase is present between the liquid film and the top
stator.

Aoune and Ramshaw (1999) showed that the gas-liquid mass transfer on a spinning disc
increases with increasing rotational disc speed and liquid flow rate. Peev et al. (2007) pre-
sented a Sherwood correlation of the gas-liquid mass transfer, based on a laminar film model
that was modified to account for the presence of surface waves. Their correlation describes
the experimental data of Aoune and Ramshaw (1999) well, with an average error of 6% and
a maximum error of 18%. The original \( Sh \)-correlation (Peev et al., 2007) has been rewritten
\( Re \) is used for \( \frac{1}{Ek} \) and \( C_w \) for the \( Re \) defined by Peev et al. (2007)) and is described here as a
function of three dimensionless groups (see nomenclature):

\[
\overline{Sh_F} = 10.8 \cdot 10^{-4} Re^{0.94} C_w^{0.24} \beta^{\frac{1}{7}}
\]  

(3.1)

Equation 3.1 is used here to estimate the gas-liquid mass transfer coefficient, \( k_L \), in region
F. Approximately 2 mm from the rim of the rotor the film region ends, where small bubbles
shear off. The rest of the surface of the rotor is covered with the liquid film. The volume of
region F is therefore taken as the volume between the rotor and the top stator, from the centre
of the reactor to the radius of the rotor, which is \( 13.7 \cdot 10^{-6} \) m\(^3\). The gas-liquid surface area
per volume of this region is then:

\[
a_{GL} = \frac{A_{top}}{V_F} = \frac{\pi R_D^2}{h \pi R_D} = \frac{1}{h} = 1 \cdot 10^3 m^2 m^{-3}
\]

(3.2)
The effect of the increase in the surface area by the surface waves is accounted for in Equation 3.1. The liquid film on the rotor is assumed to be in plug flow. It has higher radial and tangential velocities than the gas phase (Burns et al., 2003). The no-slip condition at the liquid film interface makes the gas move rapidly with the liquid, in the radial and tangential directions, whereas the same condition makes the gas flow to be zero at the stator. The resulting large velocity gradients induce a recirculation pattern, since the rate at which the gas is dragged outwards is much larger than the net gas flow rate. The gas phase in region F is therefore considered to be ideally mixed.

### 3.2.2 Region D: Dispersed region

Small gas bubbles are sheared off from the gas phase of region F at the rim of the rotor (see Figure 3.1d), due to the radial and tangential velocity components of the liquid. The gas bubbles in the liquid surrounding the rotor have a tangential velocity, due to the movement of the liquid. The centrifugal force causes the gas bubbles to flow inwards. The gas bubbles then either flow back to region F, where they coalesce with the gas phase, or they flow into the region between the rotor and the bottom stator (see Figure 3.1e). The centrifugal force, which is directed outwards, increases the inward radial velocity of the gas bubbles, due to the density difference between the gas and the liquid. The drag force between the gas bubble and the liquid balances the centrifugal force, resulting in a radial velocity of the gas bubble which depends on its radial position and on its size. The bubble’s radial velocity increases with increasing bubble diameter, since the centrifugal force depends on the volume of the gas bubble, and the drag force depends on the projected area of the bubble perpendicular to the flow direction. The gas bubbles follow a spiral path towards the outlet in the centre of the bottom stator.

Meeuwse et al. (2010) presented the rotor-stator spinning disc reactor with a single gas inlet in the bottom stator. The shear force in the liquid, which is caused by the rotation of the rotor, tears the gas bubbles from the gas inlet. The dispersed flow in the part between the rotor and the bottom stator of region D of the co-fed spinning disc reactor closely resembles the flow of gas bubbles using this single gas inlet in the bottom stator.

Region D can be described as an ideally mixed liquid volume, due to the internal recycle of liquid that is present in this rotor-stator system (Daily and Nece, 1960, Dijkstra and van Heijst, 1983, Poncet et al., 2005). At the rotor the liquid radial velocity is directed outwards, at the stator it is directed inwards. The corresponding liquid flow rates are up to a factor 50 higher than the net liquid flow rates used in this study (Owen and Rogers, 1989). The
The gas bubbles in region D flow in a spiral pattern towards the reactor outlet in the centre of the reactor. The radial velocity is dependent on the gas bubble size, however, only a relatively small difference in residence time distribution exists. The gas phase in region D will therefore be treated as plug flow.

The gas-liquid mass transfer in the co-fed rotor-stator stator spinning disc reactor is evaluated with the reactor model shown in Figure 3.2, where regions F and D are in series. The liquid film in region F and the gas phase in region D are in plug flow, the gas phase in region F and the liquid flow of region D are ideally mixed.

3.3 Experimental

A rotating disc, made of sandblasted stainless steel, of $3 \cdot 10^{-3}$ m thickness and 0.132 m diameter, is enclosed in a cylindrical housing made of poly(methyl methacrylate) (PMMA), as shown in Figure 3.3. The distance between the stators and the rotor is $1 \cdot 10^{-3}$ m, the
distance between the outer rim of the rotor and the cylindrical wall of the reactor is $1 \cdot 10^{-2}$ m, leading to a reactor volume of $50 \cdot 10^{-6}$ m$^3$. The maximum rotational disc speed of the rotor is 179 rad s$^{-1}$ (1710 rpm). The liquid and the gas are combined in a T-junction (with internal tube diameters of $4 \cdot 10^{-3}$ m), and flow into the reactor via an inlet ($2 \cdot 10^{-3}$ m I.D.) in the top stator, at a radial position of $3.3 \cdot 10^{-2}$ m.

The gas-liquid interfacial area of the gas bubbles between the bottom stator and the rotor is determined using image analysis. A picture of the bottom of the reactor, which is transparent, is made with a Canon EOS digital 400D camera, via a mirror which is adjusted at $45^\circ$. The shutter time of the camera is 0.1 s. A Phillips PR 9113 stroboscope is used to produce a flash of less than $1 \cdot 10^{-5}$ s, which leads to the same exposure time of the camera. In this time the bubbles move less than one pixel, making it possible to get a sharp photograph.

The photographs are converted into a black and white picture with Matlab, resulting in an image in which only the gas bubbles are visible. Gas bubbles smaller than the rotor-stator spacing are assumed to be spherical; larger bubbles are assumed to be flattened. With the number of bubbles, their sizes and their shapes, the gas holdup and the gas-liquid interfacial area are calculated. A detailed description of the method to estimate the gas-liquid interfacial area can be found in Meeuwse et al. (2010)
3.3.1 Volumetric mass transfer coefficient

The volumetric mass transfer coefficient, $k_{GLaGL}$, in region F, based on plug flow behaviour of the liquid phase and ideally mixed behaviour of the gas phase, is calculated with the following equation:

$$
(k_{GLaGL})_F = \frac{\phi_L}{V_R} \ln \left[ \frac{\frac{RT}{H} C_G^{out} - C_L^{in}}{\frac{RT}{H} C_G^{out} - C_L^{out}} \right] 
$$ (3.3)

The volumetric mass transfer coefficient, $k_{GLaGL}$, in region D, is calculated from the concentration difference of oxygen between the inlet and the outlet of the region, for a gas phase which behaves as plug flow and a liquid phase which is ideally mixed, with the following equation:

$$
(k_{GLaGL})_D = \frac{\phi_G}{V_R} \frac{H}{RT} \ln \left[ \frac{\frac{RT}{H} C_G^{in} - C_L^{out}}{\frac{RT}{H} C_G^{out} - C_L^{out}} \right] 
$$ (3.4)

The gas-liquid mass transfer rate is determined by the desorption of oxygen from water using nitrogen as the gas phase. In an external $2 \cdot 10^{-3}$ m$^3$ vessel the oxygen concentration is increased, by bubbling air through the water. The liquid, saturated with oxygen up to around 85%, is then pumped into the reactor. The oxygen concentrations in the liquid at the liquid inlet, before the mixing with the gas, and at the reactor outlet are measured using fiber optic oxygen probes (AVS-OXYPROBE, Avantes). All the measurements are performed in steady state. The gas phase concentration in the outlet is in both cases calculated from the mole balance of oxygen over the volume of the region:

$$
C_G^{out} = C_G^{in} + \frac{\phi_L}{\phi_G} (C_L^{in} - C_L^{out}) 
$$ (3.5)

The inlet oxygen concentration in the liquid is measured before the gas flow and the liquid flow are combined in the T-junction. In this T-junction and in the tubing between the T-junction and the reactor inlet, a certain amount of oxygen is already transferred from the liquid phase to the gas phase. The rate of transfer in this inlet section, which depends on the gas and liquid flow rates, is determined independently by coupling the inlet and outlet tubes of the reactor. The volumetric gas-liquid mass transfer coefficient in the inlet section is calculated using the plug flow assumption for both gas and liquid:

$$
(k_{GLaGL})_{inlet} = \frac{\ln \left[ \frac{\frac{RT}{H} C_G^{in} - C_L^{in}}{\frac{RT}{H} C_G^{out} - C_L^{out}} \right]}{V_R \left( \frac{RT}{H\phi_G} + \frac{1}{\phi_L} \right)} 
$$ (3.6)
The actual concentrations of oxygen in the gas and in the liquid at the reactor inlet are calculated with this volumetric mass transfer coefficient. With these concentrations the volumetric gas-liquid mass transfer coefficient of the co-fed spinning disc reactor is corrected for the effect of mass transfer in the T-junction and in the inlet tube.

3.4 Results and discussion

Figure 3.4: Volumetric gas-liquid mass transfer coefficient for various gas flow rates in region D, at a constant liquid flow rate of $6.7 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$ as a function of the rotational disc speed.

$k_{GL}a_{GL}$ in region D is shown in Figure 3.4. It increases with increasing rotational disc speed and gas flow rate. The maximum value measured is $0.95 \text{ m}^3 \text{m}^{-3} \text{s}^{-1}$ at a gas flow rate of $7.3 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$, a liquid flow rate of $6.7 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$, and a rotational disc speed of 179 rad s$^{-1}$. The observed trend in this volumetric gas-liquid mass transfer coefficient is the same as with a single gas inlet in the bottom stator (Meeuwse et al., 2010).

The gas-liquid interfacial area when co-feeding gas and liquid, is compared with the gas-liquid interfacial area using a single gas inlet in the bottom stator in Figure 3.5. The gas-liquid interfacial area is determined by the image analysis method described in the Experimental section, and therefore only the gas bubbles between the rotor and the bottom stator are taken into account, the gas bubbles in the region surrounding the rim of the disc are not included. The difference in the gas-liquid interfacial areas for the two cases is small, only at a rotational disc speed of 179 rad s$^{-1}$ the gas-liquid interfacial area with co-feeding of gas and liquid is more than 20 % higher than by using a single gas inlet in the bottom stator.
Figure 3.5: The gas-liquid interfacial area between the rotor and the bottom stator, with a single gas inlet in the bottom stator, and with co-feeding of gas and liquid, at a liquid flow rate of $6.7 \cdot 10^{-6}$ m$^3$ s$^{-1}$ and gas flow rate of $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$.

Figure 3.6: The product of the volumetric gas-liquid mass transfer coefficient and the region volume, $k_{GL}a_{GL}V_R$, for regions F and D, and for a single gas inlet in the bottom stator, at a liquid flow rate of $6.7 \cdot 10^{-6}$ m$^3$ s$^{-1}$ and gas flow rate of $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$.

The gas-liquid mass transfer coefficient, $k_{GL}$, between the rotor and the bottom stator, depends on the amount of turbulence in the liquid (Fortescue and Pearson, 1967, Lamont and Scott, 1970), and thus on the rotational disc speed. The gas-liquid mass transfer between the
rotor and the bottom stator in region D is therefore expected to be similar to the gas-liquid mass transfer using a single inlet in the bottom stator. The product of the volumetric gas-liquid mass transfer coefficient and the region volume, $k_{GL}a_{GL}V_R$, for a single gas inlet, and for the regions F and D with co-feeding of gas and liquid, are shown in Figure 3.6. This quantity represents the overall gas-liquid mass transfer rate divided by the (average) driving force in concentration between the gas and the liquid. In region D $k_{GL}a_{GL}V_R$ is comparable to the situation with the single gas inlet. Only at rotational disc speeds higher than 150 rad s$^{-1}$ a difference of more than 20 % is observed. $k_{GL}a_{GL}V_R$ thus shows the same trend as the gas-liquid interfacial area between the rotor and the bottom stator. The main part of the gas-liquid mass transfer in region D therefore occurs in the region between the rotor and the bottom stator. The region surrounding the rotor only provides a small contribution to the rate of gas-liquid mass transfer, in spite of its large volume, which is 1.6 times larger than the part of region D which is in between the rotor and the bottom stator. The shear force, and thus the energy dissipation, is much smaller in this rim region, because the distance from the rim of the rotor to the cylindrical wall is 10 times larger than the distance between the rotor and the stator. As a result of this, the volumetric gas-liquid mass transfer coefficient in the complete region D is lower than for a single gas inlet, as shown in Figure 3.7.

![Figure 3.7: The volumetric gas-liquid mass transfer coefficient, for regions F and D, and for the case of a single gas inlet in the bottom stator, at a liquid flow rate of $6.7 \cdot 10^{-6}$ m$^3$ s$^{-1}$ and gas flow rate of $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$. In the case of the single gas inlet in the bottom stator, the gas-liquid mass transfer only takes place between the rotor and the bottom stator; the volume of this region is therefore used for the calculation of the volumetric gas-liquid mass transfer coefficient.](image)

The rate of formation of small gas bubbles at the border between the regions F and D
Large gas bubble

Figure 3.8: Photographs of the region between the rotor and the bottom stator, at a liquid flow rate of $6.7 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$, a gas flow rate of $7.3 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$ and a rotational disc speed of $179 \text{ rad s}^{-1}$. A large gas bubble, which has a larger radial velocity than the rest of smaller the bubbles, crosses the region to the outlet at the centre of the disc. The gas bubble in this picture is approximately $2 \cdot 10^{-2} \text{ m}$ in diameter.

depends on the rotational disc speed. If the volume rate of the gas bubbles that are sheared off at the rim of the disc is lower than the gas flow rate, a volume of gas will build up in region F. The excess of gas leaves region F periodically as a large gas bubble (see Figure 3.8), with a diameter between $1.5 \cdot 10^{-2}$ and $3 \cdot 10^{-2} \text{ m}$. This large gas bubble crosses the region between the rotor and the bottom stator much faster than the small bubbles. With increasing rotational disc speed, the frequency of the formation of these large gas bubbles decreases, because the small gas bubbles are sheared off faster at higher rotational disc speeds. These large gas bubbles have only a small influence on the gas-liquid interfacial area because the number of large gas bubbles is very small in comparison to the number of small gas bubbles. However, the number of small bubbles that are formed will also be smaller than for a single gas inlet, due to these large gas bubbles. The gas-liquid mass transfer coefficient in region D will therefore also be smaller, since large bubbles have a smaller interfacial area per gas volume and a shorter residence time. The gas-liquid mass transfer coefficient between the rotor and the bottom stator is therefore lower for the co-fed spinning disc reactor than for the case of the single inlet in the bottom stator. The influence of the region surrounding the rotor is therefore larger than expected only on the base of Figure 3.6.
Figure 3.9: Dimensionless liquid concentration at the outlet of the reactor for the three spinning disc configurations, for a saturated liquid and a nitrogen gas phase at the inlet. The concentrations are calculated with the $k_{GL}a_{GL}$ values presented in this paper, at a liquid flow rate of $6.7 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$ and gas flow rate of $7.3 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$.

In Figure 3.6 it is shown that $k_{GL}a_{GL}V_R$ is a factor 4 to 6 higher in region D than in region F, at a gas flow rate of $7.3 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$. The gas-liquid mass transfer in region F does not depend on the gas flow rate, contrary to the gas-liquid mass transfer in region D, which is strongly influenced by the gas flow rate (see Figure 3.4). The influence of region F on the overall mass transfer therefore decreases with increasing gas flow rate. At a gas flow rate of $1.8 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$, $k_{GL}a_{GL}V_R$ in region D is only a factor 1.8 to 3.2 higher than in region F.

In Figure 3.6 it is shown that $k_{GL}a_{GL}V_R$ in region D is higher than in the case of the single gas inlet in the bottom stator. The co-fed spinning disc reactor is the combination of regions F and D, the mass transfer performance is therefore also larger than in the case of a single inlet or a spinning disc reactor with a liquid film on the rotor. The values of $k_{GL}a_{GL}V_R$, as shown in Figure 3.6, are used to calculate the oxygen concentration in the liquid at the outlet of the three spinning disc configurations for a stripping experiment, starting with oxygen saturated liquid. The results of this simulation are shown in Figure 3.9, as a function of the rotational disc speed. The rotor-stator spinning disc reactor with co-feeding of gas and liquid decreases the dimensionless liquid concentration to 0.09, at a rotational disc speed of 179 rad s$^{-1}$. The concentrations in the spinning disc reactor with a liquid film on the rotor, and the rotor-spinning disc reactor with a single gas inlet in the bottom stator only reach values of 0.43 and 0.21, respectively. The co-fed rotor-stator spinning disc reactor therefore shows a much better mass transfer performance.
3.5 Concluding remarks

The rotor-stator spinning disc reactor with the co-feeding of gas and liquid combines the flow as observed in the spinning disc reactor with a liquid film on the rotor (Aoune and Ramshaw, 1999) and the rotor-stator spinning disc reactor with dispersed gas flow (Meeuwse et al., 2010). A reactor model is presented in which the two regions are evaluated individually: the film region, where the liquid phase is in plug flow, and the dispersed region, where the liquid phase is ideally mixed. $k_{GL}a_{GL}V_R$ in the dispersed region is a factor 1.8 to 6 higher than in the film flow region. With increasing gas flow rate the relative contribution of the dispersed region increases. The gas-liquid interfacial area between the rotor and the bottom stator with co-feeding of gas and liquid is comparable to the situation with a single gas inlet in the bottom stator. The gas-liquid mass transfer coefficient $k_{GL}$ is expected to be the same for the two situations, since it depends on the rotational disc speed. Most of the mass transfer in region D will therefore take place between the rotor and the bottom stator. A simulation of a stripping process results in an outlet concentration of 9% in the co-fed spinning disc reactor, while this is 43% and 21% for a conventional spinning disc reactor, and the rotor-stator spinning disc reactor with a single gas inlet, respectively. The co-fed rotor-stator spinning disc reactor therefore outperforms these other configurations in terms of gas-liquid mass transfer.

Bibliography


Effect of rotor-stator distance and rotor radius on the rate of gas-liquid mass transfer in a rotor-stator spinning disc reactor

This chapter has been submitted as:

Abstract

This chapter describes the effect of rotor radius, rotor-stator distance, liquid flow rate and rotational disc speed on the rate of gas-liquid mass transfer in a rotor-stator spinning disc reactor. Rotor radii of 0.066 m and 0.135 m are studied with rotor-stator distances of 1, 2 and 5 mm, at rotational disc speeds up to 209 rad s$^{-1}$. At rotational disc speeds lower than 70 rad s$^{-1}$, elongated gas bubbles are formed, that are larger than the rotor-stator distance. At rotational disc speeds above 100 rad s$^{-1}$, spherical gas bubbles are formed that are smaller than the rotor-stator distance. The volumetric gas-liquid mass transfer coefficient increases with increasing rotational disc speed and decreases with increasing liquid flow rate. This decrease is larger than predicted by the Wallis drift flux model because of the complex two-phase flow pattern. The rate of gas-liquid mass transfer per unit of reactor volume increases with decreasing rotor-stator distance. The maximum observed volumetric mass transfer coefficient in case of the 0.135 m rotor is a factor 3 higher than in case of the 0.066 m rotor, while the rate of energy dissipation is a factor 15 higher.
4.1 Introduction

The rotor-stator spinning disc reactor is a novel type of multiphase reactor, which is shown to have high gas-liquid and liquid-solid mass transfer rates compared to conventional reactor equipment (Meeuwse et al., 2010a,b,c). The spinning disc reactor consists of a rotating disc (rotor) and a reactor wall (stator) on both sides of the disc (see Figure 4.1), with a small gap in between. Liquid is injected to the reactor through the top stator, gas is injected via a gas inlet in the bottom stator, near the rim of the disc. The high rotational disc speed creates a large shear force between the rotor and the stator, thereby breaking up gas bubbles. This results in a high gas-liquid interfacial area, $a_{GL}$. The turbulence induced in the system due to the rotation results in high values of the gas-liquid mass transfer coefficient, $k_{GL}$. The combination gives a high volumetric gas-liquid mass transfer coefficient, for example, $(k_{GL} a_{GL}) = 0.43 \text{ m}^3 \text{ m}^{-3} \text{s}^{-1}$ with a rotor radius of 0.066 m and a rotational disc speed of 179 rad s$^{-1}$. This is a factor 3 higher than in conventional reactors like bubble columns and stirred tanks, in spite of the low gas holdup, which is less than 2% (Meeuwse et al., 2010b). The gas holdup, and thus the efficiency of the reactor is increased by using multiple gas inlets. However, this may not be preferable if the reactor has to be scaled up to a multiple disc system, since a (re)distribution of the gas is then needed. If gas and liquid are fed together through the inlet in the top of the reactor, a liquid film will form on top of the rotor. Small gas bubbles will be sheared off at the rim of the rotor, the gas-liquid dispersion will fill the rest of the reactor, i.e. the region surrounding the rim of the disc, and the region between the rotor and the bottom stator. The reactor volume is used more efficiently in this configuration, and therefore gives rise to a significant increase in gas-liquid mass transfer (Meeuwse et al., 2010c).

The aforementioned results were all obtained using an experimental setup with a rotor radius of 0.066 m and a rotor-stator distance of 1 mm. The gas bubble diameter, and therefore also the gas-liquid interfacial area, is mainly determined by the gas flow rate and the shear force between the rotor and the stator at the gas inlet. The magnitude of the velocity gradient, and thus the shear force, depends on the rotor-stator distance, and on the tangential velocity of the rotor, which is the product of the radius and the rotational disc speed. The gas-liquid interfacial area is therefore expected to increase with increasing rotor radius and decreasing rotor-stator distance. The gas-liquid mass transfer coefficient depends on the energy input in the system per unit of reactor volume, which is also strongly affected by the rotor-stator distance and the rotor radius. An increase in the gas-liquid mass transfer coefficient is thus expected with increasing rotor radius and decreasing rotor-stator distance. As a result, the volumetric gas-liquid mass transfer coefficient is also expected to increase with increasing rotor radius and decreasing rotor-stator distance. This paper describes the influence of the
Figure 4.1: Schematic representation of the rotor-stator spinning disc reactor with a gas inlet in the bottom stator. The reactor consists of a rotating disc in a cylindrical housing. Liquid is fed to the reactor from the top inlet, near the rotating axis. Gas is injected through an orifice in the bottom stator. Gas bubbles are sheared off due to the high velocity gradient in between the rotor and the stator. The surface renewal rate near the gas bubbles is large due to the turbulence induced by the rotation of the disc, which leads to a high gas-liquid mass transfer coefficient. The same mechanism causes a high rate of mass transfer from the bulk of the liquid to the rotor and the stator. More information about the dimensions of the reactor used in this study is shown in Figure 4.3, photographs of the gas-liquid dispersion between the rotor and the stator are shown in Figures 4.5 and 4.7.

For an industrial application of the rotor-stator spinning disc reactor in a given process, the mass transfer of the reactor at a certain flow rate is most often the determining factor. In this study it is therefore chosen to compare the different reactor configurations at constant liquid and gas flow rates. The conditions used for the comparison of the rotor-stator distances used in this paper are: a liquid flow rate of $3 \times 10^{-5} \text{ m}^3 \text{ s}^{-1}$ and a gas flow rate of $1.5 \times 10^{-5} \text{ m}^3 \text{ s}^{-1}$. 
4.2 Theory

In the next section, a short summary of literature on single phase flow in rotor-stator systems is presented; in particular those references will be considered that are relevant for the work presented in this paper. Data on two-phase flow in rotor-stator systems is scarce and the gas-liquid flow in between a rotor and a stator is not yet well understood. Here we will present a simplified model, based on the Wallis drift flux model, that we will apply as a tool in the explanation of the experimental data obtained. Additionally, models for the gas-liquid mass transfer coefficient are presented.

4.2.1 Single phase flow in rotor-stator systems

Single phase flow in rotor-stator systems has been researched extensively in the past decades. Four different flow regimes can be distinguished, two laminar regimes and two turbulent regimes. The mean velocity profiles of the two turbulent regimes have approximately the same shape as the two laminar regimes. Only the turbulent regimes are described here, since all the experiments performed in the present study are in these regimes.

Figure 4.2 shows a schematic representation of the mean tangential and radial velocities in the case of single phase flow in a rotor-stator system. At a large rotor-stator distance, two boundary layers are present, one at the stator and one at the rotor. These layers are separated by a core of liquid in which no tangential or radial velocity gradient is present, which thus rotates as a solid body. In the boundary layer at the stator an inward radial velocity is present, at the rotor the radial velocity is directed outwards. The maximum radial velocity is approximately 10-20% of the tangential velocity of the rotor (Batchelor, 1951, Iacovides and Theofanopoulos, 1991, Itoh et al., 1992, Poncet et al., 2005a, Randriamampianina et al., 1997). The thickness of the boundary layers depends on the rotational disc speed and the rotor radius, and is proportional to the Reynolds number \( Re = \frac{\omega R^2}{v} \) to the power 1/5 (Djaoui et al., 2001, Owen and Rogers, 1989). When the rotor-stator distance is decreased, the central core decreases in size, until it vanishes. At a lower rotor-stator distance the boundary layers merge; no region without a tangential velocity gradient is present anymore. The transition between the two flow regimes occurs at \( GRe^{1/5} \approx 0.5 \) (Djaoui et al., 2001). The experimental setup as used in this study operates in the regime with the separate boundary layers (as shown in the top part of Figure 4.2) at 5 mm rotor-stator distance and a disc radius of 0.135 m. In the rest of the experiments, i.e. \( h = 1 \) mm and \( h = 2 \) mm at \( R_D = 0.135 \) and \( h = 1 \) mm
Figure 4.2: Schematic representation of the mean tangential (left) and radial (right) velocities in a rotor-stator system with a large rotor-stator distance (top) and a small rotor-stator distance (bottom). In the case of a large rotor-stator distance two separate boundary layers are formed, at the rotor and the stator, separated by a region rotating at a constant tangential velocity, approximately 0.3-0.45 times the velocity of the rotor (Itoh et al., 1992, Poncet et al., 2005c). No radial velocity is present in this region. The size of this region decreases with decreasing rotor-stator distance. Below a certain rotor-stator distance the two boundary layers merge, resulting in a continuous variation in tangential and radial velocity (Daily and Nece, 1960, Dijkstra and van Heijst, 1983, Schouveiler et al., 2001). The transition between the two flow regimes occurs at $GRe^{1/5} \approx 0.5$ (Djaoui et al., 2001). The drawings are schematic representations of the velocity profiles; in the region with the merged boundary layers nor accurate experimental or numerical results are found in literature.

at $R_D = 0.066$ m, the boundary layers are merged, as indicated in the bottom part of Figure 4.2.

The velocity profiles in Figure 4.2 are without a net throughflow of liquid. In the case of an inward flow, the thickness and velocity of the stator boundary layer increase, while these quantities decrease in the rotor boundary layer. A large increase in centripetal flow can lead to an inward flow in the rotor boundary layer. The tangential velocity in the core region increases with increasing centripetal flow rate. This can even lead to a tangential velocity of the liquid in this core region which is higher than the tangential velocity of the rotor (Poncet et al.,
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Although this happens at conditions which are, in the present study, only reached very close to the axis. Therefore this effect is neglected in the derivation of the holdup model presented in the next section.

4.2.2 Two phase flow and gas-liquid mass transfer

The rate of gas-liquid mass transfer depends on the product of the gas-liquid interfacial area, $a_{GL}$, and the mass transfer coefficient, $k_{GL}$. The gas-liquid interfacial area depends on the gas holdup and the gas bubble diameter. The mass transfer coefficient is determined by the energy dissipation rate in the system, or by the relative velocity of the gas bubbles and the liquid.

4.2.3 Gas bubble size, gas holdup and gas-liquid interfacial area

The gas-liquid interfacial area depends on the gas bubble size and the gas holdup in the reactor. The gas bubbles are formed at the gas inlet, the bubble size is determined by the shear force at the gas inlet. Duhar and Colin (2006) and Terasaka et al. (2003) studied the bubble detachment from an orifice in a shear flow, but only at low shear rates, in the order of $10 \text{s}^{-1}$, which is 3 orders of magnitude lower than in the rotor-stator spinning disc reactor. The gas bubbles size decreases with increasing shear rate (Duhar and Colin, 2006, Terasaka et al., 2003). This is also observed in the rotor-stator spinning disc reactor, where the gas bubble size decreases with increasing rotational disc speed (Meeuwse et al., 2010b). No bubble breakup and coalescence is observed, only at 0.02 m from the centre of the reactor the bubbles coalesce to larger gas bubbles.

The gas bubbles in the rotor-stator spinning disc reactor are split in two categories. At low rotational disc speeds (typically below 80 rad s$^{-1}$), large gas bubbles are formed, with a diameter which is larger than the rotor-stator distance. At higher rotational disc speeds (commonly above 100 rad s$^{-1}$), the gas bubbles are smaller than the rotor-stator distance. The gas holdup and the gas-liquid mass transfer mechanism are different for the two types of bubbles.

The gas holdup depends on the inward radial velocity of the gas bubbles. The radial velocity is determined by the drag force and the centrifugal force. The flow rate of liquid, which is directed radially inwards, has an influence on the inward gas bubble velocity. Near the center of the reactor the area perpendicular to the flow decreases. The influence of the two phase flow rate, i.e. the gas flow and liquid flow rate together, on the radial velocity of the gas bubble will thus increase near the axis. A common method to incorporate the effect
of the two phase flux on the gas holdup is by using the Wallis drift flux model (Wallis, 1969). This model combines the slip velocity between the phases with the two phase flux.

In the Wallis drift flux model it is assumed that the flow is one-dimensional, i.e. that only flow in the radial direction is taken into account. The liquid and gas are assumed to have the same tangential velocity, which only depends on the radial position in the reactor, and there is no axial variation in tangential and radial velocity. This is in contradiction with the flow regimes as observed in single phase rotor-stator systems (see e.g. Figure 4.2). However, in the case of large gas bubbles, with a diameter which is much larger than the rotor-stator distance (see e.g. Figure 4.13 at 1 and 2 mm rotor-stator distance), the complete axial distance is filled with the gas bubble. The influence of the presence of this gas bubble on the radial and tangential velocity profiles is unknown. Due to the large scale of the bubble, however, it is assumed that these gradients do not contribute to a large extent to the radial velocity of the gas bubble. The one-dimensional flow can thus be used to estimate the gas holdup in the rotor-stator spinning disc reactor for large (i.e. \( d_b \gg h \)) gas bubbles.

The slip velocity as used in the Wallis drift flux model is the difference in interstitial velocities, which can be expressed as a function of the superficial velocities and the gas holdup:

\[
    v_s \equiv v_{G,i} - v_{L,i} = \frac{v_{G,s}}{\varepsilon_G} - \frac{v_{L,s}}{1 - \varepsilon_G}
\]  

(4.1)

The velocities can be divided by the terminal velocity in a stagnant liquid \( (v_\infty) \), leading to:

\[
    \varepsilon_G (1 - \varepsilon_G) \frac{v_s}{v_\infty} = (1 - \varepsilon_G) \frac{v_{G,s}}{v_\infty} - \varepsilon_G \frac{v_{L,s}}{v_\infty}
\]

(4.2)

The slip velocity can be expressed as a function of the gas holdup, using the empirical relation of Richardson and Zaki (1954):

\[
    \frac{v_s}{v_\infty} = (1 - \varepsilon_G)^{n-1}
\]

(4.3)

The parameter \( n \) is a function of the Reynolds number based on the terminal velocity of a single gas bubble in a stationary liquid, and is in this case 2.39. Combining Equations 4.2 and 4.3 leads to:

\[
    \varepsilon_G (1 - \varepsilon_G)^n = (1 - \varepsilon_G) \frac{v_{G,s}}{v_\infty} - \varepsilon_G \frac{v_{L,s}}{v_\infty}
\]

(4.4)
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This relation is used to get an estimate of the gas holdup as a function of the gas flow rate and liquid flow rate, based on the terminal velocity of a single gas bubble in a stationary liquid. This velocity follows from a force balance between the centrifugal force and the drag force. Gas bubbles larger than the rotor-stator distance are assumed to behave as solid cylinders. The terminal velocity of cylindrical gas bubbles is calculated, using $C_D = 1.2$ (Janssen and Warmoeskerken, 2001):

$$v_\infty = \sqrt{\frac{\pi R_b \rho_L - \rho_G (\frac{1}{2} \omega)^2 r}{\rho_L C_D}} \quad (4.5)$$

Experimental observations with a high speed camera show that the gas bubbles have a tangential velocity which is approximately half the velocity of the rotor (Schoenmaekers, 2007). The bubbles are assumed to be uniform in size. Equations 4.4 and 4.5 are used in section 4.4.2 to get an estimate of the gas holdup, which is used in the discussion about the influence of the liquid flow rate on the gas-liquid mass transfer.

The same procedure can be used to estimate the gas holdup in the case of spherical bubbles (where $d_b < h$), which behave as solid spheres (Clift et al., 1978, Wesselingh, 1987). The terminal gas bubble velocity in a stationary liquid, also assuming the gas bubbles have a tangential velocity which is half of the tangential velocity of the rotor, is different however (Wesselingh, 1987):

$$v_\infty = 0.46 \left( \frac{\frac{1}{2} (\omega^2 r)^2 (\rho_L - \rho_G)^2}{\rho_L \mu_L} \right)^{1/3} R_b \quad (4.6)$$

The estimation of the gas holdup using Equations 4.4 and 4.6 is based on the assumption of one-dimensional flow, with no axial velocity gradient, and the same tangential velocity of the gas bubbles and the liquid. For spherical gas bubbles, especially if $d_b \ll h$, however, the diameter of the gas bubbles is in the same order, or even smaller, as the thickness of the boundary layer. The foregoing analysis is thus not valid for small gas bubbles (i.e. $d_b \ll h$). In this case the axial position of the gas bubble is important. If the gas bubble is located in the rotor boundary layer, which is centrifugal, the inward radial velocity is decreased, due to the drag force which acts outwards. In the stator boundary layer, the flow is centripetal, leading to a higher radial velocity than expected based on the centrifugal force. The radial velocity of a gas bubble is thus a strong function of the axial position of the gas bubble. It is therefore not possible to obtain a realistic estimate of the gas holdup in this case, since no information about the axial position of the gas bubbles is available. The quantitative results are thus only used when the gas bubbles are present in the core region, in the case of the flow
regime presented in the top part of Figure 4.2. An example of this is shown in section 4.4.2. This method is not applicable in case the gas bubbles are present in one of the boundary layers.

4.2.4 Mass transfer coefficient

The magnitude of the mass transfer coefficient, $k_{GL}$, depends on the rate of refreshment of the liquid surrounding the gas bubbles. Two different mass transfer mechanisms play a role in the rotor-stator spinning disc reactor, depending on the size of the gas bubbles. At low rotational disc speed large gas bubbles are present, which fill the whole reactor height between the bottom and the stator (see e.g. Figure 4.7 at $\omega = 26 \text{ rad s}^{-1}$). Most of the gas-liquid interfacial area is in this case present as a liquid film between the rotor or stator and the gas bubble. Most of the mass transfer will therefore take place from the gas bubble to this liquid film. This liquid film is not present with the small spherical bubbles, which are smaller than the rotor-stator distance (see e.g. Figure 4.7 at $\omega = 131 \text{ rad s}^{-1}$). The rate of gas-liquid mass transfer is in this case based on the rate of surface renewal at the gas-liquid interface by turbulent eddies in the liquid.

The rate of mass transfer between a large gas bubble and the liquid film can be estimated using the Higbie penetration theory, which is based on the time of contact between the gas and the liquid, before the liquid in the film is mixed with the bulk liquid (Higbie, 1935):

$$k_{GL} = 2 \sqrt{\frac{D}{\pi t_c}} \tag{4.7}$$

As mentioned before, the gas bubbles appear to have a tangential velocity of 0.5 times the velocity of the rotor. The liquid films at the rotor and the stator are assumed to have the same velocities as the rotor ($v_\theta = \omega r$) and the stator ($v_\theta = 0$). The contact times for both liquid films can thus be calculated using the length of the gas bubble perpendicular to the velocity:

$$t_c = \frac{L_b}{\omega/2} \tag{4.8}$$

From this contact time the value of $k_{GL}$ follows:

$$k_{GL} = \sqrt{\frac{2D\omega r}{\pi L_b}} \tag{4.9}$$

The value of $k_{GL}$ as calculated with Equation 4.9 depends on the radial position of the gas bubble, since this position determines the tangential velocity. This value of $k_{GL}$ has to be averaged over the whole reactor volume taking part in mass transfer, i.e. the region between
the rotor and the bottom stator, based on the assumption that the gas bubbles are evenly distributed over this region, and that the bubble length is independent of the radial position.

\[
\bar{k}_{GL} = \frac{\int_0^{V_R} k_{GL} dV}{\int_0^{V_R} dV} = \frac{\int_0^h dz \int_0^{2\pi} d\theta \int_0^{R_D} k_{GL} rdr}{\int_0^h dz \int_0^{2\pi} d\theta \int_0^{R_D} rdr} = \frac{4}{5} \sqrt{\frac{2D\omega R_D}{\pi L_b}} \tag{4.10}
\]

Eq. 4.10 is used as an estimate of the mass transfer in case of large gas bubbles, thus at low rotational disc speeds. In case of higher rotational disc speeds (commonly larger than 100 rad s\(^{-1}\)), most of the gas bubbles are spherical, and smaller than the rotor-stator distance. At these conditions the liquid is turbulent. The surface refreshment rate for the small gas bubbles, is now determined by the velocity and size of the turbulent eddies. The mass transfer coefficient is therefore determined by the energy dissipated in the system, commonly to the power 1/4 (Lamont and Scott, 1970). The energy dissipation is a function of the rotational disc speed and the rotor-stator distance (Equations 4.16-4.18, (Daily and Nece, 1960)). \(k_{GL}\) depends on the rotational disc speed, rotor-stator distance and disc radius as follows:

\[
k_{GL} \propto E_d^{1/4}, \quad E_d \propto \omega^{2/3} R_D^{4/6} h^{-5/6} \quad \Rightarrow \quad k_{GL} \propto \frac{\omega^{11/16} R_D^{1/6}}{h^{1/2} V_R^{1/4}} \tag{4.11}
\]

The reactor volume depends on the disc radius, the distance between the rim of the rotor and the reactor wall (\(w\)) and rotor-stator distance:

\[
V_R = \pi \left( 2hR_D^2 + 4wR_Dh + 2wR_DH_D + 2hw^2 + H_Dw^2 \right) \tag{4.12}
\]

The dependency of \(k_{GL}\) on the rotor-stator distance and disc radius which follows from Equations 4.11 and 4.12 is used in the Results and discussion section (Section 4.4.3) in the analysis of the experimental results.

### 4.3 Experimental

#### 4.3.1 Experimental setup

The rotor-stator spinning disc reactor, as shown in Figure 4.3, consists of a stainless steel rotating disc of 0.135 m radius, in between a top stator (stainless steel) and a bottom stator (PMMA). The maximum rotational disc speed of the rotor is 209 rad s\(^{-1}\) ( = 2000 rpm). The rotor-stator distances can be adjusted by shifting the stators up and down. In this study the
rotor-stator distances on the top and the bottom are 1, 2 and 5 mm. Liquid is fed to the reactor at the top, near the rotating axis. Gas is injected through a gas inlet (0.5 mm inner diameter) in the bottom stator, at a radial position of 0.130 m, near the rim of the disc. The gas and the liquid leave the reactor via an outlet in the center of the bottom stator. The liquid is recirculated via a 5 dm$^3$ storage vessel, which is cooled using a Lauda WKL 1200 circulation chiller, to remove the heat generated by the rotating disc. Gas-liquid mass transfer coefficients for a disc radius of 0.066 m were obtained in previous work (Meeuwse et al., 2010b).

### 4.3.2 Gas-liquid mass transfer measurement

The volumetric gas-liquid mass transfer coefficient is determined with two different methods, a steady state method and a transient method. The steady state method uses the difference in oxygen concentration in the liquid phase between the inlet and the outlet to calculate the gas-liquid mass transfer coefficient. The transient method uses the decrease in oxygen concentration in the storage vessel, which is in a recirculation loop with the reactor, to determine the gas-liquid mass transfer coefficient.

The oxygen concentration is measured at the reactor inlet and, in case of the steady state method, at the reactor outlet, using Ocean Optics FOXY-R fiber optic oxygen sensors. The temperature is measured at the same position. The oxygen sensors are calibrated in oxygen-free and oxygen-rich conditions over the whole temperature range used (5 °C - 30 °C).

In the steady state method, air is fed to the storage vessel, to increase the oxygen concen-
Nitrogen is used as the gas phase in the reactor, to strip the oxygen from the liquid. The volumetric gas-liquid mass transfer coefficient is calculated from the difference in the oxygen concentrations in the liquid at the inlet and outlet of the reactor:

$$k_{GLaGL} = \frac{\phi_G H}{V_R RT} \ln \left[ \frac{\frac{RT}{H} C_{GL}^{in} - C_{GL}^{out}}{\frac{RT}{H} C_{GL}^{in} - C_{GL}^{out}} \right]$$

This equation is based on an ideally mixed liquid phase, and a gas phase as plug flow (Meeuwse et al., 2010b). The oxygen concentration in the gas phase at the outlet follows from a mole balance:

$$C_{GL}^{out} = C_{GL}^{in} + \frac{\phi_L}{\phi_G} (C_{GL}^{in} - C_{GL}^{out})$$

The rate of gas-liquid mass transfer depends on the difference in oxygen concentration between the gas phase and the liquid phase. If the volumetric mass transfer coefficient is high, the driving force, $\frac{RT}{H} C_{GL}^{out} - C_{GL}^{out}$, will be low at the exit. A small error in the oxygen concentration measurement will then have a large influence on the value of $k_{GLaGL}$ as calculated by Equation 4.13. This situation occurs when $k_{GLaGL}$ is high, in combination with low gas and/or liquid flow rates. Under these circumstances the steady state method will not give accurate results.

In the transient method the whole liquid system is saturated with oxygen from air, at the start of the experiment. No air is fed to the storage vessel during the experiment. Nitrogen is again the gas phase in the spinning disc reactor, leading to a liquid outlet oxygen concentration which is lower than the inlet oxygen concentration. The liquid is recirculated, the oxygen concentration in the storage vessel will therefore decrease with time. This decrease is a measure for the rate of gas-liquid mass transfer. The liquid vessel is assumed to be ideally mixed, the oxygen concentration at the reactor inlet as a function of time is then:

$$C_{GL}^{in} = C_{GL}^{in, t=0} e^{\left(\frac{\phi_L}{v_{vessel}} - \frac{RT \phi_G}{\phi_G} \right) \left[ \frac{RT \phi_L}{\phi_G} \left(1-e^{-k_{GLaGL} RT \phi_R / \phi_G} \right) \right] t}$$

The decrease in oxygen concentration is fitted, with $k_{GLaGL}$ as fitting parameter. An example of a measurement, and the corresponding fit, is shown in Figure 4.4. This method can be used when the steady state method is not applicable. Measurements performed under the same conditions with the two methods showed that the $k_{GLaGL}$ values obtained were within 20% of each other, with the highest mass transfer rates, near the edge of applicability of Equation 4.13. This difference is smaller for the cases where the gas-liquid mass transfer is lower.
Figure 4.4: A typical transient oxygen measurement at the inlet and the outlet of the reactor (see Figure 4.3). The liquid concentration at the inlet of the reactor ($C_{L}^{in}$) is measured, and fitted with Equation 4.15 using the value of $k_{GLA}GL$ as fitting parameter. The outlet concentration ($C_{L}^{out}$) is measured as well; the value of $C_{L}^{out}$ calculated with Equation 4.13, based on the inlet concentration and the $k_{GLA}GL$ that gave the best fit, is also shown in the picture.

In both cases it is of the same magnitude as the experimental error of the measurements, which is generally between 10 and 20%.

4.3.3 Torque measurement

The motor used for the rotation of the spinning disc is a SEW Eurodrive CFM71M. The current, which is proportional to the torque applied by the motor, is measured. The torque applied without a rotating disc, i.e. when the engine is running on idle, is subtracted. The energy dissipation rate is calculated using the corrected torque:

$$E_d = \frac{\tau \omega}{V_R}$$ (4.16)

A comparison is made in this paper with the gas-liquid mass transfer in a system with a disc radius of 0.066 m (Meeuwse et al., 2010b). No torque measurements are available for this configuration. The torque for this case is therefore calculated using the following correlation (Daily and Nece, 1960):
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\[
\tau = C_m \frac{1}{2} \rho \omega^2 R_D^5
\]  \hspace{1cm} (4.17)

\[
C_m = 0.08 \left( \frac{h}{R} \right)^{-\frac{1}{6}} Re^{-\frac{1}{4}}
\]  \hspace{1cm} (4.18)

The applicability of this equation is checked with the torque measurements with the disc of 0.135 m radius, with 1 and 2 mm rotor-stator distance, which is also in the range where this correlation is applicable (Djaoui et al., 2001). All the measurements are within 40% of the values obtained by this correlation.

### 4.3.4 Image acquisition

![Figure 4.5: Photograph of the gas-liquid dispersion between the rotor and the bottom stator, at \( \omega = 105 \text{ rad s}^{-1}, \phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}, \phi_L = 3 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}, \) and \( h = 1 \text{ mm}. \) The black rectangle, which is 5.8 by 5.8 cm in size, indicates the position of the detail cuts in Figures 4.7 and 4.13.](image)

Pictures of the gas-liquid dispersion between the rotor and the transparent bottom stator are made via a mirror, which is adjusted at 45°. The shutter time of the Canon EOS 400D camera is 0.1 s. Due to a stroboscope flash, the exposure time of the image is only 10 \( \mu \)s. In spite of this short exposure time, the bubbles on the picture are blurred at rotational disc speeds above 130 rad s\(^{-1}\), due to the high velocity at the rim of the disc. Additionally, the bubbles on the picture sometimes overlap each other, at a rotor-stator distance of more than 1 mm; accurate determination of the bubble size is then not possible. The image analysis method as described in Meeuwse et al. (2010b) can therefore not be used to determine the
gas holdup and the gas-liquid interfacial area in this study. Only a qualitative analysis of the gas-liquid dispersion is made in this paper. An example of a picture of the gas-liquid dispersion is shown in Figure 4.5; the position of the detail cuts for Figures 4.7 and 4.13 is indicated.

## 4.4 Results and discussion

The following sections describe the effect of rotational disc speed, liquid flow rate, rotor-stator distance, and rotor radius on the volumetric gas-liquid mass transfer coefficient. The models for the gas holdup and the gas-liquid mass transfer coefficient presented in the Theory section are used as a tool for the explanation of the experimental results.

### 4.4.1 Rotational disc speed

![Figure 4.6: Volumetric mass transfer coefficient at $R_D = 0.135$, $h = 1$ mm and $\phi_L = 3 \cdot 10^{-5}$ m$^3$ s$^{-1}$. The gas-liquid mass transfer increases with increasing rotational disc speed and gas flow rate.](image)

The volumetric mass transfer coefficient, at 1 mm rotor-stator distance, as a function of the rotational disc speed, is shown in Figure 4.6. It increases with increasing rotational disc speed and increasing gas flow rate. These trends are similar to those observed with a disc radius of 0.066 m (Meeuwse et al., 2010b). The values however, which are up to 2 m$^3$ L m$^{-3}$ s$^{-1}$, are significantly higher than the highest value obtained with $R_D = 0.066$ m, which is 0.43 m$^3$ L m$^{-3}$ s$^{-1}$ at 179 rad s$^{-1}$ and $\phi_G = 7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$. The volumetric gas-liquid mass
Figure 4.7: Gas bubbles in the rotor-stator spinning disc reactor at various rotational disc speeds, at $\phi_G = 1.5 \cdot 10^{-5}$ m$^3$ s$^{-1}$, $\phi_L = 3 \cdot 10^{-5}$ m$^3$ s$^{-1}$, $h = 1$ mm. The gas bubble size decreases with increasing rotational disc speed. The position of the images in the reactor is indicated in Figure 4.5. The photographs at rotational disc speeds above 157 rad s$^{-1}$ are not shown, since the gas bubbles move during the exposure time of the camera ($\approx 10$ $\mu$s), resulting in a blurred image.

Transfer coefficient increases slowly with increasing rotational disc speed up to 100 rad s$^{-1}$, at higher rotational disc speeds it increases more rapidly. In Figure 4.7 images of the gas bubbles at different rotational speeds are shown. At a rotational disc speed of 52 rad s$^{-1}$ or lower, the gas is present as large gas bubbles, which are squeezed in between the rotor and the stator. At 79 rad s$^{-1}$, the first small gas bubbles, which have a diameter smaller than 1 mm, appear. At 131 rad s$^{-1}$ and higher rotational disc speeds, the gas is mainly present in the form of small gas bubbles, which further decrease in size with increasing rotational disc speed. The gas-liquid interfacial area, $a_{GL}$, will thus increase rapidly at rotational disc speeds of 100 rad s$^{-1}$ and higher.
Results and discussion

Figure 4.8: Volumetric mass transfer coefficient at $R_D = 0.135$, $h = 5$ mm and $\omega = 105$ rad s$^{-1}$. The volumetric gas-liquid mass transfer coefficient decreases with increasing liquid flow rate, but the effect is less pronounced than at 1 mm rotor-stator distance.

![Graph showing volumetric mass transfer coefficient vs. liquid flow rate](image)

Figure 4.9: (a) Image of the gas liquid flow at $R_D = 0.135$, $h = 5$ mm, $\omega = 105$ rad s$^{-1}$ and $\phi_L = 3 \cdot 10^{-5}$ m$^3$ s$^{-1}$. (b) Detail cut from 4.9a of 1.5 cm by 1 cm. The stroboscope lamb is adjusted at an angle of approximately 45$^\circ$. The position of the shadows gives an indication of the position of the gas bubble: the closer the shadow is, the closer the gas bubble is located to the rotor.

![Image of stroboscope and gas bubbles](image)
4.4.2 Liquid flow rate

The liquid flow rate is expected to influence the gas holdup, and thus the gas-liquid mass transfer coefficient, as described in section 4.2.3. This section describes the experimental results, and discusses the trends observed, using the models presented before.

The volumetric gas-liquid mass transfer coefficient as a function of liquid flow rate for a rotor-stator distance of 5 mm is shown in Figure 4.8. The gas-liquid mass transfer coefficient decreases significantly, by approximately a factor 2, with an increase in liquid flow rate, at higher liquid flow rates this effect seems to level off. This effect was not investigated in the experiments with a disc radius of 0.066 m. An image of the flow at 105 rad s$^{-1}$ is shown in Figure 4.9a, where a bimodal distribution in gas bubbles is observed. The large gas bubbles, which have an average gas bubble diameter of approximately 2 mm, flow in a spiral pattern towards the center of the reactor. The effect of liquid flow on the gas holdup of these bubbles is expected to be very small, only a decrease of 2.5% according to Equations 4.4 and 4.6.

The small gas bubbles ($d_b \approx 0.5$ mm), are present in the rest of the reactor, which operates in the regime where the boundary layers are separated by a core rotating as a solid body (see Figure 4.2, top drawing). The shadows behind the small gas bubbles (see Figure 4.9b) indicate that most of them are located near the rotor, thus in the boundary layer which is directed outwards. The residence time of these small gas bubbles will thus be significantly higher than for the large gas bubbles. The liquid flow rate is known to have an influence on the velocity and thickness of the rotor boundary layer, and will thus decrease the residence time. The boundary layer at the stator will increase in thickness and velocity, thereby increasing the radial velocity of gas bubbles located in this layer. An increase in liquid flow rate will thus lead to a decrease in gas holdup, which explains the decrease of gas-liquid mass transfer. Unfortunately, the drift flux model as presented in the Theory section, can not be used to predict the gas holdup in this configuration, due to the fact that the gas bubbles are much smaller than the rotor-stator distance.

The decrease in gas-liquid mass transfer as a function of rotational disc speed is even larger in case of a rotor-stator distance of 1 mm and a rotational disc speed of 52 rad s$^{-1}$, as shown in Figure 4.10a. The liquid flow rate is expected to have an influence on the inward radial velocity, and thus the gas holdup and gas-liquid interfacial area, as described in the Theory section. The estimated gas holdup, for a rotor-stator distance of 1 mm, and a rotational disc speed of 52 rad s$^{-1}$ is shown in Figure 4.11. An increase in liquid flow rate of a factor 6 decreases the overall gas holdup by 16 %. The effect of the liquid flow rate on the gas holdup is only observed at a radial position below 0.05 m (Figure 4.11b), while most of
Results and discussion

The volumetric gas-liquid mass transfer coefficient is calculated using the assumption that the liquid phase is ideally mixed. This assumption is based on the velocity profiles in single phase rotor-stator systems, as described in the Theory section. The influence of the presence of gas bubbles on the flow pattern is unknown. The presence of the gas bubbles could have a significant influence on the formation of the boundary layers at the rotor and the stator, and thus on the mixing in the reactor. In the absence of mixing, the liquid is in plug flow, which affects the values of the volumetric mass transfer coefficient obtained from experiments, as shown in Figure 4.10b. The volumetric mass transfer coefficient is then lower, but still the effect of liquid flow rate is observed. Although it is possible that the presence of gas bubbles affects the boundary layers, it is improbable that this would result in complete plug flow behaviour of the liquid phase. This effect, if present, thus does not explain the liquid flow dependency observed.

The volumetric mass transfer coefficients are obtained using two different measurement methods, as described in the Experimental section. Both methods give the same liquid flow rate dependency.
Effect of rotor-stator distance and rotor radius on the rate of gas-liquid mass transfer

Figure 4.11: (a) The gas holdup estimated using the Wallis drift flux model, combined with the slip velocity relation of Richardson and Zaki (Richardson and Zaki, 1954, Wallis, 1969), and Equation 4.5 at $h = 1$ mm, $\omega = 52 \text{ rad s}^{-1}$. The gas bubble radius used is taken from images of the gas bubbles, and is respectively 6.5 mm and 10 mm at $\phi_G = 7.3 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$ and $\phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$. (b) The gas holdup as a function of the radial position in the spinning disc reactor, at $\phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$. The influence of the liquid flow rate is only observed at low radial positions, typically below 0.05 m.

dependency, so it is improbable that the liquid flow dependency is the result of an experimental error. The inlet concentration is measured with a fiberoptic sensor, which is located in the inlet tube of the reactor. An increasing liquid flow rate will increase the pressure in this tube, which could possibly affect the measurements. However, calibration of the sensor at different flow rates, and thus pressures, showed no influence on the volumetric mass transfer coefficient. The pressure in the reactor itself does not depend on the liquid flow rate, since the outlet tube is relatively large, and therefore only has a small pressure drop. This is thus not the source of the influence of the liquid flow rate on the mass transfer coefficient.

An estimate of $k_{GL}$ is made using Equation 4.10, leading to a value of $5 \cdot 10^{-4} \text{ m}^3 \text{ m}^{-2} \text{ s}^{-1}$. With a gas holdup of 0.07 (estimated from the images for $\phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$), this leads to $k_{GL}a_{GL} = 0.06 \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$. The measured value of $k_{GL}a_{GL} = 0.16 \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$, which is thus more than a factor two higher, which means that Equation 4.10 underestimates the mass transfer coefficient. To obtain the value of $k_{GL}a_{GL}$ at the lowest rotational disc speed, the $k_{GL}$ has to be an order of magnitude higher than predicted by Equation 4.10, which indicates that other effects play a role.
The cause of the liquid flow rate dependency is thus not clear. More information has to be available about the local gas and liquid velocities in the rotor stator spinning disc reactor, which is out of scope of this paper, but part of future research. All the other measurements presented in this paper are therefore performed at the same liquid flow rate \((3 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1})\), where the mass transfer coefficient reaches a constant value.

### 4.4.3 Rotor-stator distance

![Graphs showing volumetric mass transfer coefficient and product of volumetric mass transfer coefficient and reactor volume as functions of rotational disc speed.](image)

Figure 4.12: (a) Volumetric mass transfer coefficient at \(R_D = 0.135, \phi_L = 3 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1},\) and \(\phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}\). \(k_{GL}a_{GL}\) increases with decreasing rotor-stator distance. (b) Volumetric mass transfer coefficient multiplied by the reactor volume at \(R_D = 0.135, \phi_L = 3 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1},\) and \(\phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}\). At rotational disc speeds below 70 rad s\(^{-1}\), the mass transfer at 1 mm rotor-stator distance is twice as high as at 2 mm; above 70 rad s\(^{-1}\) the mass transfer rates are equal. The mass transfer at 5 mm rotor-stator distance is lower in the whole range of rotational disc speeds.

The volumetric mass transfer coefficient increases with decreasing rotor-stator distance, as is shown in Figure 4.12a. The product of the volumetric mass transfer coefficient and the reactor volume is shown in Figure 4.12b; this quantity represents the overall mass transfer rate, divided by the driving force. \(k_{GL}a_{GL}V_R\) is a factor 2 higher at 1 mm rotor-stator distance than at 2 mm for rotational disc speeds below 70 rad s\(^{-1}\). In this flow regime all the gas is present as flat gas bubbles (see Figure 4.7), where the main gas-liquid mass transfer will take place from the gas bubble to the liquid film on the rotor and the stator. The gas-liquid interfacial area per unit reactor volume of this liquid film will approximately double when
Effect of rotor-stator distance and rotor radius on the rate of gas-liquid mass transfer

Figure 4.13: Gas bubbles at 1, 2 and 5 mm rotor-stator distance, at $\phi_G = 1.5 \cdot 10^{-6} \text{ m}^3 \text{s}^{-1}$, $\phi_L = 3 \cdot 10^{-5} \text{ m}^3 \text{s}^{-1}$ and $\omega = 26 \text{ rad s}^{-1}$. Liquid ripples are observed at 2 mm, in contrary to the situation with 1 mm rotor-stator distance. At 5 mm rotor-stator distance, the gas bubbles are not squeezed in between the rotor and stator. The position of the pictures in the reactor is indicated in Figure 4.5.

the rotor-stator distance decreases from 2 mm to 1 mm. The liquid film at 2 mm rotor-stator distance however, shows a different behaviour than at 1 mm, as can be seen in Figure 4.13. Ripples are observed in the liquid film, which are not present at 1 mm. No effect of this on the mass transfer rate is observed, however, only the decrease by a factor of 2, due to the factor 2 decrease in gas-liquid interfacial area. The gas-liquid mass transfer at 5 mm rotor-stator distance is significantly lower than at 1 and 2 mm. The large gas bubbles, at low rotational disc speeds, do fill a large part of the whole reactor height at 5 mm, but no film layer at the stator is present, as can be seen in Figure 4.13. The rate of mass transfer is therefore less than a factor of 5 lower than at 1 mm rotor-stator distance.

Surprisingly, the product of the volumetric mass transfer coefficient and the reactor volume at rotational disc speeds higher than 70 rad s$^{-1}$, is approximately equal for 1 and 2 mm rotor-stator distance. The total rate of mass transfer is independent of the rotor-stator distance in this region, which is unexpected. The bubbles are smaller than the rotor-stator distance in both cases, Equation 4.11 is therefore applied to get an estimate of the gas holdup ratio between the experiments at 1 and 2 mm rotor-stator distance. The bubbles are assumed to be spherical and uniform in size, the gas-liquid interfacial area is then:

$$a_{GL} = \frac{6 \varepsilon_G}{d_b}$$  \hspace{1cm} (4.19)

Combined with Equation 4.11, this leads to the following relation for the volumetric mass transfer coefficient:
The product of the reactor volume and the volumetric mass transfer coefficient for 1 and 2 mm rotor-stator distance is equal:

\[ [k_{GL}a_{GL}V_R]_{h=1} = [k_{GL}a_{GL}V_R]_{h=2} \] (4.21)

The ratio of the gas holdups is therefore:

\[
\frac{[\varepsilon_G]_{h=1}}{[\varepsilon_G]_{h=2}} = \left( \frac{h^{\frac{11}{2}} d_b}{h^{\frac{11}{2}} d_b} \right)_{h=1} \left( \frac{V_R^{3/4}}{V_R^{3/4}} \right)_{h=2}
\] (4.22)

The ratio of reactor volumes is known, the ratio of bubble diameters can be estimated from the images of the gas-liquid flow (e.g. Figure 4.7). At 157 rad s\(^{-1}\), the average gas bubble diameter at 1 and 2 mm rotor-stator distance is 0.6 and 0.9 mm, respectively. This leads to:

\[
\frac{[\varepsilon_G]_{h=1}}{[\varepsilon_G]_{h=2}} = \frac{0.6}{0.9} \cdot 0.97 \cdot 1.55 = 1.0
\] (4.23)

The gas holdup is thus equal for the two rotor-stator distances, in spite of the change in reactor volume, which means that the total amount of gas in the case of 2 mm rotor-stator distance is 1.8 times higher than for 1 mm. This is not expected based on the Wallis drift flux model, as shown in Figure 4.14, where the difference in gas holdup is more than a factor 2. As mentioned in the Theory section, the Wallis drift flux model is not valid in the case of small gas bubbles in the boundary layers, since the residence time of the gas bubbles is strongly dependent on the axial position. Gas bubbles in the rotor boundary layer have a lower inward radial velocity, while gas bubbles in the stator boundary layer have an increased radial velocity due to the centripetal boundary layer. An increase in liquid flow rate will result in a thinner rotor boundary layer with a lower radial velocity and a thicker stator boundary layer with a higher radial velocity. This will decrease the residence time, and thus the gas holdup, for both the gas bubbles in the rotor boundary layer and the stator boundary layer. The area perpendicular to the liquid flow is proportional to the rotor-stator distance; the influence of the liquid flow rate will thus be higher for 1 mm rotor-stator distance than for 2 mm rotor-stator distance. The increase in thickness and velocity of the stator boundary layer and the decrease in thickness and velocity of the rotor boundary layer is apparently large enough to decrease the gas holdup for 1 mm rotor-stator distance, and thus the gas-liquid mass transfer, to the value obtained at a rotor-stator distance of 2 mm.
Effect of rotor-stator distance and rotor radius on the rate of gas-liquid mass transfer

Figure 4.14: The gas holdup estimated using the Wallis drift flux model (Wallis, 1969), combined with the slip velocity relation of Richardson and Zaki (1954), Wallis (1969) and Equation 4.6, at \( \omega = 157 \text{ rad s}^{-1} \) and \( \phi_G = 1.5 \cdot 10^{-5} \text{ m}^3 \text{s}^{-1} \). The gas bubble radius used is taken from images of the gas bubbles, and is respectively 0.3 mm and 0.45 mm for 1 and 2 mm rotor-stator distance.

In the case of a rotor-stator distance of 5 mm, at rotational disc speeds above 100 rad s\(^{-1}\), the gas bubbles are approximately a factor 2 to 3 larger than at 1 mm rotor-stator distance, which results in a lower gas-liquid interfacial area, and thus in a lower mass transfer rate. The difference in gas bubble size is due to the change in flow regime, when increasing the rotor-stator distance from 2 to 5 mm. In the case of 5 mm rotor-stator distance two boundary layers are formed, one at the rotor and one at the stator, separated by a region with a constant tangential velocity, as shown in the top part of Figure 4.2. These boundary layers are merged at 1 and 2 mm rotor stator distance, as shown in the bottom part of Figure 4.2. The shear force, causing the break off of the gas bubbles at the gas inlet, is thus much lower for 5 mm rotor-stator distance than for 1 and 2 mm rotor-stator distance, resulting in larger gas bubbles, and thus a lower gas-liquid interfacial area and volumetric mass transfer coefficient.

Figure 4.15 shows the volumetric gas-liquid mass transfer coefficient as a function of the rate of energy dissipation in the system. The values for 1 and 2 mm rotor-stator distance are on the same line, the energy dissipation thus determines the mass transfer rate. Higher mass transfer rates can be reached with a rotor-stator distance of 1 mm, though, since the energy dissipation rate per unit volume is higher at the same rotational disc speed. The values of \( k_{GL}a_{GL} \) in the small bubble region are fitted with a power law model, with the energy dissipation rate as independent variable, resulting in an exponent of 0.75 ± 0.08. The surface
The gas-liquid interfacial area is, in this regime, thus a function of the rotational disc speed to the power 1.4 ± 0.2. The gas-liquid interfacial area is a combination of the gas holdup and the gas bubble size (see Equation 4.19). Meeuwse et al. (2010b) showed that, for the disc with 0.066 m radius, the gas holdup increases with increasing rotational disc speed. The gas-liquid interfacial area increases more rapidly, due to the decreasing gas bubble size. The same thus holds for the rotor-stator with a disc of 0.135 m radius. The gas-liquid mass transfer per unit of energy dissipation at 5 mm rotor-stator distance is much lower. This decrease is probably due to the significantly lower gas-liquid interfacial area, which is a result

\[
k_{GL}a_{GL} \propto E_d^{0.75}, \quad k_{GL} \propto E_d^{1.4} \Rightarrow
\]

\[
a_{GL} \propto E_d^{0.5}, \quad E_d \propto \frac{\omega^{3.4} h^{-\frac{1}{8}}}{V_R} \Rightarrow
\]

\[
a_{GL} \propto \frac{\omega^{1.4}}{h^{0.08} V_R^{0.5}} \quad (4.24)
\]
from the transition from the regime with a merged boundary layer to the regime with the separated boundary layers.

4.4.4 Rotor radius

Figure 4.16: Volumetric mass transfer coefficient at $h = 1$ mm, and $\phi_G = 7.3 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$.

$\phi_L = 3 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$ at $R_D = 0.135$ m, and $\phi_L = 6.7 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$ at $R_D = 0.066$ m. At a rotational disc speed below 100 rad $\text{s}^{-1}$, the volumetric mass transfer coefficient for the 0.135 m disc is smaller than for the 0.066 m disc, at higher rotational disc speeds it is higher.

The volumetric gas-liquid mass transfer coefficient as a function of rotational disc speed, for two rotor diameters, is shown in Figure 4.16. At rotational disc speeds above 100 rad $\text{s}^{-1}$, the volumetric gas-liquid mass transfer coefficient for the disc with a radius of 0.135 m is higher than for the 0.066 m disc. Below 100 rad $\text{s}^{-1}$, however, the volumetric gas-liquid mass transfer coefficient for the 0.066 m disc is higher. The reactor volume differs by a factor 3.4, therefore the overall mass transfer, $k_{GL} \phi_{GL} V_R$, is at 80 rad $\text{s}^{-1}$ larger for $R_D = 0.135$ m. At 52 rad $\text{s}^{-1}$, both have the same value, which is unexpected, since the gas bubbles have a longer path to travel in the case of $R_D = 0.135$ m. The liquid flow rate does have an influence, since it is more than four times higher at $R_D = 0.135$ m, leading to a decrease in gas holdup and thus in gas-liquid interfacial area. Additionally, smaller gas bubbles are formed at $R_D = 0.066$ m; this decreases the contact time of the gas with the liquid film on the rotor and the stator, thereby increasing $k_{GL}$.

Figure 4.17 shows the gas-liquid mass transfer coefficient as a function of the rate energy dissipation, for the two disc radii, and the different rotor-stator distances used. The gas-liquid
mass transfer, at equal energy dissipation rate, is higher for the disc with $R_D = 0.066$ m than for the reactor with $R_D = 0.135$ m. The rotational disc speed is much larger with the small disc at the same energy dissipation rate, therefore the small disc system is filled with small bubbles with surface renewal due to turbulent eddies, while the large disc system has the large gas bubbles with the mass transfer occurring in the liquid film at the rotor and at the stator. The energy dissipation rate in the large disc system can be increased tremendously, up to 6 MW $m^{-3}$, compared to 0.4 MW $m^{-3}$ in the small disc system. This 15-fold increase in energy dissipation rate leads to a 3-fold ($0.43 m^3_L m^{-3}_R$ s$^{-1}$ with $R_D = 0.066$ m to $1.36 m^3_L m^{-3}_R$ s$^{-1}$ with $R_D = 0.135$ m) increase in gas-liquid mass transfer rate. The mass transfer per unit of energy dissipation $\left( \frac{k GLaGL}{Ed} \right)$ for the disc with a radius of 0.066 m is $1.1 m^3_L MJ^{-1}$, while it is $0.2 m^3_L MJ^{-1}$ for the 0.135 m disc at the same gas flow rate.

The choice of the disc radius in an industrial scale spinning disc reactor depends on the process itself. When high mass transfer coefficients are needed, e.g. to obtain a higher selectivity, or to decrease the reactor volume for safety purposes, it is beneficial to use a larger disc size. However, the energy dissipation rate, and thus the operational costs, are higher than in the case of a larger number of smaller discs. For processes where the energy costs during operation play an important role in the overall cost of the process, the use of smaller discs is preferred.
4.5 Conclusions

4.5.1 Liquid flow rate

The volumetric mass transfer coefficient decreases with increasing liquid flow rate. A model for the gas holdup, based on the Wallis drift flux, predicts a decrease in gas holdup, and thus in mass transfer, but the effect observed experimentally is much larger. The model does not incorporate the complex flow pattern in the two-phase rotor-stator spinning disc reactor. More has to be known about the local velocities to accurately describe the effects of the liquid flow rate on the gas-liquid mass transfer in the spinning disc reactor.

4.5.2 Rotor-stator distance

The volumetric gas-liquid mass transfer coefficient in the spinning disc reactor increases with increasing rotational disc speed, increasing gas flow rate, and decreasing liquid flow rate. At rotational disc speeds below 70 rad s\(^{-1}\), the total gas-liquid mass transfer with 1 mm rotor-stator distance is twice as high as for 2 mm rotor-stator distance, since the mass transfer mainly occurs at the liquid film between large bubbles and the rotor/stator. At rotational disc speeds above 100 rad s\(^{-1}\), the gas is dispersed as small spherical bubbles. The total gas-liquid mass transfer for 1 and 2 mm rotor-stator distance is equal in this case, for 5 mm it is much lower. The volumetric gas-liquid mass transfer coefficient as a function of energy dissipation rate is also equal for 1 and 2 mm rotor-stator distance. At 1 mm, however, higher values of the energy dissipation rate, and thus the volumetric gas-liquid mass coefficient, can be obtained.

4.5.3 Rotor radius

The volumetric gas-liquid mass transfer coefficient of the disc with 0.135 m radius is lower than with the 0.066 m disc below 100 rad s\(^{-1}\). Above this rotational disc speed it is significantly higher. The gas-liquid mass transfer per unit of energy dissipation (\(\frac{k_{GL,agi}}{E_d}\)) is around 0.2 m\(^3\) MJ\(^{-1}\) at 0.135 m radius, while it is 1.1 m\(^3\) MJ\(^{-1}\) at a radius of 0.066 m. The maximum volumetric gas-liquid mass transfer coefficient obtained with the 0.135 m disc is 2.5 m\(^3\) m\(^{-3}\) s\(^{-1}\), at \(\phi_L = 1.5 \cdot 10^{-5}\) m\(^3\) s\(^{-1}\), and \(\phi_G = 1.5 \cdot 10^{-5}\) m\(^3\) s\(^{-1}\) and a rotational disc speed of 209 rad s\(^{-1}\). The maximum volumetric mass transfer coefficient thus increases with increasing disc radius, but from energetic point of view scaling up by increasing the number of rotors is preferred above scaling up in rotor size.
Bibliography


Richardson, J. F., Zaki, W. N., 1954. The sedimentation of a suspension of uniform spheres
Effect of rotor-stator distance and rotor radius on the rate of gas-liquid mass transfer under conditions of viscous flow. Chemical Engineering Science 3 (2), 65–73.
Liquid-solid mass transfer and reaction in a rotor-stator spinning disc reactor

Abstract

The heterogeneously catalyzed oxidation of glucose is performed in a rotor-stator spinning disc reactor. One side of the rotor is coated with a Pt/C and Nafion® catalytic layer, resulting in a liquid-solid interfacial area of $274 \text{ m}^2 \text{ m}^{-3}$. At the lowest rotational disc speed, 26 rad s$^{-1}$, the reaction is liquid-solid mass transfer limited; at the highest rotational disc speed, 180 rad s$^{-1}$, the intrinsic kinetics are rate determining. The experimental overall reaction rates are fitted with a resistances in series model, with the activation energy, pre-exponential factor and volumetric liquid-solid mass transfer coefficient as parameters. The volumetric liquid-solid mass transfer coefficient, $k_{LS}a_{LS}$, increases from 0.02 to 0.22 $\text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$ for a rotational disc speed of 26 to 180 rad s$^{-1}$. These values are high in comparison to conventional reactors, like packed beds, in spite of the low liquid-solid interfacial area used in this study. The values of the liquid-solid mass transfer coefficient $k_{LS}$ are one order of magnitude higher compared to values reported for packed beds. The Sherwood number for the liquid-solid mass transfer in the rotor-stator spinning disc reactor depends on the Reynolds number to the power 2 in the range $1 \cdot 10^5 < Re < 7 \cdot 10^5$. In this range the transition of laminar flow to turbulent flow takes place, resulting in a change of the mass transfer mechanism.
5.1 Introduction

The chemical industry is continuously searching for smaller, safer and more efficient reactors. Gas-liquid mass transfer and liquid-solid mass transfer are often the rate limiting steps in multiphase processes, e.g. heterogeneously catalyzed reactions. Multiphase reactors with increased mass transfer rates can therefore be smaller, and thus safer, and more cost efficient. Intensified processes, with higher mass transfer rates, can also lead to lower energy costs and capital costs, due to the smaller equipment.

The rate of mass transfer in a reactor is determined by the mass transfer coefficient, $k$, the interphase interfacial area, $a$, and the driving force in concentration. The mass transfer coefficient can be determined by the diffusion of the solute, or by the surface renewal rate of the fluid near the interface. In process intensification in general, the surface renewal rate is increased by agitating the fluid. This can be done by stirring, but also by flowing the liquid with a high velocity through the reactor. In the rotor-stator spinning disc reactor used in this work, the high velocity of the rotor with a narrow rotor-stator gap induces intense turbulence, and therefore leads to high mass transfer coefficients.

High volumetric gas-liquid mass transfer coefficients in the rotor-stator spinning disc reactor have been presented in previous papers (Meeuwse et al., 2010a,b). The high shear forces between the rotor and the stator create small gas bubbles with a high specific interfacial area. This, combined with the high gas-liquid mass transfer coefficients, gives the good gas-liquid mass transfer performance.

The liquid-solid mass transfer, which is the mass transfer from the bulk of the liquid towards the catalytic sites, plays an important role in heterogeneously catalyzed systems. The catalyst in a reactor can either freely move with the fluid, e.g. in a slurry bubble column, or it can be fixed, with the fluid flowing around, e.g. in packed beds or solid foams. The liquid velocity, and thus the flow rate, determines the liquid-solid mass transfer in packed beds. An increased flow rate therefore increases the liquid-solid mass transfer, but also decreases the residence time; a larger reactor volume is then required. The values of the liquid-solid mass transfer coefficient in the rotor-stator spinning disc reactor as presented in this paper are independent of the residence time in this reactor.

In this study the liquid-solid mass transfer is determined with a liquid phase only. However, the gas holdup in the spinning disc reactor is relatively low, typically up to 2 volume percent (Meeuwse et al., 2010a); the influence of a gas phase on the liquid-solid mass trans-
fer is therefore expected to be low. The values of the liquid-solid mass transfer coefficients presented in this paper are therefore also considered to be valid in gas-liquid-solid reactions, e.g. heterogeneously catalyzed reactions.

The liquid-solid mass transfer is determined from the rate of the oxidation of glucose in the rotor-stator spinning disc reactor. A layer of Platinum on carbon with Nafion® is applied to the rotor as a catalyst. The rate of the reaction is determined by the intrinsic kinetics and the liquid-solid mass transfer to the catalyst present on the rotor. The reaction rates are used to quantify the liquid-solid mass transfer coefficients in the spinning disc reactor as a function of rotational disc speed.

5.1.1 Liquid-solid mass transfer and reaction

The overall reaction rate of a heterogeneously catalyzed reaction can be described as resistances in series of the mass transfer steps and the intrinsic kinetics. The overall reaction rate coefficient for a first order reaction with liquid-solid mass transfer is as follows:

\[
K_{ov} = \left( \frac{1}{k_{LS}\alpha_{LS}} + \frac{1}{\eta k_0 e^{-\frac{E_{act}}{RT}}} \right)^{-1}
\]  

(5.1)

The first term describes the liquid-solid mass transfer, the second term the intrinsic kinetics together with diffusion limitation in the catalyst support layer, expressed by the effectiveness factor. The effectiveness factor is defined as the observed reaction rate divided by the reaction rate if the whole catalyst volume would have the same concentration as at the surface. For a flat plate geometry and first order reaction it is defined as:

\[
\eta = \frac{\tanh(\Phi)}{\Phi}
\]

(5.2)

with the Thiele modulus:

\[
\Phi = \delta \sqrt{\frac{k_0 e^{-\frac{E_{act}}{RT}}}{D_{eff}}}
\]

(5.3)

The pre-exponential factor in the kinetic term also contains the catalyst loading:

\[
k_0 = k_A \frac{N_{Pt}}{V_R}
\]

(5.4)

The experimental data are fitted to Equation 5.1, with \( k_0, E_{act} \) and \( k_{LS}\alpha_{LS} \) as parameters to be fitted. The kinetic part is strongly temperature dependent, the mass transfer term is only slightly temperature dependent. A change in temperature will therefore predominantly
affect the kinetic part. The values of $k_{LSaLS}$ are determined for each rotational disc speed separately. The effect of the mass transfer and of the intrinsic kinetics can then be quantified for the rotational disc speeds and temperatures used in this study.

## 5.2 Experimental

### 5.2.1 Preparation of catalytically active disc

A catalytically active rotor is prepared, to study the reaction and the liquid-solid mass transfer in the spinning disc reactor. A Nafion® coating, with Platinum on carbon particles is used for this purpose. Platinum on carbon (8.7 wt%, determined by inductively coupled plasma spectroscopy (ICP)) is prepared using hexachloroplatinic acid and Norit active carbon CA1 (particle size $D_{50} = 30 \mu m$) (Auer et al., 1999). The Pt/C is added to a Nafion® in propane-1,2-diol solution (De Heer and De Bruijn, 2004). The content of Pt/C, Nafion® and propane-1,2-diol in the coating was varied, to get a coating which is easy to apply and which adheres strongly to the surface, resulting in an ink with 5.4 wt% Nafion®, 18 wt% Pt/C and 77 wt% propane-1,2-diol. The ink was applied with a pipette to the aluminum rotor, which gave a homogeneous and uniform coating. The coating stayed on the rotating disc without an observable loss of catalyst or coating for several months of experiments, and thus could resist the large shear forces in the spinning disc reactor. The catalyst activity did not change in this period. The Nafion® glues the Pt/C particles together, resulting in an open structure, as can been seen in the scanning electron microscope (SEM) image in Figure 5.1, leading to accessible catalytic sites. The results of energy-dispersive X-ray spectroscopy (EDX) are shown in Figure 5.2. The particles that are visible in the SEM picture are the active carbon particles (Figure 5.2b). The particles are glued together with the Nafion®, which contains fluorine (Figure 5.2c). The platinum particles are dispersed in the whole layer, as shown in Figure 5.2d.

### 5.2.2 Glucose oxidation

The reaction used in this study needs to be a fast reaction, to be able to determine the liquid-solid mass transfer coefficient. The housing of the spinning disc reactor is made of PMMA, an aqueous environment is therefore desired. The oxidation of glucose to gluconic acid over the Pt/C catalyst deposited on the rotating disc is chosen:

$$\text{Glucose} + \frac{1}{2} \text{O}_2 \rightarrow \text{Gluconic acid}$$

The kinetics of this reaction are quite complex (Besson et al., 1996; Dam et al., 1987; Gangwal et al., 2004; Wilt and Baan, 1972). The conditions selected in this study result
Figure 5.1: SEM image of the surface of the catalytically active disc. The structures shown on the image are the carbon particles, glued together by the Nafion® polymer.

Figure 5.2: EDX results for the catalytic layer. a) Original SEM picture b) Carbon; the large particles mainly consist of carbon. c) Fluorine; the main content of Nafion®, which acts as glue. d) Platinum; the platinum is dispersed in the whole layer.
in pseudo first order behaviour in oxygen. Glucose is present in large excess in the reactor (1 kmol m\(^{-3}\)) and its concentration does not change significantly during the experiment; equation (5.1) can thus be used. The pH influences the reaction rate, and is controlled at pH 8 with a 0.1 kmol m\(^{-3}\) NaOH solution. The overall reaction rate coefficient can be calculated from the measured oxygen concentrations at the inlet and the outlet of the reactor as follows:

\[
K_{ov} = \frac{\phi L}{V_R} \left( \frac{C_{in}^{O_2}}{C_{out}^{O_2}} - 1 \right)
\]

This equation is based on an ideally mixed volume. This assumption is based on the high radial inward and outward liquid velocities present in a rotor-stator spinning disc reactor (Daily and Nece, 1960, Dijkstra and van Heijst, 1983, Owen and Rogers, 1989, Poncet et al., 2005), which results in a liquid recirculation time more than one order of magnitude smaller than the average residence time of the liquid in the reactor (Meeuwse et al., 2010a).

The oxygen concentration at the catalytic surface is known to have an inhibiting effect on the reaction rate (Dam et al., 1987). At high oxygen concentrations the catalyst surface can overoxidize, thus reducing the number of active sites for the reaction (Gangwal et al., 2004). The reaction is then less than first order in oxygen, or even of negative reaction order. Equation 5.1, and thus the approach used in this paper, would then not be valid anymore. The oxygen concentrations used in this study are therefore kept low, corresponding to 5 kPa of oxygen in the gas phase. The effect of overoxidation in time can still be observed, see Figure 5.3. Here, the overall reaction rate coefficient is shown for a series of measurements at 50 °C and 5 kPa oxygen saturation pressure. The first measurement is taken at 97 rad s\(^{-1}\). Every measurement is done for a period of three minutes; after that the rotational disc speed is increased. The measurement at 180 rad s\(^{-1}\) is directly repeated, followed by a decrease in rotational disc speed. The overall reaction rate coefficient increases with increasing rotational disc speed. However, it decreases faster in the second part of the series, where the rotational speed is decreased. The overall reaction rate thus decreases as a function of time, see Figure 5.4. In 4 minutes a decrease in activity of 10% can be observed. This effect is due to the overoxidation of the catalyst, which depends on the oxygen concentration at the solid interface. Catalyst deactivation is observed, even at the low oxygen concentrations. The reaction measurements are therefore kept short, to decrease the influence of the overoxidation, and a reduction step is introduced in the experimental sequence, to fully regenerate the catalyst. After each regeneration step the initial catalyst activity is again observed. The procedure used in this study is described in the next section.
Figure 5.3: Overall reaction rate coefficient at 50 °C and 5 kPa O₂, showing deactivation of the catalyst by overoxidation. The measurements start at 97 rad s⁻¹, the rotational speed is increased every 3 minutes; after reaching 180 rad s⁻¹, the rotational speed is decreased.

Figure 5.4: A typical graph of the measured overall reaction rate coefficient, at 180 rad s⁻¹ and 52 °C. Approximately 1 minute is needed to reach the steady state, after that the overall reaction rate coefficient decreases gradually due to overoxidation of the catalyst.
5.2.3 Experimental setup and procedure

A schematic drawing of the experimental setup used is shown in Figure 5.5. The rotor-stator spinning disc reactor consists of a rotating disc of 0.066 m in radius, enclosed in a cylindrical housing, with a rotor-stator distance of $1 \cdot 10^{-3}$ m. The volume of the reactor is $5 \cdot 10^{-5}$ m$^3$. The catalytic coating of Pt/C with Nafion$^\text{®}$ is only present on the bottom side of the rotating disc. The liquid-solid interfacial area $a_{LS}$ is therefore $274 \text{ m}^2 \text{ m}^{-3}$. The inlet of the reactor is in the top stator, near the rotating axis, the outlet is in the center of the bottom stator. The liquid is pumped through the reactor with a pump with a Bronkhorst Coriolis mass flow controller (M55), at $7 \cdot 10^{-6}$ m$^3$ s$^{-1}$. The oxygen concentration and the temperature are measured before and after the reactor. The reactor can be bypassed, while the measurements of the oxygen concentration and the temperature can still be performed. Two external vessels, with temperature control, are present in the setup. In one of the vessels nitrogen flows through, the other one is aerated by a flow of nitrogen with 5 vol% of oxygen.
The reactor forms a recycle loop with one of the vessels; two manual valves can be used to switch from the oxygen-rich to the oxygen-free liquid vessel.

The oxygen concentrations are measured with fiber optic oxygen sensors (Ocean Optics (FOXY-R) and Avantes (AVS-OXYPROBE-1.5)), at the inlet and at the outlet. The temperature is measured at the same position, using K-type thermocouples. The output signal of the oxygen sensor is strongly influenced by the local temperature. The oxygen sensors are calibrated at 0 and 5 kPa of oxygen saturation pressure, for a range of temperatures of a few degrees C, which is the range of temperatures for a measurement series. In this range the temperature dependence of the oxygen sensors is linear. The oxygen concentrations are corrected for the measured temperature at the position of the probe, which can vary during the experiments, especially after a switch of one of the valves. The concentrations with the oxygen-free liquid and oxygen-rich liquid reach the right concentration, of 0 and 1 respectively, within 2% absolute deviation, in every measurement sequence. The temperature at the outlet is taken as the temperature of the experiments, since this is equal to the reactor temperature for an ideally mixed system.

![Figure 5.6: The oxygen concentrations at the inlet (dashed line) and outlet (solid line) of the reactor during a measurement cycle. The graphs starts with oxygen-rich liquid through the bypass, which is afterwards switched to the reactor, which operates at 5 rad s$^{-1}$. The measurement is done at 180 rad s$^{-1}$, afterwards the catalyst is regenerated with oxygen free liquid.](image)

The oxygen concentrations during a typical measurement series are shown in Figure 5.6.
The measurement starts with the reactor bypassed, and the oxygen-rich glucose solution connected to the recycle loop; the measured oxygen concentrations reach the saturation concentration corresponding to 5 kPa, which is denoted as 1 in Figure 5.6. After 5 minutes this flow is switched towards the reactor, which was at oxygen free conditions, operating at 5 rad s\(^{-1}\). A sudden temperature drop occurs, resulting in a high measurement value for a few seconds, since the temperature exceeds the range of the linear temperature dependence of the concentration. The oxygen-free liquid, from the previous regeneration step, first has to leave the reactor, causing a drop in concentration at the outlet, which rapidly increases again, since the mass transfer, and thus oxygen consumption, is very low at 5 rad s\(^{-1}\). One minute after the switch to the reactor the desired rotational disc speed is set. After 50 seconds, which corresponds to 5 times the average residence time in the reactor, 99% of the steady state value is reached. After that the oxygen concentration slowly increases again, due to the gradual overoxidation of the catalyst. The values reported in this paper are the maximum overall reaction rate coefficients determined in this pseudo steady state period. After 5 minutes of measurement the oxygen-free liquid is switched to the reactor again, for the reduction step, where oxygen-free glucose solution is flushed through the spinning disc reactor, which is rotating at the maximum rotational disc speed of 180 rad s\(^{-1}\). This takes 6 minutes, after that the oxygen-rich liquid is pumped through the bypass again, and the next measurement can start.

5.3 Results and discussion

5.3.1 Mass transfer and reaction

The values of the overall reaction rate coefficient as a function of rotational disc speed are shown in Figure 5.7, for the investigated temperatures. \(K_{ov}\) increases a factor 4 to 6 by increasing the rotational disc speed from 26 to 180 rad s\(^{-1}\), up to 0.13 m\(^3\) L\(^{-3}\) m\(^{-3}\) s\(^{-1}\). The reaction rate is thus determined by the mass transfer from the liquid to the catalyst. Also a significant effect of temperature is observed, \(K_{ov}\) at 180 rad s\(^{-1}\) increases with a factor 2.5 when increasing the temperature from 38 °C to 52 °C. The reaction is therefore also determined by the intrinsic kinetics at the catalytic site.

The values of the overall reaction rate coefficient are fitted with Equations 5.1, 5.2, and (5.3), where \(k_0\) and \(E_{act}\) are the same for all measurements, and each rotational disc speed has its own \(k_{LS}a_{LS}\) value. It was found that \(k_0 = 1.1 \cdot 10^{11} m_L^3 m_R^{-3} s^{-1}\) and \(E_{act} = 73 \text{ kJ mol}^{-1}\) gave the best fit. The fitted value of the activation energy is close to the literature value of
Results and discussion

Figure 5.7: Overall reaction rate coefficient as a function of rotational disc speed. The reaction rate increases with increasing rotational disc speed and increasing temperature, indicating that both liquid-solid mass transfer and reaction play a role. The lines drawn are based on the fitting of the $k_{LSaLS}$ values and $k_0$ and $E_{act}$ in Equation 5.1.

77.4 kJ mol$^{-1}$ for the oxidation of glucose on platinum on carbon (Wilt and Baan, 1972). The effectiveness factor depends on temperature, and varies from 0.82 at 52 °C to 0.92 at 38 °C.

The values of the volumetric liquid-solid mass transfer coefficient are shown in Figure 5.8, together with the obtained values of $K_{ov}$. At low rotational disc speeds the values of $k_{LSaLS}$ are close to the values of $K_{ov}$, the kinetics therefore play only a minor role in this region. At higher rotational disc speeds the liquid-solid mass transfer coefficient is much higher than the experimental values of $K_{ov}$; the intrinsic kinetics are thus limiting the reaction rate under these circumstances.

In Figure 5.9 the mass transfer resistance and the resistance of the intrinsic kinetics are plotted. At low rotational disc speeds the mass transfer is mainly determining the reaction rate, the influence of the liquid-solid mass transfer decreases with increasing rotational disc speed. At a rotational disc speed of 180 rad s$^{-1}$ the kinetic resistance at 52 °C is a factor 2 higher than the mass transfer resistance, while it is a factor 7 higher at 38 °C. The accuracy of the $k_{LSaLS}$ values obtained thus decreases with increasing rotational disc speed. Especially the very high value of $k_{LSaLS}$ at 180 rad s$^{-1}$, 0.65 m$^3$ L m$^{-3}$ R s$^{-1}$, can have a relatively large error, since the impact on the value of $K_{ov}$ is small. This value of the liquid-mass transfer coefficient at 180 rad s$^{-1}$ is therefore not considered in the rest of the paper.
Figure 5.8: The volumetric liquid-solid mass transfer coefficient and the overall reaction rate coefficients as a function of rotational disc speed. The values are approximately equal at low rotational speeds, indicating that the reaction rate is mass transfer limited. The liquid-solid mass transfer coefficients at high rotational speeds are much higher than the overall reaction rate coefficient, indicating that here the intrinsic kinetics have a much larger influence.

Figure 5.9: The resistances to mass transfer and reaction as a function of rotational disc speed. The reaction is clearly mass transfer limited at low rotational disc speeds (typically below 60 rad s\(^{-1}\)); at high rotational disc speeds, typically larger than 100 rad s\(^{-1}\), the kinetics are the rate limiting factor, especially for the lower temperatures.
5.3.2 Comparison with conventional reactors

The liquid-solid mass transfer coefficient is calculated by the division of the fitted volumetric mass transfer coefficient \( k_{LSaLS} \) by the liquid-solid interfacial area \( a_{LS} \). The value of the liquid-solid mass transfer coefficient at low rotational speeds is comparable to a cocurrent downflow fixed bed column, which has reported \( k_{LS} \) values between \( 2 \cdot 10^{-6} \) and \( 2 \cdot 10^{-4} \) m\(^3\)L m\(^{-2}\) i s\(^{-1}\) (Shah, 1979). The liquid-solid mass transfer coefficient in the spinning disc reactor at 157 rad s\(^{-1}\) of \( 7.9 \cdot 10^{-4} \) m\(^3\)L m\(^{-2}\) i s\(^{-1}\), is an order of magnitude higher than in this type of conventional reactor. In agitated systems liquid-solid mass transfer coefficients are reported up to \( 1 \cdot 10^{-3} \) m\(^3\)L m\(^{-2}\) i s\(^{-1}\) (Ramachandran and Chaudhari, 1983), the values obtained in the spinning disc reactor have the same order of magnitude and are known to increase even more with increasing rotational disc speed.

The highest value of \( k_{LSaLS} \) obtained in this study is \( 0.22 \) m\(^3\)L m\(^{-3}\) R\(^{-1}\) s\(^{-1}\). This is higher than the values obtained in packed columns, which range from \( 0.01 \) to \( 0.1 \) m\(^3\)L m\(^{-3}\) R\(^{-1}\) s\(^{-1}\) (Comiti et al., 2000, Delaunay et al., 1980, Shah, 1979). In agitated tanks volumetric liquid-solid mass transfer coefficients up to \( 0.5 \) m\(^3\)L m\(^{-3}\) R\(^{-1}\) s\(^{-1}\) are reported (Ramachandran and Chaudhari, 1983, Trambouze et al., 1988). The liquid-solid interfacial area in this study in the spinning disc reactor, of 274 m\(^2\) m\(^{-3}\) R\(^{-1}\) is low in comparison to the aforementioned conventional reactors. This area, however, can easily be increased by a factor 2, by coating both sides of the rotating disc. The stators can also be coated with a catalytic layer; the mass transfer coefficient will probably be lower than at the rotor, but in the same order of magnitude (Jansson and Ashworth, 1977). If the gap between the rim of the disc and the inner diameter of the stator is minimized, the liquid-solid interfacial area can thus be increased to 2000 m\(^2\) m\(^{-3}\) R\(^{-1}\), almost a factor 8 higher than in the current setup. \( k_{LSaLS} \) will increase correspondingly, leading to volumetric gas-liquid mass transfer coefficients up to \( 1.5 \) m\(^3\)L m\(^{-3}\) R\(^{-1}\) at 157 rad s\(^{-1}\).

The reaction rate in gas-liquid-solid reactions is also influenced by the gas-liquid mass transfer rate. The volumetric gas-liquid mass transfer coefficient, \( k_{GLaGL} \), measured in the rotor-stator spinning disc reactor using a single gas inlet in the bottom stator, ranges from 0.1 to 0.43 m\(^3\)L m\(^{-3}\) s\(^{-1}\) (Meeuwse et al., 2010a). This is the same order of magnitude as the volumetric liquid-solid mass transfer coefficient presented in this paper, which is not expected to be influenced significantly by the presence of gas, due to the low gas holdup. In case of a gas-liquid-solid reaction, Equation 5.1 would be extended with a gas-liquid mass transfer term. The overall mass transfer resistance would approximately be doubled, if oxygen would be supplied via the gas phase instead of the liquid phase. However, at the highest rotational disc speed, the reaction rate would still be mostly influenced by the intrinsic kinetics, especially
at the lowest temperature (38 °C) used. The rotor-stator spinning disc reactor is therefore clearly suitable to perform heterogeneously catalyzed reactions, with one of the components present in the gas phase.

5.3.3 Liquid-solid mass transfer correlation

Figure 5.10: $Sh$ as function of $Re$. The solid line is fitted to the experimental data. The dashed line is the theoretical Sherwood relation for a free rotating disc with laminar flow (Bird et al., 2002, Levich, 1962), which describes the data at low $Re$. The dashed-dotted line is the Sherwood correlation for turbulent flow for a free rotating disc (Daguenet, 1968).

The Sherwood number for liquid-solid mass transfer as a function of the Reynolds number is shown in Figure 5.10. The experimental data in Figure 5.10 are fitted with a power law model:

$$Sh = a Re^b + c$$

(5.6)

The fitted coefficients are: $a = 2 \cdot 10^{-8}$ ($4 \cdot 10^{-9} \leq a \leq 1 \cdot 10^{-7}$), $b = 2.02 \pm 0.13$ and $c = 8.7 \cdot 10^2 \pm 2.5 \cdot 10^2$. The exponent in $Re$ is approximately 2, which is high compared to other correlations, where the power is below 1 (Green and Perry, 2008). The mass transfer coefficient in turbulent systems is commonly related to the rate of energy dissipation, which is a measure for the velocities and the sizes of the turbulent eddies, and thus for the surface renewal rate near the disc surface. For isotropic turbulence the liquid solid-mass transfer coefficient is shown to be proportional to the rate of energy dissipation to the power 1/4 (Lamont and Scott, 1970). In empirical correlations this power, or a value close to it, is also
observed, e.g. in bubble columns (Sano et al., 1974) or packed beds (Comiti et al., 2000, Delaunay et al., 1980). In a rotor-stator system as used in this study, the energy dissipation $E_d$ depends as follows on $Re$ (Daily and Nece, 1960):

$$E_d \propto Re^{2.3}$$ (5.7)

Combining Equations 5.6 and 5.7 shows that the mass transfer coefficient is proportional to the rate of energy dissipation to the power 0.72, almost a factor 3 higher than the power of 1/4 in the case of isotropic turbulence. This can be explained by comparing with a free rotating disc, where a similar phenomenon was observed. Heat transfer in rotor-stator systems is known to have a similar dependency on rotational disc speed as systems with a rotating disc in a large medium, although the values obtained in rotor-stator systems are higher, and the transition from laminar to turbulent flow occurs at a different value of $Re$ (Boutarfa and Harmand, 2005, Owen and Rogers, 1989). In analogy, the same behaviour is expected with mass transfer. At $Re < 3 \cdot 10^5$, $Sh$ can be described with the theoretical relation for a rotating disc in an infinite medium with laminar flow, which is based on the axial flow of liquid towards the rotor induced by the rotation of the rotor (Bird et al., 2002, Levich, 1962):

$$Sh = 0.620Re^{1.2}Sc^{1.3}$$ (5.8)

For turbulent mass transfer towards a free rotating disc the exponent in $Re$ is commonly 0.9 (Daguenet, 1968, Ellison and Cornet, 1971, Levich, 1962). A widely used correlation is proposed by Daguenet (1968):

$$Sh = 0.00707Re^{0.9}Sc^{1.3}$$ (5.9)

Daguenet states that this correlation is valid for $Re > 2.7 \cdot 10^5$. However, Mohr and Newman (1976) show that a transition region from laminar to turbulent mass transfer is present, which can be described by the correlation:

$$Sh = (0.8910^5Re^{-\frac{1}{2}} + 9.710^{-15}Re^{3})Sc^{1/3}$$ (5.10)

Figure 5.11 shows the laminar and turbulent correlations, and the transition between the two situations. With increasing $Re$, the laminar flow first becomes unstable, only after increasing $Re$ further the flow becomes fully turbulent (Mohr and Newman, 1976). Besides, turbulence first occurs near the rim of the disc, the liquid flow at small disc radius is then still laminar (Hanna et al., 1988). In the transition regime the observed power in $Re$ is 3.0, which describes the transition from laminar to turbulent flow, and is thus not a fundamental property of the mass transfer mechanism, as the value of 0.5 in laminar flow, and the value of 0.9 in
Laminar
Turbulent

\[ Sh = 0.62 \text{Re}^{0.5} \text{Sc}^{1/3} \] (Laminar)
\[ Sh = 0.00707 \text{Re}^{0.9} \text{Sc}^{1/3} \] (Turbulent)
\[ Sh = (0.89 \times 10^5 \text{Re}^{-0.5} + 9.7 \times 10^{-15} \text{Re}^3) \text{Sc}^{1/3} \] (Transition)

Figure 5.11: \( Sh \) as function of \( Re \) for a rotating disc in a large medium. The dashed line is the theoretical Sherwood relation for a rotating disc in an infinite medium with laminar flow (Bird et al., 2002, Levich, 1962). The dashed-dotted line is the Sherwood correlation for turbulent flow for a free rotating disc (Daguenet, 1968). The solid line shows the correlation for the transition region between laminar and turbulent (Mohr and Newman, 1976). The correlations are also plotted outside the region of applicability, to show the transition more clearly.

The transition of laminar flow to turbulent flow is also present in rotor-stator systems, roughly in the range \( 10^5 < Re < 10^6 \) (Owen and Rogers, 1989). Howey et al. (2010) measured the heat transfer as a function of the radius in rotor-stator systems with gap ratios of \( G = \frac{h}{RD} = 0.01 \) and 0.02, comparable to \( G = 0.015 \) in this study. When increasing \( Re \) above \( 1 \cdot 10^5 \) the heat transfer coefficient increases rapidly near the rim of the disc, but not at smaller radii. The transition of laminar to turbulent flow first occurs at the rim of the disc; only at higher \( Re \) the complete rotor-stator cavity is in turbulent flow. Since the measurements in this study are in the transition regime, the fraction of turbulent behaviour in the rotor-stator cavity increases with increasing \( Re \). This is thus a similar effect as in the free disc situation.

The exponent in \( Re \) of 2.0 in this study can be attributed to the transition of laminar flow to turbulent flow in the rotor-stator spinning disc reactor. Experiments at higher rotational disc speed are needed to determine the dependency of \( Sh \) on \( Re \) in the fully turbulent regime, where the exponent in \( Re \) is expected to be lower. Equation 5.6 is thus not valid outside the range \( 1 \cdot 10^5 < Re < 7 \cdot 10^5 \). The free disc correlations for laminar and turbulent flow are also shown in Figure 5.10. For \( Re < 3 \cdot 10^5 \), the values of \( Sh \) in the spinning disc reactor coincide with the laminar flow near a free disc. The values at \( Re > 5 \cdot 10^5 \) are larger than in the free
disc situation, which is in agreement with literature (Boutarfa and Harmand, 2005, Owen and Rogers, 1989).

5.4 Conclusions

The oxidation of glucose is performed in a rotor-stator spinning disc reactor, with a Pt/C with Nafion® coating on one side of the rotating disc as catalyst. The reaction is limited by the liquid-solid mass transfer at low rotational disc speeds (typically lower than 60 rad s\(^{-1}\)). The liquid-solid mass transfer increases with increasing rotational disc speed; the intrinsic kinetics are the rate determining step at the highest rotational disc speeds (typically higher than 100 rad s\(^{-1}\)). The overall reaction rate coefficient is fitted with a resistances in series model; the fitted activation energy is close to the value reported in literature. The values of the volumetric liquid-solid mass transfer coefficient vary from 0.02 m\(^3\) L\(^{-1}\) m\(^{-3}\) R\(^{-1}\) s\(^{-1}\) at 26 rad s\(^{-1}\) to 0.22 m\(^3\) L\(^{-1}\) m\(^{-3}\) R\(^{-1}\) s\(^{-1}\) at 157 rad s\(^{-1}\), which is higher than in conventional reactors such as a packed bed. The liquid-solid interfacial area is relatively low, 274 m\(^2\) m\(^{-3}\), the maximum value of \(k_{LS}\) of \(7.9 \cdot 10^{-4} \) m\(^3\) L\(^{-1}\) m\(^{-2}\) s\(^{-1}\) is therefore one order of magnitude higher than in packed columns. The liquid-solid interfacial area in the rotor-stator spinning disc reactor can easily be increased, up to 2000 m\(^2\) m\(^{-3}\), leading to \(k_{LS}\) of one order or magnitude higher than in packed beds.

\(Sh\) for the liquid-solid mass transfer in the rotor-stator spinning disc reactor depends on \(Re\) to the power 2 in the range \(1 \cdot 10^5 < Re < 7 \cdot 10^5\). In this range the transition of laminar flow to turbulent flow takes place; the mass transfer mechanism therefore also changes in this region gradually from laminar to turbulent. The high power in \(Re\) is thus not a fundamental property of the mass transfer mechanism, but a result of the laminar to turbulent transition.

Bibliography


Multistage rotor-stator spinning disc reactor

This chapter has been published as:

Abstract

The scale up of a rotor-stator spinning disc reactor by stacking single stage rotor-stator units in series is demonstrated. The gas-liquid mass transfer per stage is equal to the mass transfer in a single stage spinning disc reactor. The pressure drop per stage increases with increasing rotational disc speed and liquid flow rate. The pressure drop is more than a factor 2 higher for gas-liquid flow than for liquid flow only, and is up to 0.64 bar at 459 rad s$^{-1}$. The high mass and heat transfer coefficients in the (multistage) rotor-stator spinning disc reactor make it especially suitable for reactions with dangerous reactants, highly exothermic reactions and reactions where selectivity issues can be solved by high mass transfer rates. Additionally, the multistage rotor-stator spinning disc reactor mimics plug flow behaviour, which is beneficial for most processes.
6.1 Introduction

The productivity of reactors for multiphase processes is often limited by the rate of the mass transfer steps, from the gas to the liquid and from the liquid to the solid (catalyst). A higher mass transfer rate can thus lead to a higher productivity, or to a smaller reactor with the same productivity. Additionally, the mass transfer rates can influence the selectivity of reactions.

The gas-liquid mass transfer coefficient \( (k_{GL}a_{GL}) \) is the product of the gas-liquid interfacial area \( (a_{GL}) \) and the gas-liquid mass transfer coefficient \( (k_{GL}) \). The former is commonly increased by increasing the surface area where liquid has to flow over, e.g. in a falling film reactor or in a (structured) packing, or by decreasing the gas bubble size, which is e.g. done by a stirrer. The gas-liquid mass transfer coefficient often depends on the energy dissipation rate, and can thus be increased by increasing the flow rate, e.g. in a packed bed reactor, or by dissipating energy into the reactor directly, e.g. by using a stirrer.

The rotor-stator spinning disc reactor is a new type of multiphase reactor, which shows high mass transfer rates compared to conventional equipment (Meeuwse et al., 2010a,b,c,d). The rotor-stator spinning disc reactor consists of a rotating disc, enclosed in a cylindrical housing. The distance between the rotor and the reactor wall is small, commonly 1 mm. A high velocity gradient is present in the gap between the rotor and the stator, which acts as a shear force which breaks up gas bubbles, leading to a high gas-liquid interfacial area. The high energy input in the system, due to the rotation of the disc, leads to small turbulent eddies, with a high velocity, that increase the gas-liquid and liquid-solid mass transfer coefficients. Two different configurations of the rotor-stator spinning disc reactor have been investigated. In the first case, gas is injected through an orifice in the bottom stator, near the rim of the rotor; liquid is injected from the top of the reactor (Meeuwse et al., 2010a,c). In the second case, gas and liquid are fed together to the spinning disc reactor (Meeuwse et al., 2010d).

Meeuwse et al. (2010c) describe the gas-liquid mass transfer in case of a single gas inlet in the bottom stator. The gas bubbles are sheared off at the gas inlet, due to the velocity gradient; the gas bubble size decreases with increasing rotational disc speed, leading to an increase in gas-liquid mass transfer (Meeuwse et al., 2010c). The volumetric gas-liquid mass transfer coefficient increases with increasing rotor radius, up to \( 2.5 \text{ m}^3 \text{L}^{-1} \text{m}_R^{-3} \text{s}^{-1} \), at a rotational disc speed of 209 rad s\(^{-1}\), using a rotor with 0.135 m radius and 1 mm rotor-stator distance (Meeuwse et al., 2010a). The energy dissipation rate, however, increases more than the mass transfer rate; scaling up by stacking multiple rotor-stator units in series is therefore, from energetic point of view, preferred over scaling up in rotor size. For scaling up by numbering
up the reactor configuration with the gas inlet in the bottom stator, however, a complicated gas (re)distribution system may be needed.

![Diagram of the rotor-stator spinning disc reactor](image)

**Figure 6.1:** Schematic drawing of the concept of the rotor-stator spinning disc reactor with co-feeding of gas and liquid (not on scale). Gas and liquid are added to the top of the reactor. The liquid flows as a thin film over the rotor; the gas phase is present above, this region is called the film flow region. Near the rim of the rotor, gas bubbles are sheared off. The region surrounding the rim of the rotor and the region between the rotor and the bottom stator, therefore has liquid as continuous phase, with small gas bubbles dispersed in it. This region is denoted as the dispersed flow region. A gas liquid dispersion leaves the reactor via the bottom outlet; it is expected that feeding this dispersion to the next stage will lead to the same flow behaviour.

In the other configuration used, gas and liquid are both fed to the top of the reactor (Meeuwse et al., 2010d), as shown schematically in Figure 6.1. On top of the rotor a liquid film will form, with the gas phase present between this liquid film and the top stator; this is called the film flow region. Small gas bubbles are sheared off near the rim of the rotor. The rest of the reactor, the dispersed flow region, is thus filled with small gas bubbles dispersed in the liquid. The volumetric mass transfer coefficient multiplied by the reactor volume \( k_{GL}\omega_{GL}V_R \) in this configuration is up to a factor 2 higher than in the case of the single gas inlet in the bottom stator. This is mainly because the whole reactor volume is used, contrary to the case with the gas inlet in the stator, where only the region between the rotor and the bottom stator contributes to the mass transfer. Near the center of the reactor the gas
bubbles coalesce to larger bubbles, a gas-liquid dispersion leaves the reactor via the exit in the bottom. If the gas-liquid dispersion leaving the dispersed flow region is added to a next stage, it is therefore expected that the same flow regime, i.e. the film flow and the dispersed flow, is obtained. Scaling up this concept to a multistage spinning disc reactor is thus less complicated, since no gas redistribution system is needed.

The scale up of a reactor is an important step towards an industrial process. The reactor volume has to be increased, either to increase the residence in the reactor, or to increase the production capacity. In this study the rotor-stator spinning disc reactor is scaled up by stacking multiple (up to 3) rotor-stator stages in series, where the rotors are mounted on a common axis. It is expected that the same flow behaviour is obtained as in the single stage rotor-stator spinning disc reactor. A gas-liquid dispersion is fed to the reactor, which immediately separates in the film flow and the gas phase on top. At the exit of the reactor the gas bubbles coalesce to larger gas bubbles, and a similar gas-liquid dispersion is formed as which is fed. It is thus highly probable that the same behaviour will be present when fed to the next stage. The strategy to scale up by using rotor-stator stages in series is, for most reactions, preferred above scaling up in rotor size, since the mass transfer per unit of energy dissipation is higher. Additionally, in a multiple rotor-stator spinning disc reactor the liquid and the gas will mimic plug behaviour. If necessary, the stages in the multistage spinning disc reactor can operate under different reaction conditions, e.g. temperatures and catalysts, and it can thus be used for multi-step synthesis. This study shows that the scale up by stacking multiple rotor-stator stages in series is a viable method to increase the residence time and/or the production capacity in the rotor-stator spinning disc reactor.

This study describes the multistage rotor-stator spinning disc reactor, with 2 or 3 rotor-stator stages, where the gas and the liquid are fed together to the first stage. It is compared with the single stage unit, to investigate whether the same flow regime, with the film flow region and the dispersed flow region, is obtained. This is done by quantifying the mass transfer rate in the dispersed flow region, and comparing this to a single stage unit. Additionally, the energy dissipation rate and the pressure drop of this multistage rotor-stator spinning disc reactor are described. The last section summarizes the main advantages and disadvantages of the multistage spinning disc reactor and compares it with reactors commonly used in industry, and gives a few examples of potential applications.
6.2 Experimental

6.2.1 Experimental setup

Figure 6.2: Schematic drawing of the 3-stage multistage spinning disc reactor. The rotors are attached to an axis which has a maximum rotational speed of 470 rad s$^{-1}$. The radius of the rotors is 0.066 m, the inner reactor radius is 0.076 m. The axial rotor-stator distance is 1 mm. Gas and liquid are added to the reactor from the top, the gas-liquid dispersion leaves the reactor via the bottom. Cooling liquid (water in this study) flows through the hollow stators, countercurrently with the process fluids. The pressure differences as denoted in the drawing are measured using pressure sensors connected to the outer rim of the reactor.

Figure 6.2 shows a schematic representation of the multistage spinning disc reactor, which is made of stainless steel. It consists of an axis (radius 0.017 m) with 3 rotors attached to it, which has a maximum rotational disc speed of 470 rad s$^{-1}$. The radius of the rotors is 0.066 m, the inner reactor radius is 0.076 m, the radial distance between the rotor and the reactor wall is thus 0.01 m. The height of a stage is 8 mm, the thickness of the discs is 6 mm. The rotor-stator distance is thus 1 mm on both sides. The distance between the axis and the stator plates is 1 mm as well. The rotor size, the inner reactor radius and the rotor-stator distance are the same as in the single stage rotor-stator spinning disc reactor (Meeuwse et al., 2010c,d). Gas is injected at the top of the reactor, into a gas chamber. The liquid is injected by four channels (only two of them are shown in the drawing) onto the axis. The outlet of the reactor is on the bottom.
The motor which drives the axis uses up to 2.5 kW of energy, of which most will be converted into heat. To keep the reactor temperature approximately constant, the heat has to be removed from the reactor: cooling liquid (tap water) continuously flows through the hollow stators. The stators contain redistribution walls on the inside, to increase the liquid velocity, and thus the heat transfer coefficient, on the coolant side. The stator walls are 5 mm thick. The coolant is fed to the bottom stator, at the outlet it is fed to the next stator. The coolant thus flows countercurrently with the process fluids.

The motor used to rotate the rotor is a SEW Eurodrive CFM71M. The torque is proportional to the current applied to the motor, which is measured. The torque with the motor running idle, i.e. without rotors, is subtracted from the total torque. The energy dissipation rate per unit volume of reactor follows from the torque:

$$E_d = \frac{\tau \omega}{V_R} \quad (6.1)$$

The gas-liquid mass transfer rates in this study are measured by the desorption of oxygen from water. The gas (nitrogen) and liquid (water) flow rates are controlled by a Bronkhorst gas mass flow controller and a Coriflow, respectively. The oxygen concentration at the inlet liquid and at the outlet is measured with Ocean Optics FOXY-R fiber optic sensors and an Avantes spectrometer. The oxygen sensors are calibrated as a function of temperature, which is measured simultaneously and ranges from 15 to 30 °C. The gauge pressure (relative to atmospheric conditions) is measured in the inlet tube of the liquid, as well as at the rim of reactor stage 2. Additionally, the pressure difference between the rim of stage 1 and the rim of stage 3 is measured. The differential pressures as indicated in Figure 6.2 are calculated from these pressures:

$$\Delta P_{\text{stage}} = \frac{P_0}{3} = \frac{P_1 - P_3}{2} \quad (6.2)$$
$$\Delta P_{\text{top}} = P_2 - 2\Delta P_{\text{stage}} \quad (6.3)$$
$$\Delta P_{\text{bottom}} = \Delta P_{\text{top}} + \Delta P_{\text{stage}} \quad (6.4)$$

A liquid flow rate of $2.7 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$ is fed to the reactor from a 2 dm$^3$ stirred vessel. The vessel is aerated to increase the oxygen concentration. The outlet liquid is recycled to the vessel. The cooling liquid is tap water of approximately 15 °C. Nitrogen is used as the gas phase in the reactor, the flow rate is varied up to $3 \cdot 10^{-5} \text{ m}^3 \text{ s}^{-1}$ at normal flow conditions.
6.2.2 Gas-liquid mass transfer

Meeuwse et al. (2010d) described the gas-liquid mass transfer in a single stage spinning disc reactor, where gas and liquid are fed together to the bottom of the reactor. A liquid film is present on top of the rotor, the gas phase fills the rest of the region, see Figure 6.1. The liquid film is in plug flow behaviour, the gas phase is assumed to be ideally mixed, since a recirculation of the gas is induced by the radial and tangential velocity of the liquid. A Sherwood correlation for the gas-liquid mass transfer coefficient on a spinning disc with a liquid film present is used for the calculation of the gas-liquid mass transfer coefficient in the film flow region (Aoune and Ramshaw, 1999, Meeuwse et al., 2010d, Peev et al., 2007):

$$Sh = 10.8 \cdot 10^{-4} Re^{0.94} C_{w}^{0.24} Sc^{\frac{1}{2}}$$ (6.5)

The gas-liquid interfacial area is the top area of the rotor divided by the volume of the film flow region:

$$a_{GL} = \frac{A_{top}}{V_F} = \frac{\pi R_D^2}{h \pi R_D^2} = \frac{1}{h} = 1 \cdot 10^3 \text{m}^2 \text{m}^{-3} R$$ (6.6)

Small gas bubbles are sheared off from the gas phase in the film flow region, at the rim of the rotor. The region surrounding the rim of the rotor and the region between the rotor and the bottom stator is thus filled with gas bubbles dispersed in liquid, as shown schematically in Figure 6.1. The gas bubbles in the dispersed flow region flow radially inwards, due to the centrifugal force. No coalescence of gas bubbles is observed, and most of the gas bubbles have approximately the same residence time; the gas phase is thus assumed to be in plug flow. A recirculation of the liquid is induced by the rotation of the rotor (Dijkstra and van Heijst, 1983, Meeuwse et al., 2010d, Poncet et al., 2005a), the liquid phase in the dispersed flow region is therefore assumed to be ideally mixed.

A representation of the reactor model used for the combination of the film flow region and the dispersed flow region is shown in Figure 6.3, where the number of stages, N, is one. The oxygen concentrations are measured at the inlet and the outlet. The $k_{GLaGL}$ in the film flow region is calculated from Equation 6.5, the rest of the mass transfer thus occurs in the dispersed flow region. In this way the value of the volumetric gas-liquid mass transfer coefficient, $k_{GLaGL}$, is obtained.

The same flow behaviour is expected in the multistage spinning disc reactor ($N > 1$) as in the single stage version ($N = 1$). The gas and liquid which leave the first stage are again expected to form a liquid film on the rotor of the next stage, with the gas phase present above.
In the film flow region (see Figure 6.1), gas is ideally mixed and liquid is in plug flow, in the dispersed region the gas phase is in plug flow and the liquid phase is ideally mixed. The gas and the liquid from the dispersed region of the 1st stage flow to the film flow region of the next stage. The number of stages, $N$, is 1, 2 or 3 in this study. The oxygen concentration is measured in the liquid phase at the inlet and at the outlet.

The reactor model as presented in Figure 6.2 is thus valid again, but now the sequence is executed $N$ times. The concentrations in the film flow region are calculated using time dependent differential equation for the ideally mixed gas phase, coupled with a differential Equation for the liquid, which is in plug flow. The exit concentrations of the phase are the input concentrations of the dispersed flow region, where the differential equation of the ideally mixed liquid phase is coupled with the differential Equation of the gas phase. This procedure gives the output concentration as a function of time. The steady state value of the calculated concentration is fitted to the experimentally measured outlet concentration, with the value of $k_{GLaGL}$ in the dispersed flow region as fitting parameter.

### 6.3 Results and Discussion

#### 6.3.1 Gas-liquid mass transfer

Figure 6.4 shows the volumetric gas-liquid mass transfer coefficient in the dispersed flow region as a function of the rotational disc speed. Figure 6.4a shows the values with a 2-stage rotor-stator spinning disc reactor. The volumetric mass transfer increases with increasing rotational disc speed, which was also observed in the single stage unit (Meeuwse et al., 2010d).
An increase in gas flow rate leads to an increase in mass transfer as well. The same trends are observed in Figure 6.4b, for the 3-stage spinning disc reactor. At rotational disc speeds above 80 rad s$^{-1}$, however, the increase in mass transfer as a function of rotational disc speed seems to level off, and the mass transfer even decreases at higher rotational disc speeds. The leveling off and the decrease at higher rotational disc speeds is explained as follows. The gas-liquid mass transfer rates in the multistage spinning disc reactor are high, which means that equilibrium between the gas and the liquid will be reached at high rotational disc speeds. The measured oxygen concentration, however, is not exactly the same as the oxygen concentration in the gas phase calculated with the mass balance, but a little bit higher, due to a bias in the measurements. This slight overestimation of the oxygen concentration is probably due to the calibration method used, and has only a significant effect when the gas and the liquid are near equilibrium. The mass transfer rate is thus underestimated, and gets constant with increasing rotational disc speed. The value of the mass transfer in the film flow region, however, calculated with Equations 6.5 and 6.6 will increase with increasing rotational disc speed. This thus leads to a decrease in the volumetric gas-liquid mass transfer coefficient in the dispersed flow region. The measurements where the gas and the liquid concentrations are too close together are therefore discarded, if $(C_{out}^L - \frac{RT}{H} C_{out}^G) < 0.01$ mol m$^{-3}$.

The effect of the bias in the measurements is obviously larger if the number of stages increases. In the single stage spinning disc reactor the decrease of mass transfer coefficient was not observed, up to the maximum rotational disc speed of 180 rad s$^{-1}$ used (Meeuwse
et al., 2010d). The method used, the desorption of oxygen from water, is thus not suitable to measure the gas-liquid mass transfer at rotational disc speeds higher than 80 rad s$^{-1}$ for a 3-stage spinning disc reactor. The problem that the gas and the liquid reach equilibrium at higher rotational disc speeds can be overcome by using a (fast) reaction in the liquid bulk, which keeps the bulk concentration in the liquid low, preferably at a concentration of zero.

Figure 6.5: The volumetric gas-liquid mass transfer coefficient in the dispersed flow region, for a single stage, 2-stage and 3-stage rotor-stator spinning disc reactor, at a gas flow rate of $7.3 \cdot 10^{-6}$ m$^3$ s$^{-1}$.

Figure 6.5 shows the volumetric gas-liquid mass transfer coefficient in the dispersed flow region, for the single stage, 2-stage and 3-stage rotor-stator spinning disc reactor. The values of $k_{GL}a_{GL}$ are equal for the 2-stage and the 3 stage reactor below 80 rad s$^{-1}$. Above this value, the increase with the 3-stage reactor levels off, which is due to the measurement method, as explained before. The values obtained in the single disc reactor are approximately the same, and follow the same trend, although the steep increase in mass transfer as a function of rotational disc speed starts at a somewhat higher rotational disc speed. The values for the single stage spinning disc reactor are obtained in a different setup (Meeuwse et al., 2010d), since it was not possible to use one stage in the setup used in the current study. The setup used by Meeuwse et al. (2010d) was of similar dimensions, it only had a rotating disc with a thickness of 4 mm instead of 6 mm. The measurements were performed at the same gas flow rate, but at a lower liquid flow rate of $\phi_L = 6.7 \cdot 10^{-6}$ m$^3$ s$^{-1}$.

The fact that the obtained mass transfer coefficients are of the same magnitude as in the single disc system, is a strong indication that the gas-liquid flow (liquid film on rotor, gas
bubbles dispersed in the rest of the reactor) is the same, for 2 or 3 stages. Unfortunately, no measurements for the multistage system could be done above 110 rad s\(^{-1}\), due to the high mass transfer rates. At higher rotational disc speeds it can thus not be shown that the same flow configuration is obtained, but it is plausible that the behaviour will be the same. The co-fed rotor-stator spinning disc reactor can thus be scaled up by stacking multiple rotor-stator stages in series, where every stage has the same mass transfer performance as in a single stage reactor.

6.3.1.1 Energy dissipation rate

![Energy dissipation rate plots](image)

Figure 6.6: The energy dissipation rate in the multistage spinning disc reactor as function of the rotational disc speed. (a) The energy dissipation rate for the single stage, 2-stage and 3-stage reactor. (b) The energy dissipation rate for a 3 stage reactor with and without gas flow. The results of the correlation by Daily and Nece (1960) are shown as well.

Figure 6.6a shows the energy dissipation in the spinning disc reactor as a function of rotational disc speed, for the 1, 2 and 3-stage spinning disc reactor. The energy dissipation rate is approximately equal for the 1, 2 and 3-stage reactor, which is another indication that the flow is the same in all stages. The energy dissipation rate decreases with 20% in the presence of gas, as shown in Figure 6.6b, the gas flow rate does not have an influence. A decrease in energy dissipation rate is expected, since a significant part of the reactor is filled with gas, which has a lower density and viscosity, and thus less energy is needed to accelerate the fluid. Figure 6.6b also shows the correlation for the energy dissipation from Daily and Nece (1960). The values obtained in the single phase multistage rotor-stator spinning disc reactor are a factor 2.7 higher than those obtained from the correlation. This could be caused by the
higher liquid throughflow rates used in this paper. The ingoing liquid has to be accelerated to 
the velocity of the rotor, leading to a higher energy input. However, this is not experimentally 
proven, and no evidence of this is found in the literature.

6.3.1.2 Pressure

Figure 6.7: The pressure differences in the second stage of the 3-stage spinning disc reactor. 
The pressures differences are denoted in Figure 6.2 and are: the pressure increase from the 
axis on top of the rotor towards the rim of the reactor, the pressure decrease from the rim 
of the reactor to the axis below the rotor, and the pressure drop over the stage, which is the 
difference between the two. The values are for the middle stage of the 3-stage reactor, with 
a liquid flow rate of $2.7 \times 10^{-5}$ m$^3$ s$^{-1}$ (a) The pressures in the case of liquid only. (b) The 
pressures in case of liquid flow and a gas flow rate of $7.3 \times 10^{-6}$ m$^3$ s$^{-1}$.

Figure 6.7 shows the pressure differences in the second stage of the 3-stage spinning disc 
reactor, with a liquid phase only (a), and with gas and liquid together (b). The pressure at 
the rim of the reactor is always higher than near the axis, due to the centrifugal pressure. 
The pressure thus increases from the axis on top of the disc to the rim of the reactor ($\Delta P_{top}$), 
up to 1.0 bar at 459 rad s$^{-1}$. The pressure decrease between the rim of the reactor and the 
axis below the rotor ($\Delta P_{bottom}$), is larger, up to 1.3 bar at 459 rad s$^{-1}$. The difference is the 
pressure drop per stage ($\Delta P_{stage}$), which is thus up to 0.26 bar at 459 rad s$^{-1}$.

The pressure drop is not caused by the friction of the fluid, as would be the case in the 
flow between two flat plates. The centrifugal pressure plays a large role, as is seen by the
influence of the rotational disc speed. The liquid flow rate has a large influence as well. The pressure drop per stage in the case of a liquid flow rate which is a factor 4 lower ($\phi_L = 6.7 \cdot 10^{-6} \text{ m}^3 \text{ s}^{-1}$), is 0.024 bar at 459 rad s$^{-1}$, which is an order of magnitude lower. $\Delta P_{\text{top}} = 1.1 \text{ bar}$, at 459 rad s$^{-1}$, in this case, and $\Delta P_{\text{bottom}}$ is thus almost the same. If no liquid flow rate would be present, no pressure drop over a stage would be expected of course, since the buildup of centrifugal pressure is then the same above the rotor as below the rotor. In the case of a (large) liquid flow rate, the pressure buildup above the rotor will be lower, due to the centrifugal flow, while the pressure will decrease more below the rotor, where the flow is centripetal (Debuchy et al., 1998, 2008, Poncet et al., 2005b).

The region between the rotor and the top stator is mainly filled with gas in the case of co-feeding of gas and liquid. The liquid flows over the rotor as a liquid film. The region between the rotor and the bottom stator is filled with liquid, with small gas bubbles dispersed in it. The gas holdup in the bottom part will thus be much lower than in the top part. The centrifugal pressure strongly depends on the density of the phases, $\Delta P_{\text{top}}$ will thus be much smaller than $\Delta P_{\text{bottom}}$, as shown in Figure 6.7, with values of 0.33 bar and 0.97 bar at 459 rad s$^{-1}$. The resulting pressure drop per stage is thus 2.5 times higher than in the case with liquid only, with 0.64 bar at 459 rad s$^{-1}$.

Energy is needed to overcome the pressure drop in the reactor. This energy input also has to be taken into account in the total energy dissipation rate of the system. The energy needed to pressure the liquid, for a 3-stage reactor with a pressure drop of $1.9 \cdot 10^5 \text{ Pa}$, follows from the work applied:

$$E_{d,L} = \int_{P_{\text{atm}}}^{P_0} \frac{\phi_L dP}{V_R} = \frac{\phi_L (P_{\text{atm}} - P_0)}{V_R} = 2.7 \cdot 10^4 \text{ W m}^{-3}$$

(6.7)

The isentropic work for compression can be used for the gas phase, assuming the ideal gas law:

$$E_{d,G} = \int_{P_{\text{atm}}}^{P_0} \frac{\phi_G dP}{V_R} = \int_{P_{\text{atm}}}^{P_0} \frac{nRT dP}{P} = \frac{nRT \ln \frac{P_0}{P_{\text{atm}}}}{V_R} = \frac{P_{\text{atm}} \phi_G RT \ln \frac{P_0}{P_{\text{atm}}}}{V_R} = 1.7 \cdot 10^4 \text{ W m}^{-3}$$

(6.8)

The total energy dissipation rate resulting from the pressure drop is thus:

$$E_{d,\Delta P} = E_{d,L} + E_{d,G} = 4.4 \cdot 10^4 \text{ W m}^{-3}$$

(6.9)
This is two orders of magnitude lower than the energy dissipation rate due to the rotation of the rotor, and can therefore be neglected.

6.4 Reactor comparison

The rotor-stator spinning disc reactor has advantages and disadvantages, compared to conventional reactor equipment. The following sections describe the most important characteristics of the spinning disc reactor, and compares it to conventional reactors. The choice of a reactor for a certain process depends on the characteristics of the process; examples are presented of processes where the spinning disc is a suitable alternative for reactors commonly used in industry.

6.4.1 Mass transfer

The mass transfer coefficients presented in this paper for the multistage rotor-stator spinning disc reactor are only up to a rotational disc speed of 110 rad s$^{-1}$. It is shown that the single disc behaviour repeats itself at each stage, and that scale-up by stacking rotor-stator units is possible. The mass transfer coefficient in a single stage rotor-stator spinning disc reactor is measured up to a rotational disc speed of 180 rad s$^{-1}$. The volumetric mass transfer coefficient in the film flow region is up to $0.4 \text{ m}^3 \text{ m}_R^{-3} \text{s}^{-1}$, in the dispersed phase it is up to $0.95 \text{ m}_L^3 \text{ m}_R^{-3} \text{s}^{-1}$ (Meeuwse et al., 2010d). This is 3 to 7 times higher than in conventional reactors such as bubble columns or stirred tank reactors (Cents et al., 2005, Van der Schaaf et al., 2007). The same mass transfer behaviour is thus expected for the multistage spinning disc reactor. It is shown for the spinning disc reactor with a single gas inlet that the mass transfer coefficient can be increased even further by increasing the rotor size; an increase in rotor radius of a factor 2 leads to a 3-fold increase in mass transfer (Meeuwse et al., 2010a). Additionally, the liquid-solid mass transfer coefficient is, with a value of $8 \cdot 10^{-4} \text{ m}_L^3 \text{ m}_R^{-2} \text{s}^{-1}$ at a rotational disc speed of 157 rad s$^{-1}$, an order of magnitude higher than e.g. in a packed bed reactor (Shah, 1979). The liquid-solid interfacial area, which can be up to 2000 m$^2$ m$^{-3}$R is comparable or even higher than in this type of conventional reactor.

6.4.2 Heat transfer

From literature it is known that the heat transfer coefficients in a rotor-stator system like the one used in this study, are up to $2 \cdot 10^4 \text{ W m}^{-2} \text{K}^{-1}$ (Owen et al., 1974), where the heat transfer towards the rotor is commonly up to 50% higher than to the stator (Howey et al., 2010, Owen and Rogers, 1989), as described in the Appendix. This is up to an order of magnitude higher than in conventional reactors, such as stirred tanks, bubble columns, and
packed bed reactors. The interfacial area available for heat transfer is much higher than in conventional systems, if the rotor and the stator are used it can be up to $2000 \text{ m}^2 \text{ m}^{-3} \cdot \text{R}$. The overall heat transfer coefficient is therefore expected to be one or two orders of magnitude higher. The (multistage) rotor-stator spinning disc reactor is thus very promising for reactions where heat transfer plays a key role.

### 6.4.3 Energy requirements

Figure 6.8: Volumetric mass transfer coefficient as function of energy dissipation, for a variety of multiphase reactors. (Stemmet et al., 2007, Trambouze and Euzen, 2004) The values of $k_{GL}a_{GL}$ obtained in the spinning disc reactor are more than one order of magnitude higher than in conventional reactor systems, however, the energy input is around 3 orders of magnitude higher.

This paper shows the mass transfer coefficient in the multistage rotor-stator spinning disc reactor. The mass transfer rates obtained are high in comparison with conventional reactors. This is due to the high velocities, and the high degree of turbulence which is obtained in the rotor-stator spinning disc reactor. The energy input needed for the rotation of the rotors, together with the energy needed to overcome the pressure drop, influences the operation costs of this reactor. The energy dissipation rate in the spinning disc reactor is thus a factor that plays a significant role in the decision whether a spinning disc reactor will be chosen instead of a different type of reactor.

The maximum volumetric gas-liquid mass transfer coefficient obtained in the rotor-stator spinning disc reactor is approximately a factor 20 higher than the coefficient obtained for
stirred tank reactors (Meeuwse et al., 2010a, Tschentscher et al., 2010). The energy dissipation rate, however, is up to a factor 2000 higher than for this type of conventional equipment. The energy dissipation rate, however, is up to a factor 2000 higher than for this type of conventional equipment. The energy input is converted into heat, and can thus be used to heat up the reactants, or this heat can be used for other process steps.

The rate of gas-liquid mass transfer per unit of energy dissipation \( \frac{k_{GLaGL}}{E_d} \) is around 0.4 \( \text{m}^3 \text{L MJ}^{-1} \) for a rotor with 0.135 m radius (Meeuwse et al., 2010a) and 1.1 \( \text{m}^3 \text{L MJ}^{-1} \) for a rotor with 0.066 m radius, while it is around 80 \( \text{m}^3 \text{L MJ}^{-1} \) for a stirred tank reactor with a Rushton stirrer (Tschentscher et al., 2010). The mass transfer per unit of energy is thus much lower than for most conventional reactors (Stemmet et al., 2007). The mass transfer coefficient, however, is much higher than for conventional equipment, as is shown in Figure 6.8. A high mass transfer coefficient can have a large influence on the selectivity in the case of competitive or consecutive reactions. Mass transfer coefficients which are one order of magnitude higher, can give a new scope for reactions that were not economically feasible in conventional equipment, due to selectivity issues. The rotor-stator spinning disc reactor is thus mainly suited for this type of processes. Additionally, a large reduction in reactor size and volume can be achieved by using the rotor-stator spinning disc reactor, which can increase the safety of a process, due to the decreased inventory of (dangerous) chemicals. The smaller volume also makes it feasible to work at higher pressures, which opens a new window of reactor operation.

6.4.4 Reactor operation

A single rotor-stator spinning disc reactor consists of a part where the gas is ideally mixed and the liquid is in plug flow (the film flow region) and a part where the gas is in plug flow and the liquid ideally mixed (dispersed flow region). For all positive order reactions, and thus also all reactions which are limited by mass transfer, which behave as a first order reaction, plug flow behaviour is preferred. The ideally mixed regions in the spinning disc reactor are thus not beneficial. However, if the single stage reactor is expanded towards a multistage system, it behaves similarly to a cascade of mixers. If the number of stages is high enough, this mimics plug flow behaviour.

Most of the conventional reactor systems that are used in industry which behave more or less as plug flow (e.g. packings or trickle bed reactors) have a mass transfer coefficient which is directly linked to the velocity in the reactor. Increasing the velocity gives a higher mass transfer rate, but this also decreases the residence time in the reactor. A longer column is then
needed to get the same conversion. In a (multistage) rotor-stator spinning disc reactor the
mass transfer is determined by the rotation of the discs, and not by the flow rate of gas and
liquid in the reactor. An extra degree of freedom is therefore present which is unavailable in
most conventional systems.

6.4.5 Applications

The multistage rotor-stator spinning disc reactor requires more sophisticated equipment than
conventional reactor systems. This will result in higher equipment costs, and a higher risk
of mechanical failure, due to the rotating parts. Apparatuses with rotating parts are widely
used in industry (e.g. pumps or compressors), but they have higher costs for maintenance,
to prevent failure of the mechanical parts. Additionally, the energy requirements are higher
then for conventional equipment. The rotor-stator spinning disc reactor is therefore only eco-
nomically attractive if the high heat and mass transfer rates justify the high investment and
operational costs. This reactor is therefore probably not the best choice for large scale pro-
cesses. Examples of processes in which the rotor-stator spinning disc reactor does have a
large potential are given below.

An important advantage of the multistage rotor-stator spinning disc reactor is the high
mass transfer rate, which significantly increases the conversion, or decreases the reactor vol-
ume needed for a process. The smaller reactor volume thus has a lower amount of chemicals,
which is especially beneficial in the case of dangerous reactants and/or products, since the
risk of a spill or an explosion is decreased. For reactions at a higher pressure a smaller reactor
volume is also beneficial, since this decreases the outer surface area of the reactor. The gas
holdup in the reactor is relatively low in the dispersed region (typically a few percent), and
somewhat higher in the film flow region. For reactions with dangerous gases this increases
the safety even further. These aspects typically play a role in the production of pharmaceu-
ticals and in fine-chemistry (Chaudhari and Mills, 2004, Mills and Chaudhari, 1999), e.g.
with hydrogenation, oxidation and hydroformylation reactions. The energy dissipation rate,
which is high in the spinning disc reactor compared to conventional reactors, is not playing
an important role in the total costs of these processes, especially in pharmaceutical industry,
where time to market, selectivity and conversion are much more important than the operation
costs.

In parallel or serial reactions, the selectivity towards the desired product is an important
issue. It is important to have a good control over the reactor conditions, especially if the mass
transfer determines the selectivity. A (very) high mass transfer rate can have a positive influ-
ence on the selectivity; a (very) high energy dissipation rate is then not necessarily a problem. A higher selectivity can make a process economically feasible, e.g. since less separation effort is required.

Another potential application of the multistage rotor-stator spinning disc reactor is for highly exothermic reactions, since both the surface area and well as the heat transfer coefficient are high compared to other reactors. The formation of hot spots in the reactor is unlikely, since either the rotor or the stator is always close by. If the reaction is heterogeneously catalyzed and the catalyst is supported on the rotor and/or the stator, the heat can be removed locally, through the rotor and/or the stator.

6.5 Conclusions

The volumetric mass transfer coefficient in the dispersed flow region of a rotor-stator spinning disc reactor is the same for a single stage reactor, a 2-stage reactor and a 3-stage reactor. This proves the principle that the spinning disc reactor can be scaled up by using multiple rotor-stator units in series, where the rotors are mounted on a common axis. The gas-liquid mass transfer is measured up to a rotational disc speed of 100 rad s$^{-1}$, where the volumetric gas-liquid mass transfer coefficient is $0.7 \text{ m}^3 \text{ m}^{-3} \text{ s}^{-1}$. The mass transfer rates at higher rotational disc speeds could not be obtained, since the mass transfer is so high that the gas and liquid are virtually in equilibrium.

The pressure drop in the multistage spinning disc reactor increases with increasing liquid flow rate and rotational disc speed, up to 0.64 bar, at 459 rad s$^{-1}$. The pressure drop in the reactor is higher with the presence of gas and liquid then with liquid phase only.

The high mass and heat transfer rates, in combination with the high energy dissipation rate in the reactor, make the multistage rotor-stator spinning disc reactor mainly suitable for reactions with dangerous chemicals, reactions at high pressures, highly exothermic reactions and reactions where a significant increase in selectivity can be obtained due to the high mass transfer rates.

Appendix: Heat transfer

For the heat transfer in rotor-stator spinning disc reactors a distinction is made between the heat transfer towards the rotor and the heat transfer towards the stator. In general, the heat
transfer coefficient increases with increasing rotational disc speed and increasing radial position. For the use in the spinning disc reactor, the heat transfer coefficient averaged over the whole rotor or whole stator is used. Most of the research on heat transfer in rotor-stator system is performed using air; a correction with $Pr^{\frac{1}{3}}$ is used to get an estimation of the heat transfer coefficient in the case of a liquid filled cavity.

The stator heat transfer coefficient in a rotor-stator system increases with decreasing gap ratio. With a gap ratio ($G$) between 0.01 and 0.02 it varies from $2 \cdot 10^3$ W m$^{-2}$ K$^{-1}$ at $\omega = 25$ rad s$^{-1}$ to $7 \cdot 10^3$ W m$^{-2}$ K$^{-1}$ at $\omega = 130$ rad s$^{-1}$ (Howey et al., 2010, Yuan et al., 2003), and is expected to increase further with higher rotational disc speeds (Owen et al., 1974).

The rotor heat transfer coefficient decreases with decreasing gap ratio. At a gap ratio of 0.0115, it increases with increasing rotational disc speed, from $1.0 \cdot 10^4$ W m$^{-2}$ K$^{-1}$ at $\omega = 100$ rad s$^{-1}$, to $2.2 \cdot 10^4$ W m$^{-2}$ K$^{-1}$ at $\omega = 400$ rad s$^{-1}$ (Owen et al., 1974). The heat transfer coefficients towards the rotor are thus higher than towards the stator, although they are of the same order of magnitude (Howey et al., 2010, Owen and Rogers, 1989).

In the design as presented in this article, the cooling or heating is present in the stators, due to practical reasons, in spite of the fact that the heat transfer coefficient is higher at the rotor. The overall rate of heat transfer depends on the resistances towards heat transfer in series. This are, respectively, the heat transfer from the fluid in the spinning disc reactor towards the stator, the heat conduction through the stator wall, and the heat transfer from the stator wall to the cooling water:

$$\frac{1}{\alpha_{\text{tot}}} = \frac{1}{\alpha_{\text{SDR}}} + \frac{d_{\text{stator}}}{k_{\text{stator}}} + \frac{1}{\alpha_{C}}$$  \hspace{1cm} (6.10)

The heat transfer coefficient towards the stator is calculated via a correlation (Nikitenko, 1963, Owen and Rogers, 1989), which is corrected for the use of water ($Pr = 8$) instead of air ($Pr = 0.71$):

$$Nu = 0.0178Re^\frac{4}{5} Pr_{w}^{\frac{1}{2}} Pr_{air}^{\frac{1}{2}}$$  \hspace{1cm} (6.11)

The stator wall is made of stainless steel, has a thickness of 5 mm, and a heat conductivity ($k$) of 50 W m$^{-1}$ K$^{-1}$. The heat transfer coefficient inside the stator is calculated using a correlation for flow in tubes (Bird et al., 2002):

$$Nu_c = 0.035ReCPr_{w}^{\frac{1}{3}}$$  \hspace{1cm} (6.12)
Multistage rotor-stator spinning disc reactor

Figure 6.9: The resistances towards heat transfer in the rotor-stator spinning disc reactor, which is cooled with water. At rotational disc speeds above 50 rad s\(^{-1}\) the heat transfer is mainly determined by the heat transfer coefficient of the cooling water.

The hydraulic diameter is 6.6 \(\times\) 10\(^{-3}\) m, the superficial velocity of the cooling water is 0.5 m. The heat transfer resistances are plotted in Figure 6.9. At low rotational disc speeds (\(\omega < 50\) rad s\(^{-1}\)) the resistance is the largest on the reactor side, at higher rotational disc speeds the coolant side is determining the heat transfer rate. To increase the rate of this heat transfer step, and thus profit of the high heat transfer coefficient at the reactor side, modifications have to be made for the cooling side, e.g. another cooling fluid, a higher flow rate or a smaller hydraulic diameter.

The heat transfer coefficients in the rotor-stator spinning disc reactor are expected to be up to 2 \(\times\) 10\(^4\) W m\(^{-2}\) K\(^{-1}\), which is much higher than in conventional reactors, e.g. bubble columns (up to 5 \(\times\) 10\(^3\) W m\(^{-2}\) K\(^{-1}\)) (Fair et al., 1962), packed bed (up to 4 \(\times\) 10\(^3\) W m\(^{-2}\) K\(^{-1}\)) (Lamine et al., 1992) or stirred tank reactors (up to 1 \(\times\) 10\(^3\) W m\(^{-2}\) K\(^{-1}\)) (Steiff et al., 1980, Trambouze et al., 1988). Additionally, the area available for mass transfer is much higher, up to two orders of magnitude. In case either only the rotor or only the stators are used for heat transfer, an interfacial area of 1000 m\(^2\) m\(_R^{-3}\) can be reached, using a rotor-stator distance of 1 mm.

In case a heterogeneously catalyzed reaction is performed, the catalyst can be supported on the rotor and/or the stator. If the reaction is highly exothermic, the combination of the catalyst and the cooling liquid inside the rotor and/or the stator, the catalyst can be cooled locally. This prevents the formation of hotspots, or the large (radial) temperature gradient.
which is observed in packed bed reactors, which can have a large effect on the selectivity of the reaction, and on catalyst deactivation.

Bibliography


Meeuwse, M., van der Schaaf, J., Schouten, J. C., 2010d. Mass transfer in a rotor-stator


Conclusions and outlook

7.1 Conclusions

This thesis describes the research on the mass transfer and hydrodynamics in a new type of multiphase reactor, the rotor-stator spinning disc reactor. The rotor-stator spinning disc reactor consists of a rotating disc, enclosed in a cylindrical housing, with only a small gap, typically 1 mm, between the rotor and the stator. The gas-liquid mass transfer, liquid-solid mass transfer and the hydrodynamics are studied, in various reactor configurations.

In the first reactor configuration studied, the liquid is fed to the reactor via the top, near the rotating axis. The gas is added through a small orifice in the bottom stator, near the rim of the rotor. Small gas bubbles are sheared off due to the velocity gradient between the rotor and the stator, which acts a shear force. The gas bubble diameter therefore decreases with increasing rotational disc speed. The gas bubbles have an inward radial velocity, due to the centrifugal force; the gas holdup in the spinning disc reactor is thus only a few percent. The radial velocity decreases with decreasing gas bubble diameter. The residence time, and thus the gas holdup, increases therefore with increasing rotational disc speed. Combined with the decreasing gas bubble size, this gives an increase in gas-liquid interfacial area.

The gas bubbles formed at low rotational disc speeds are larger than the rotor-stator distance. The main part of the mass transfer will take place between the gas bubble and the liquid films on the rotor and the stator. At higher rotational disc speeds, the gas bubbles are smaller than the rotor-stator distance; the mass transfer rate is then determined by the size and velocity of the turbulent eddies. The volumetric mass transfer coefficient is therefore proportional to the energy dissipation rate.
The volumetric gas-liquid mass transfer coefficient increases with increasing gas flow rate, increasing rotational disc speed, increasing rotor radius, and decreasing rotor-stator distance, up to $2.5 \text{ m}^3 \text{ m}^{-3} \text{s}^{-1}$, at a gas flow rate of $1.5 \times 10^{-5} \text{ m}^3 \text{s}^{-1}$, a rotational disc speed of $209 \text{ rad} \text{s}^{-1}$, using a rotor with $0.135 \text{ m}$ radius and $1 \text{ mm}$ rotor-stator distance. This is one order of magnitude higher than conventional reactors, such as bubble columns or stirred tank reactors. The increase in energy dissipation rate with increasing rotor radius is higher than the increase in mass transfer; based on energy consumption it is therefore preferred to scale up the reactor by stacking multiple rotor-stator stages in series, instead of scaling up by increasing the rotor size. For the scale up of this reactor configuration, however, a system has to be developed to redistribute the gas at each stage.

Alternatively, gas and liquid can be fed together to the spinning disc reactor. A liquid film will form on the rotor surface, flowing outwards. The gas phase is present, as a gas bubble the size of the rotor, above the liquid film in this film flow region. At the rim of the rotor small gas bubbles are sheared off the gas bubble. The dispersed flow region, which is the remainder of the reactor volume, i.e. the region surrounding the rim of the disc and the region between the rotor and the bottom stator, is filled with liquid, with small gas bubbles dispersed in it. The main part of the mass transfer takes place in the dispersed flow region; the mass transfer is also higher than in case of a single gas inlet in the bottom stator. The mass transfer performance, $k_{GL}a_{GL}V_R$, which is the mass transfer rate divided by the driving force, is a factor 2 higher with the configuration where gas and liquid are fed together to the reactor compared with the situation with a single gas inlet in the bottom stator.

An additional advantage of co-feeding of gas and liquid is the easy scale up. It is shown that a 2-stage and a 3-stage rotor stator spinning disc reactor have the same mass transfer coefficients as the single stage reactor. This is a strong indication that the gas-liquid flow in the multiple stage reactor is the same as in a single stage reactor. The pressure drop in the reactor increases with increasing rotational disc speed and liquid flow rate, and is higher for gas-liquid flow than for single phase flow; the highest value obtained is $0.64 \text{ bar per stage}$, at $459 \text{ rad s}^{-1}$.

The liquid-solid mass transfer coefficient towards the rotor increases with increasing rotational disc speed, up to $7.9 \times 10^{-4} \text{ m}^3 \text{m}^{-2} \text{s}^{-1}$ at $157 \text{ rad s}^{-1}$. This is one order of magnitude higher than in conventional reactors, such as a packed bed reactor. The liquid-solid interfacial area can be increased up to $2000 \text{ m}^2 \text{ m}^{-3}$, by using the stator as well, which is known to have mass transfer rates of the same order of magnitude. At low rotational disc speeds the flow
in the rotor-stator system is laminar. When increasing the rotational disc speed, the laminar flow becomes unstable, until the flow is fully turbulent. This is observed in the values of the liquid-solid mass transfer coefficient as well.

The high values of the mass transfer coefficients, and the high expected values of the heat transfer coefficient, make the rotor-stator spinning disc reactor a promising reactor for multiphase reactions, in spite of the high energy input needed. Especially reactions with dangerous chemicals or reactions at high pressure can benefit from the high mass transfer rates, since this leads to a significant decrease in reactor volume. The expected high heat transfer coefficients, and high surface area for heat transfer, make the spinning disc reactor suitable for highly exothermic reactions. Furthermore, the high mass transfer coefficients can lead to a significant increase in selectivity in case of serial or parallel reactions.

7.2 Outlook

To explore the full potential of the gas-liquid mass transfer in the (multi-stage) rotor-stator spinning disc reactor, measurements have to be performed at rotational disc speeds that are higher than 200 rad s\(^{-1}\). The oxygen desorption method as used in this thesis is not sufficient for this, since the mass transfer rates are too high; the gas and the liquid are virtually in equilibrium with each other. The mass transfer rates can be measured using a reaction, which has to be fast enough to keep the concentration in the liquid bulk low, preferably at zero. This can for example be done using a homogeneously catalyzed reaction.

The liquid-solid mass transfer coefficient has to be determined up to higher rotational disc speeds in the multi-stage reactor as well. For this a heterogeneously catalyzed reaction is the best choice, since it allows the determination of the liquid-solid mass transfer coefficient, and it is a proof of principle for an industrial heterogeneously catalyzed process. For this a (very) fast reaction is required, to avoid kinetic limitations (as shown in Chapter 5). A hydrogenation, with a catalyst supported on the rotor would be a good choice for this. If no kinetic effects are observed it is also possible to perform the reaction for a gas-liquid-solid reaction as well. This gives the possibility to see whether this is the same as the gas-liquid and liquid-solid mass transfer steps in series.

Heat transfer plays in important role in a large number of industrial reactions. The heat transfer in the rotor-stator spinning disc reactor is expected to be high, based on literature in single phase rotor-stator systems, as described in the Appendix of Chapter 6. The influence of the presence of the gas phase on the heat transfer is not known, although this is an important
factor, and should thus be part of future research.

The residence time distribution of the liquid and the gas is an important point in reactor operation. Assumptions, based on visual observations in combination with information from literature, are used throughout this thesis for the analysis of the measurements. Preliminary residence time experiments show that the reactor models used are valid in most conditions, but not at all conditions, more research is needed to improve the models used. Plug flow behaviour would be favorable for most processes. Based on the residence time an estimation can be made of the number of stages needed to approach plug behaviour in the multiple spinning disc reactor.

The explanations given in this thesis for the observed phenomena, were mostly based on simplified theories, due to the lack of knowledge about the exact conditions inside the rotor-stator system. Almost no experimental and numerical data is present about the radial, tangential and axial velocities in a single phase system under the conditions used in this paper, mainly since the rotor-stator distance used here is smaller than the distance used in most of the investigations found in literature. A proper experimental and numerical study, under the conditions used in the rotor-stator spinning disc reactor, could help to increase the understanding of the observed phenomena. Additionally, the influence of the presence of a gas phase on the velocity profiles in the rotor-stator spinning disc reactor would be worth investigating, since this has a large influence on the hydrodynamics of both gas and liquid in the rotor-stator spinning disc reactor.
Nomenclature

Roman Symbols

$A_b$  
Gas bubble interfacial area  
m$^2$

$A_{top}$  
Area on top side of the rotor  
m$^2$

$a_{GL}$  
Gas-liquid interfacial area per unit volume of reactor  
m$_i^2$ m$^{-3}$

$a_{LS}$  
Liquid-solid interfacial area per unit volume of reactor  
m$_i^2$ m$^{-3}$

$A_{top}$  
Area on top side of the rotor  
m$^2$

$C$  
Oxygen concentration  
mol m$^{-3}$

$C_{cat}$  
Catalyst concentration  
m$_{cat}$ m$^{-3}$

$C_D$  
Drag force coefficient  
-

$C_{max}$  
Maximum solubility of oxygen in the liquid at 1 atm of air  
m$_L^{-3}$

$C_m$  
Torque coefficient  
-

$C_w$  
Flow rate coefficient, $C_w = \frac{\phi_L}{2\pi\nu R_D}$  
-

$D$  
Diffusion coefficient of oxygen in water  
m$_L^2$ s$^{-1}$

$d_b$  
(Equivalent) gas bubble diameter  
m$_G$

$D_{eff}$  
Effective diffusion coefficient in porous structure  
m$^2$ s$^{-1}$

$d_{stator}$  
Thickness stator wall  
m

$E_{act}$  
Activation energy  
J mol$^{-1}$

$E_d$  
Rate of energy dissipation  
J m$^{-3}$ s$^{-1}$

$G$  
Gap ratio, $G = \frac{h}{R_D}$  
-

$H$  
Henry coefficient  
m$_L^3$ Pa mol$^{-1}$

$h$  
Rotor-stator distance  
m$_R$

$H_D$  
Rotor thickness  
m$_R$

$k$  
Heat conductivity  
W m$^{-1}$ K$^{-1}$

$k_0$  
Pre-exponential factor  
m$_L^3$ m$^{-3}$ s$^{-1}$
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
<th>Unit</th>
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<tbody>
<tr>
<td>$k_A$</td>
<td>Pre-exponential factor Arrhenius equation</td>
<td>$m^3_L \text{mol}^{-1} \text{Pt}^{-1} \text{s}^{-1}$</td>
</tr>
<tr>
<td>$k_G$</td>
<td>Gas side mass transfer coefficient</td>
<td>$m^3_G \text{mol}^{-2} \text{s}^{-1}$</td>
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<td>$k_{GL}$</td>
<td>Gas-liquid mass transfer coefficient</td>
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<td>Liquid-solid mass transfer coefficient</td>
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<td>$L_b$</td>
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<td>$N$</td>
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<td>-</td>
</tr>
<tr>
<td>$n$</td>
<td>Empirical coefficient Richardson and Zaki model</td>
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<tr>
<td>$N_{Pt}$</td>
<td>Number of Platinum sites</td>
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<td>$Nu$</td>
<td>Nusselt number $Nu = \frac{\alpha R_h}{k}$ or $Nu_c = \frac{\alpha D_{\text{hydro}}}{k}$</td>
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<tr>
<td>$P_N$</td>
<td>Gauge pressure of stage N</td>
<td>Pa</td>
</tr>
<tr>
<td>$Pr$</td>
<td>Prandtl number $Pr = \frac{\mu C_p}{k}$</td>
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<td>Gas constant $R = m^3_G \text{Pa} \text{mol}^{-1} \text{K}^{-1}$</td>
<td>m$^3_R$</td>
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<tr>
<td>$r$</td>
<td>Radial position</td>
<td>m</td>
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<tr>
<td>$r_A$</td>
<td>Reaction rate</td>
<td>mol m$^{-3} \text{s}^{-1}$</td>
</tr>
<tr>
<td>$R_{ax}$</td>
<td>Radius of rotor axis</td>
<td>m$^3_R$</td>
</tr>
<tr>
<td>$R_b$</td>
<td>Bubble radius</td>
<td>m$^3_G$</td>
</tr>
<tr>
<td>$R_D$</td>
<td>Rotor radius</td>
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<tr>
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<tr>
<td>$R_{in}$</td>
<td>Radial position reactor inlet</td>
<td>m$^3_R$</td>
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<tr>
<td>$Sc$</td>
<td>Schmidt number, $Sc = \frac{\nu}{D}$</td>
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<tr>
<td>$Sh$</td>
<td>Sherwood number, $Sh = \frac{k_{LS} R_D}{D}$</td>
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<td>$Sh_F$</td>
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<tr>
<td>$T$</td>
<td>Temperature</td>
<td>K</td>
</tr>
<tr>
<td>$t$</td>
<td>Time</td>
<td>s</td>
</tr>
<tr>
<td>$t_c$</td>
<td>Contact time between gas and liquid</td>
<td>s</td>
</tr>
<tr>
<td>$v$</td>
<td>Superficial velocity</td>
<td>m s$^{-1}$</td>
</tr>
<tr>
<td>$V_b$</td>
<td>Gas bubble volume</td>
<td>m$^3_G$</td>
</tr>
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$V_D$ Volume dispersed flow region $\text{m}^3_R$

$V_F$ Volume film flow region $\text{m}^3_R$

$v_{G,i}$ Interstitial velocity gas phase $\text{m} \text{s}^{-1}$

$v_{G,s}$ Superficial velocity gas phase, $v_{G,s} = \frac{\phi_G}{2\pi rh}$ $\text{m} \text{s}^{-1}$

$v_{L,i}$ Interstitial velocity liquid phase $\text{m} \text{s}^{-1}$

$v_{L,s}$ Superficial velocity liquid phase, $v_{L,s} = \frac{\phi_L}{2\pi rh}$ $\text{m} \text{s}^{-1}$

$v_r$ Radial velocity $\text{m} \text{s}^{-1}$

$V_R$ Reactor volume $\text{m}^3_R$

$v_s$ Slip velocity between gas and liquid phase $\text{m} \text{s}^{-1}$

$V_{vessel}$ Volume storage vessel $\text{m}^3_R$

$v_\theta$ Tangential velocity $\text{m} \text{s}^{-1}$

$v_{\infty}$ Terminal velocity of gas bubble in stationary liquid $\text{m} \text{s}^{-1}$

$w$ Distance between the rim of the rotor and the reactor wall $\text{m}$

**Greek Symbols**

$\alpha$ Heat transfer coefficient $\text{W m}^{-2} \text{K}^{-1}$

$\delta$ Layer thickness $\text{m}$

$\varepsilon_G$ Gas holdup $\text{m}^3_G \text{m}^{-3}_R$

$\eta$ Effectiveness factor -

$\mu_L$ Dynamic viscosity of liquid $\text{Pa} \text{s}$

$\nu$ Kinematic viscosity, $\nu = \frac{\mu}{\rho}$ $\text{m}^2_L \text{s}^{-1}$

$\rho$ Density $\text{kg m}^{-3}_L$

$\tau$ Torque $\text{Nm}$

$\Phi$ Thiele modulus -

$\phi_G$ Gas flow rate $\text{m}^3_G \text{s}^{-1}$

$\phi_L$ Liquid flow rate $\text{m}^3_L \text{s}^{-1}$

$\omega$ Rotational disc speed rad s$^{-1}$

**Superscripts**

$in$ Inlet

$out$ Outlet

**Subscripts**

$atm$ Atmospheric

$C$ Cooling side
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<td>$D$</td>
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<td>Film flow region $F$</td>
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<td>$G$</td>
<td>Gas phase</td>
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<td>Interface</td>
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<td>$R$</td>
<td>Reactor</td>
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List of Publications

Journal Publications

Oral Presentations

• Meeuwse, M., Van der Schaaf, J., Schouten, J. C., 2010. Multiple rotor-stator spinning disc contactor: compact equipment for intensified multiphase processes. 19th International Congress of Chemical and Process Engineering (CHISA 2010) and the 7th European Congress of Chemical Engineering (ECCE-7) (pp. 1887-1888), 28 August - 1 September 2010, Prague, Czech Republic.


• Meeuwse, M., Van der Schaaf, J., Kuster, B. F. M., Schouten, J. C., 2008. Spinning disc reactor: Intensification of mass transfer and reaction rate. IXth Netherlands Catalysis and Chemistry Conference (NCCC IX) (p. 120), 3-5 March 2008, Noordwijkerhout, Netherlands.


Poster presentations


• Meeuwse, M., Van der Schaaf, J., Schouten, J. C., 2010. Multiple rotor-stator spinning disc contactor: compact equipment for intensified multiphase processes. 19th Inter-
national Congress of Chemical and Process Engineering (CHISA 2010) and the 7th European Congress of Chemical Engineering (ECCE-7) (pp. 365-366), 28 August - 1 September 2010, Prague, Czech Republic.


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Het schrijven van een proefschrift mag dan wel een individuele bezigheid zijn, de inhoud van dit proefschrift kon niet tot stand komen zonder de hulp van anderen. Daarbij valt te denken aan wetenschappelijke begeleiding en technische ondersteuning, maar ook aan koffiepauzes met collega’s en de broodnodige ontspanning in de avonduren. Ik wil daarom een ieder van harte bedanken die, op welke wijze dan ook, een positieve bijdrage heeft geleverd aan mijn promotie(tijd)!

Allereerst wil ik Jaap bedanken. Na mijn afstuderen kon ik direct doorstromen in dit promotieproject, waar ik de pionier kon worden van de rotor-stator spinning disc reactor. In dit project heb ik altijd de vrijheid gekregen om datgene te onderzoeken, waarvan ik dacht dat het relevant was. Op de juiste momenten heb ik echter wel de aansporing en bijsturing gekregen, die ervoor zorgde dat mijn project gestroomlijnd bleef lopen en dat ik ook artikelen ging produceren. Ik sta nog steeds versteld van het feit hoe snel je een artikel (inhoudelijk en taaltechnisch!) kunt corrigeren, al hebben de kleinste opmerkingen van jou (“waarom?” of “hoe?”) mij vaak de meeste hoofdbreken gekost. Jouw vaardigheid om in de huid van de reviewer te kruipen, heeft de volledigheid en leesbaarheid van mijn artikelen en abstracts zodanig omhoog gekrikt, dat ik mij nooit zorgen hoefde te maken of het wel geaccepteerd werd! Denise, bedankt voor het mij herinneren aan allerlei deadlines (hoewel ik daar niet altijd blij van was), en het regelen van allerhande formaliteiten, zoals het plannen van een promotiedatum, wat niet meeviel, maar gelukkig toch gelukt is!

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prullenbak had moeten verwijzen. Uiteindelijk zijn we daar in de meeste gevallen in geslaagd en is mede dankzij jouw bijdrage dit proefschrift er uitgekomen!

Ben, ook al geniet je al geruime tijd van je pensioen, je hebt in de eerste helft van mijn project nog een bijdrage kunnen leveren. Ik moet eerlijk toegeven dat ik lang niet al je ideeën tot uitvoer heb kunnen brengen (ik had tenslotte maar 4 jaar), maar deze input is toch zeker gewaardeerd. Daarnaast waren de projectbespreking soms buitengewoon amusant, als jij weer een discussie met John had over welke muziek (de Beatles of de Rolling Stones) de hoogste stofoverdracht in de spinning disc reactor zou hebben...

Ik wil technologiestichting STW bedanken voor het financieel mogelijk maken van dit project. Tevens wil ik de bedrijven uit de industriële gebruikerscommissie bedanken voor hun bijdrage tijdens de voortgangsbesprekingen: Alfa Laval, DSM, Akzo Nobel, TNO en MSD/Organon. I would like to thank especially Alfa Laval, for the support which made it possible to construct our beautiful multistage spinning disc reactor.

Onze vakgroep mag zich gelukkig prijzen met de werkplaats en het technische personeel wat er rondloopt. Chris, Erik en Dolf: ik ben heel vaak in de werkplaats geweest, en ondanks het feit dat jullie het vaak druk hadden, werd ik meestal toch heel snel geholpen. Jullie kennis, kunde en de inhoud van jullie voorraadkastjes hebben mijn project verder op weg geholpen! Anton, bedankt voor de hele lading spullen die jij voor mij hebt besteld en voor de planning van de werkzaamheden. Paul, bedankt voor het installeren en programmeren van de uitlezing en aansturing. Ik kan me voorstellen dat ik af en toe wel eens vervelend was, als ik weer belde of je een paar dingen aan kon passen, maar je kwam toch iedere keer weer snel langs, zodat ik weer verder kon. Ik kijk daarom met veel tevredenheid terug op onze samenwerking!

Madan, jij verdient een aparte alinea in dit dankwoord. Zonder jou waren de twee opstellingen, de “grote” spinning disc, en de “multidisc” er nooit gekomen, en waren ze helemaal niet zo goed en mooi geworden! Ik heb onze samenwerking altijd als zeer prettig ervaren, en we hebben een goed team gevormd: jij voor de technische details en de tekeningen, ik voor de berekeningen en de vervelende vragen om je scherp te houden. Ook al ging er ook wel eens iets mis (heb je de “kromme schijf” al verdrongen?), we mogen toch wel trots zijn op het eindresultaat. Het is niet voor niets dat iedere rondleiding langs de spinning disc opstellingen gaat! En aan mijn opvolger Frans wil ik ook wel wat meegeven: laat mijn mooie reactors heel! Maar ik heb er wel vertrouwen in dat dat (nu wel) gaat lukken, en ben ervan overtuigd dat jouw spinning disc project een succes wordt!
Het onderzoek op de spinning disc reactor heb ik niet alleen uitgevoerd, ik heb ook de hulp gehad van een aantal afstudeerders. Patrice, ook al heb jij niet zo heel veel aan de spinning disc kunnen meten, je hebt mijn project toch een vliegende start kunnen geven! Jordy, het video analyse script wat jij in elkaar gezet hebt, heeft de basis gevormd voor de beeldanalyse, een van de pijlers onder dit onderzoek! Sanne, het maken van een katalytische schijf heeft je bloed, zweet en tranen gekost, maar het heeft een mooi eindresultaat opgeleverd, wat zonder jou nooit was gelukt! Kevin, je indrukwekkende oplossingen van de Navier-Stokes vergelijking bleken helaas toch niet bruikbaar in de praktijk. Toch denk ik dat het een mooie exercitie is geweest, waar we veel van geleerd hebben, en ik weet zeker dat je wiskundige kwaliteiten tijdens je promotie wel tot resultaat zullen leiden! Edwin, de vele uren die jij achter de spinning disc hebt gezeten hebben mij geen windeieren gelegd, en ik denk dat je trots mag zijn op het resultaat! Voor jullie allemaal geldt: ik heb met veel plezier met jullie samengewerkt en ik wil jullie bedanken voor jullie bijdrage aan mijn project!

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De koffiekamer is het kloppend hart van onze afdeling, in het begin vooral een Nederlandse enclave, maar mede dankzij het Coffee Integration Program verworden tot het toonbeeld van een succesvolle multiculturele samenleving. Stijn, hulde voor het initiatief en het doorzettingsvermogen! Ook de andere notoire koffiedrinkers zal ik niet vergeten: Maurice, Niek, Patrick, Peter, Mart, Marlies, Anton, Christine, Jack, Joost, Carlo, Frans, Dulce en vele anderen.

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De congressen die ik heb mogen bezoeken zijn onvergetelijke ervaringen geweest. Ik wil graag mijn reisgenoten bedanken voor de geweldige tijd die we hebben beleefd in Veld-
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About the Author

Marco Meeuwse was born on the 15th of September, 1983, in Rheden. He finished secondary school (VWO) in 2001, and started with Chemical Engineering at Eindhoven University of Technology, where he graduated in 2006 (Cum Laude). His MSc. thesis was entitled: "Overall volumetric gas-liquid mass transfer coefficient in solid foam packings". He started in October 2006 with his PhD project in the Laboratory of Chemical Reactor Engineering (SCR), of Eindhoven University of Technology, under supervision of dr.ir. J. van der Schaaf, dr.ir. B.F.M. Kuster and prof.dr.ir J.C. Schouten. His PhD project involved reactor design, mass transfer and hydrodynamics in the rotor-stator spinning disc reactor. At January 1st 2011 he started as an Associate Scientist Reactor Engineering at the Advanced Chemical Engineering Solutions department of DSM Research, Technology and Analysis in Geleen.