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A review on process intensification in HiGee distillation

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Keywords

HiGee, distillation, gas-liquid contactor, heterogeneous catalyst, rotating packed bed

Abstract

This review paper describes the state-of-the-art in the field of HiGee contactors used for gas-liquid mass transfer processes, with a special focus on distillation, and for heterogeneously catalyzed reactions.

Several types of rotating beds are discussed, including single-block rotating packed-bed, split-packing rotating bed, rotating zigzag bed, two-stage counter-current rotating packed bed, blade packing rotating packed bed, rotating bed with blade packing and baffles, counter-flow concentric-ring rotating bed and crossflow concentric-baffle rotating bed.

The working principles of HiGee technology, as well as the modeling, design and control aspects, and practical applications are explained and discussed. In addition, this paper addresses the advantages and disadvantages with respect to mass-transfer performance, pressure drop, rotor complexity and suitability to perform continuous distillation and to be filled with catalyst packing for heterogeneous reactions.

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1 **1. Introduction**

2 The idea of exploiting high-gravity fields to intensify gas-liquid mass transfer has received
3 special attention over the last three decades, after a patent was granted to Ramshaw and
4 Mallinson in 1981¹. However, this concept has been around for more than a century, since
5 Elsenhans filed a patent in 1906² for a non rotor-stator rotating zigzag bed for purifying gases.
6 Schmidt patented the first rotating packed bed with wire mesh packing in 1913³. Then, Placek
7 was granted several patents, between 1933 and 1944, for a spiraling rotating bed⁴, a
8 corrugated plates with holes rotating bed^{5,6} and a concentric rings with holes rotating bed⁷.
9 Thereafter, Kapitza patented the first rotor-stator zigzag bed in 1952⁸ and Pilo and Dahlbeck
10 filed several patents⁹⁻¹¹ during the 60's for rotating packed beds for gas absorption and
11 desorption, distillation and reaction. However, Podbielniak was the first to address the effect
12 of gravity on the tallness of the distillation column in 1935¹² and continued his efforts on
13 process intensification in distillation and extraction for more than two decades¹³⁻²⁴.
14 HiGee contactors operate in a high-gravity field (100-1000 times gravity) in order to enhance
15 mass-transfer and throughput up to 1-2 orders of magnitude, allowing for a reduction in size
16 of up to 10 times with respect to conventional packed columns for the same separation²⁵⁻²⁷.
17 This size reduction has been attributed to their higher volumetric mass-transfer coefficients
18 and intensified momentum and heat transfer rates due to the formation of thinner liquid films
19 and smaller droplets which result in larger surface areas and improved micromixing
20 performance with respect to conventional packed bed equipment^{28,29}, offering the possibility
21 to use very high specific surface area packing. Additionally, high gravity relaxes flooding
22 limits that allow operation at higher gas velocities.

23 The high-gravity (HiGee) gas-liquid contactor most commonly mentioned in the literature is
24 the single-block rotating packed bed (RPB). However, subsequent developments have
25 resulted in several HiGee contactors with varying hydraulic and mass transfer characteristics,
26 depending mostly on their rotor design. These rotors can be rather complex due to the
27 combination of moving and stationary disks or even two disks moving in counter-direction.
28 The rotating zigzag bed seems to be of the best rotating beds for performing continuous
29 distillation because it allows for intermediate feeds within one single rotor and because of its
30 favorable combination of high mass transfer performance and higher liquid residence time
31 when compared to other rotating beds. However, its rotor is formed by concentric rings or
32 baffles that do not allow for catalyst to be used as packing. An upgraded version of the
33 rotating zigzag bed, the two-stage counter-current rotating packed bed^{30,31}, combines the
34 advantages of the rotating zigzag bed with the capability to use packing and therefore is seen,

1 together with the much simpler conventional RPB¹ (two-stage), as the most appropriate
2 equipment to attempt heterogeneous-catalyzed reactions among all HiGee contactors available
3 in the literature^{30,31}.

4 A disadvantage of HiGee gas-liquid contactors is their higher pressure drop compared to
5 conventional packed beds, however, the main disadvantages are the rotating parts, including
6 the rotor, bearings and dynamic seals, which result in long-term reliability concerns²⁶. This
7 disadvantage is mitigated by the fast and simple shutdown and startup of these equipment³²,
8 making it easy to perform preventive and corrective maintenance, without incurring high
9 production losses.

10 Several papers discussing HiGee designs have been published so far, including multi-stage
11 spraying rotating packed bed³³, RPB with wave-form disk packing³⁴, helical rotating
12 absorber³⁵, RPB with split packing (SP-RPB)^{30,36-39}, rotating zigzag bed (RZB)^{27,40,41}, two-
13 stage counter-current rotating packed bed (TSCC-RPB)^{30,31,36}, blade-packing rotating packed-
14 bed (BP-RPB)⁴²⁻⁴⁷, counter-flow concentric-ring rotating bed⁴⁸, cross-flow concentric-baffle
15 rotating bed⁴⁹. This review paper provides an overview of the HiGee technology available
16 both for gas-liquid mass-transfer -with a special focus on distillation- and for heterogeneously
17 catalyzed reactions. First, the working principles of different rotating beds reported in the
18 literature are explained. Next, modeling, design and control aspects of HiGee equipment are
19 presented. Finally, a current list of industrial applications is presented and discussed.

20

21 **2. Working principles**

22 The essence of HiGee technology is replacing the gravitational field by a high centrifugal
23 field achieved by rotating a cylindrical rigid bed. Figure 1 illustrates the working principle of
24 HiGee distillation in a rotating packed bed (RPB) with a vertical axis. The vapor-liquid
25 counter-current flow is horizontal in case of HiGee distillation and not vertical, as typical for
26 conventional operation. This means that the separation extent is determined by the diameter of
27 the rotor while its capacity is limited by its axial height – in contrast with conventional
28 distillation where the diameter determines the capacity and the height of the column gives the
29 separation extent^{50,51}. The rotor is an annular, cylindrical packed bed, a series of concentric
30 perforated baffles or a combination of both, housed in a casing and driven by a motor. Several
31 different HiGee devices have been developed over the last 30 years and the most relevant
32 ones are described in the next section. They differ mainly in their rotor design, since this
33 determines the main characteristics of rotating beds.

34

1 3. HiGee equipment

2 3.1 Single-block rotating packed-bed (RPB)

3 A single-block RPB is formed by a casing containing a rotor filled with packing, a shaft, a
4 liquid distributor and gas and liquid inlet/outlets. The rotor can be made up of diverse porous
5 media, such as wound wire mesh, foam metal, and corrugated sheet metal. The liquid is
6 injected onto the center of the rotor through a stationary set of nozzles (distributor) and is
7 thrown out into the packing. Then, it flows radially outwards as thin films, rivulets, or
8 droplets by centrifugal force and leaves the packing as a shower of droplets, which is
9 collected by the casing wall and runs downwards along the walls by the action of gravity,
10 leaving the casing. The gas, on the other hand, can flow in co-current, counter-current or
11 cross-current, with respect to the liquid; being the counter-current and cross-current
12 arrangements the most commonly used for gas-liquid contacting. These two different gas flow
13 arrangements are described in more detail down below.

14 Chen et al.⁵² and Yang et al.⁵³ demonstrated that the highest mass transfer rates in an RPB
15 take place at the inner-end zone of the rotor (r_i) due to several reasons: Firstly, because the
16 most violent gas-packing collisions occur at this location due to the high relative velocity
17 between the incoming liquid jets and the rotating packing. Secondly, because at r_i , the cross-
18 sectional area for flow is minimum and the gas flux is maximum. And lastly, because the
19 most rapid resupply of fresh liquid from the liquid distributor occur at this place.

20 *Counter-current flow rotating packed bed*

21 A simplified drawing of an RPB operating in counter-current flow is shown in Figure 2 (top).
22 The liquid is injected onto the center of the rotor and flows radially outwards as previously
23 described. The gas enters the equipment casing and then the rotor at the outer periphery and is
24 forced to flow radially inwards due to the pressure gradient. The gas and the liquid contact
25 each other counter-currently while mass transfer occurs. The gas leaves the packing at the eye
26 of the rotor through the outlet pipe.^{27,50} A mechanical seal is required in order to block any
27 gas bypass flow around the rotor.⁵⁴ In a counter-current flow rotating packed bed the gas flow
28 rate is limited by the flow area at the eye of the rotor where flooding is most likely to occur
29 since it is where the gas and liquid velocities are highest.

30 *Cross-current flow rotating packed bed*

31 A simplified drawing of a cross-current flow RPB is shown in Figure 2 (btm). The liquid is
32 injected onto the center of the rotor and flows radially outwards as previously described. The
33 gas enters at the bottom, flows axially through the packing and leaves the rotor from the top.

1 Therefore, the gas and the liquid contact each other in a cross-current flow in the rotor. In
2 contrast with the counter-current RPB, in the cross-current flow RPB the gas is not withdrawn
3 from the eye of the rotor and therefore the flooding constraint can be relaxed, enabling its
4 operation at higher gas flow rates^{55,56}. For instance, Guo et al.⁵⁶ studied the hydrodynamics
5 and mass transfer characteristics in a cross-current flow rotating packed bed and found that it
6 could operate at gas flow rates as high as 15m/s without flooding. At such high gas flow rates,
7 the droplets formed at the top of the packing are likely to be instantaneously entrained by the
8 gas. This can be prevented by having a section of non-irrigated packing at the downstream side
9 to separate out the droplets by inertial impact⁵⁶. The cross-current flow arrangement has been
10 reported to result in larger volumetric mass transfer coefficients than counter-current flow
11 RPBs but with a lower pressure drop at the same operational conditions^{57,58}.

12 Both counter-current and cross-current flow single-block RPBs have some drawbacks. The
13 liquid residence time inside the rotor is very short, which limits the separation extent despite
14 the very high mass transfer rates⁴⁰. Since the whole packing is rotating, it is difficult to insert
15 middle-feed streams and therefore, multiple rotors are required for continuous distillation.
16 This is not, however, a crucial downside since the limited amount of equilibrium stages
17 available per rotor makes multiple rotors necessary for distillation anyway. A coaxial
18 multirotor configuration (i.e. multiple rotors coaxially installed in one casing) is structurally
19 complicated due to the need of dynamic seals and liquid collectors⁴⁰ and for this reason,
20 multirotor configurations usually involve the use of two or three rotating packed beds driven
21 by individual motors. In addition to that, Rao et al.²⁵ and Sandilya et al.⁵⁹ showed that
22 although both volumetric mass transfer coefficients ($k_G a$ and $k_L a$) increase with the rotational
23 speed, this is due to larger interfacial areas (a), to enhanced liquid-side mass transfer
24 coefficients (k_L) and to the intense mass transfer in the entry region (end effect^{52,53}), but not
25 to enhancements in the gas-side mass transfer coefficients (k_G), which are only marginal.
26 They suggest that this occurs because the gas acquires the same angular velocity of the
27 packing soon as it enters the rotor and starts rotating as a rigid body along with the packing.
28 Sandilya et al.⁵⁹ compared experimental results of k_G in an RPB with those calculated using
29 the correlation proposed by Onda et al.⁶⁰ for conventional packed columns, given by

$$30 \quad k_G = C \left(\frac{D_G a_t}{RT} \right) Re_G^{0.7} Sc_G^{1/3} (a_t d_p)^{-2} \quad (1)$$

31 They found that the k_G values in the RPB were even lower than the ones estimated using
32 Onda's correlation and attributed this to liquid maldistribution. Thus the gas flow is similar to

1 that through a stationary rotor, the gas-side mass transfer coefficients lie in a similar range
2 than for conventional trickle beds and no intensification of the gas-side mass transfer is
3 expected other than that due to the larger interfacial area. The gas velocity, however, can
4 increase significantly compared to trickle beds under the high-gravity field, and, according to
5 equation (1), this leads to larger gas-side mass transfer coefficients since k_G depends on the
6 gas Reynolds number.

8 **3.2 Rotating packed-bed with split packing (SP-RPB)**

9 Chandra et al.⁶¹ came up with an alternative rotating packed bed design (Figure 3) in which
10 the rotor was split into two sets of alternate annular rings of packing (wire mesh or metal
11 foam) with gaps in between. One of the sets was fixed to the top disk and the other one to the
12 bottom disk. The two sets of rings were rotated with two motors in a counter- or co-direction
13 to promote gas tangential slip inside the packing and increase the gas-side mass transfer
14 coefficient, as suggested by Rao et al.⁵⁹.

15 The SP-RPB design is much more complex than the single-block RPB, especially due to the
16 need for two rotors spinning independently³⁹. In spite of this, recent studies³⁹ have suggested
17 that the split-packing RPB design may be superior over conventional single-block RPB only
18 for gas-side resistance controlled mass transfer processes. Similar to the single-block RPB, it
19 is difficult to insert middle-feed streams in a SP-RPB since both the upper and lower disks are
20 rotating during operation^{61,62}. Additionally, a multistage configuration of rotor-rotor stages on
21 a single axis is mechanically impossible.

23 **3.3 Rotating zigzag bed (RZB)**

24 In 1952, Kapitzka⁸ patented the so-called rotating zigzag bed. Later, in 2008, Ji et al.⁶³ patented
25 the rotating zigzag bed^{40,41} again. The RZB is composed of a rotating and a stationary disk as
26 shown in Figure 4. In the rotating zigzag bed, concentric circular baffles are fixed on the
27 rotational and stationary disks and serve as the contacting elements of gas and liquid phases.
28 These baffles are assembled together, alternating between rotational and stationary baffles.
29 The rotational baffles have perforations on their upper part and are fixed on the lower
30 (rotational) disk. The stationary baffles are fixed on the upper disk. The gas and liquid flow in
31 zigzag through the clearance between the rotational baffles and the upper disk and through the
32 clearance between stationary baffles and the lower disk. As in the single-block RPB, the gas is
33 fed through the casing and is forced to flow to the rotor and radially inwards through the

1 packing due to the pressure gradient. The liquid is fed at the center of the rotor and flows
2 radially outwards, contacting the gas in counter-current, due to the centrifugal force.
3 In a RZB the liquid is thrown by centrifugal force from the rotational baffles into the
4 stationary baffles, resulting in very fine droplets. For this reason, the rotating zigzag bed can
5 function without liquid distributors^{27,40,41}. In addition to that and thanks to its upper disk being
6 stationary, the dynamic seal can be eliminated and intermediate feeds can be easily introduced
7 at any radial length, making continuous distillation possible without the need of two rotors,
8 provided that the required number of theoretical stages can be reached within a single rotor.
9 The mass transfer performance of a rotating zigzag bed is comparable to that of a single-block
10 rotating packed bed but with better operability at a higher turndown ratio⁴⁰ (ratio of the
11 highest and lowest achievable flow rates). However, its separation ability can be further
12 improved by simply installing multiple rotors in one casing. The RZB, on the other hand, has
13 a higher power consumption than the single-block RPB since every time the liquid contacts
14 the static baffles it is brought to a halt and must be accelerated again by the rotating baffles.
15 Additionally, the zigzag path of the gas results in a higher gas pressure drop compared to an
16 RPB^{40,41,64}. The fact that no packing is used in the RZB, significantly reduces its available
17 surface area for gas-liquid contacting^{30,40,41}, making it unsuitable for heterogeneously
18 catalyzed reactions.

19

20 **3.4 Two-stage counter-current rotating-packed bed (TSCC-RPB)**

21 Luo et al.³⁰ developed and investigated a design that combined the packed bed and the zigzag
22 rotor design. The TSCC-RPB (Figure 5) has two stages and each stage has a rotor³¹. The two
23 rotors are installed on one shaft driven by one motor and each rotor is made up of a rotating
24 disk, fixed to the shaft, and a stationary disk, fixed to the housing. Packing rings and
25 concentric rings are attached to the rotating disk and the stationary disk, respectively, to
26 enhance the collision between the liquid and the packing and to lengthen the contact time.
27 Conventional packing or catalysts can be loaded into the concentric rotating rings for
28 distillation, heterogeneously catalyzed reactions and/or catalytic (homogenous and
29 heterogeneous) reactive distillation³⁰.

30 The liquid is fed into the upper rotor through a stationary distributor, and it moves outwards
31 due to the centrifugal force, passing through both the porous stationary rings and packed
32 rotating rings. The liquid is then collected at the bottom of the upper housing and flows into
33 the eye at the center of the lower rotor. It then flows into the lower rotor and radially
34 outwards, leaving the RPB from the liquid outlet.

1 The gas is tangentially introduced into the TSCC-RPB from the gas inlet and flows in
2 sequence through the lower rotor, the eye of the lower rotor and the upper rotor before leaving
3 the rotating packed-bed through the gas outlet. The liquid and the gas are thus contacted
4 counter-currently while mass transfer takes place.

5 Since the TSCC-RPB has two stages, it can be used for continuous distillation, with the upper
6 rotor serving as a rectifying section and the lower rotor as a stripping section. Besides,
7 intermediate feeds can also be introduced by installing liquid distributors on top of the
8 stationary disk, making continuous distillation possible with a single rotor, provided the
9 number of equilibrium stages per rotor is enough for the required separation.

10 The main drawback of the TSCC-RPB is its complex rotor structure with the combination of
11 rotational packing and static rings that require high manufacture precision, resulting in high
12 costs that brings additional to its industrial application³⁶.

14 **3.5 Blade packing-rotating packed-bed (BP-RPB)**

15 Lin et al.⁴² developed an RPB equipped with blade packings in order to achieve a low gas
16 pressure drop compared to previous HiGee contactors. The packings were made up of 12
17 blades covered in stainless steel wire mesh that were installed inside the rotor, keeping a
18 separation angle of 30 degrees among each other (Figure 6). Liquid left the distributor at a
19 relatively high velocity and then entered the inner side of the rotor and moved radially
20 outwards while contacting the gas counter-currently. However, the structure of the rotor with
21 blades is limited by the number of blades and therefore has a lower surface area compared to a
22 conventional rotating packed bed⁶⁵.

23 Other variations of HiGee gas-liquid contactors combining packing and blades⁴⁴, and blade
24 packing and baffles⁴⁵ have been also developed. Luo et al.⁴⁴ came up with a rotor with
25 packing and blades shown in Figure 7. This rather complex rotor was developed based on
26 previous observations about the end effect in RPBs^{52,53}. In their work, Luo et al.⁴⁴ built and
27 tested five different rotors equipped with packing and blades to artificially create multiple end
28 zones inside the rotors and thus enhance mass transfer in the so-called bulk zone (the zone of
29 an RPB rotor where the end effect is not dominant). These rotors had three rings of packing
30 sections separated by two rings of blades. The design parameter changed among the five
31 rotors was the angle between the plane of their blades and the equatorial line of the rotor
32 (Figure 7). The experimental results reported⁴⁴ indicate that the rotors with packing and
33 blades can intensify the mass transfer process over a range of gas-liquid ratios, resulting in
34 both larger mass transfer coefficients and specific surface areas than single-block rotating

1 packed beds. They attributed this to the disintegration of the liquid into tiny liquid droplets
2 caused by the more energetic gas-liquid interactions in the so-called “artificially-created end
3 zones” between the high-voidage packing and the blades. The main drawback of this design is
4 the higher complexity of its rotor structure compared to a conventional RPB rotor⁴⁴.

5 Sung and Chen⁴⁵ came up with another variation of the blade-packing rotating packed-bed
6 which has a rotor with blade packings and static baffles. A schematic drawing of this rotor is
7 shown in Figure 8. The blade packings and stationary baffles are alternately aligned in the
8 radial direction of the rotor and are fixed to a rotational and a stationary disk, respectively.
9 The stationary baffles were fixed at the clearance between sets of blade packings keeping a
10 distance of 2 cm between the two disks. These baffles retard gas rotation and provide a high
11 annular slip velocity between the gas and packing while producing a lower pressure drop than
12 in an RPB and a rotating bed with blade packings⁴⁵.

14 **3.6 Counter-flow concentric-ring rotating bed**

15 The rotor of the counter-flow concentric-ring rotating bed, CFCR-RPB (Figure 9), was
16 developed by Li et al.⁴⁸ in an attempt to improve the RZB by perforating all parts of the
17 rotating baffles (not only the top, as in the RZB) and by eliminating the stationary baffles. The
18 rotor comprises a rotating disk, driven by a motor, and a stationary disk. A set of concentric
19 circular metal rings with small perforations acting as gas and liquid channels is fixed to the
20 rotating disk with equal radial spacing. Concentric circular grooves are made on the lower
21 surface of the stationary disk such that when the two disks are assembled, the top of the
22 concentric rotating rings extends into the concentric grooves, forming a tight labyrinth seal to
23 prevent gas from bypassing the rotating rings. Intermediate feeds can be introduced on top of
24 the stationary disk, which is attached to the casing.

25 The liquid enters through the liquid inlet of the rotating bed and flows to the rotating liquid
26 distributor, where it is dispersed as fine liquid droplets into the center of the bed. The liquid
27 droplets flow radially outwards through the perforations on the rotating rings due to the
28 centrifugal force and they are discharged through the liquid outlet of the casing. The gas is
29 tangentially introduced into the casing and flows radially inwards through the perforations of
30 rotating rings due to the pressure difference and is discharged through the gas outlet. Gas and
31 liquid contact each other counter-currently while mass transfer takes place.

32 Li et al.⁴⁸ performed total reflux distillation experiments at atmospheric pressure using an
33 ethanol-water system in a counter-flow concentric-ring rotating bed. They compared the
34 equipment performance with that of an RZB and found that even though the counter-flow

1 concentric-ring rotating bed had a lower mass transfer performance, its gas-liquid throughput
2 was at least 5.6 times greater than that of RZB, which reached its flooding limits at an F-
3 factor of $0.66 \text{ m/s (kg/m}^3)^{0.5}$ while the counter-flow concentric ring rotating bed could operate
4 without flooding up to an F-factor of $3.68 \text{ m/s (kg/m}^3)^{0.5}$. Additionally, its pressure drop per
5 discrete step was comparable to that of the RZB at different F-factors until the flooding limit
6 of the RZB was reached and suddenly increased its pressure drop.

7 8 **3.7 Cross-flow concentric-baffle rotating bed (CRB)**

9 Another modified version of the RZB is the crossflow concentric-baffle rotating bed (CRB)
10 developed by Wang et al.⁴⁹ (Figure 10). In the CRB the gas flows in zigzag towards the center
11 of the bed while the liquid flows radially outwards, contacting the gas in cross-current flow.

12 The rotor comprises a set of perforated concentric baffles, which are fixed to the rotational
13 disk and extend into concentric grooves made into the lower surface of the stationary disk, as
14 described previously for the counter-flow concentric ring rotating bed. Each baffle is divided
15 into three zones in the axial direction: gas-hole zone, liquid-hole zone, and non-hole zone.
16 The baffles are fixed to the rotational disk in such a way that alternate baffles are in axially
17 opposite directions. Due to pressure difference, gas flows through the zigzag flow channels
18 formed by the gas-hole zones on all baffles and the annular space between the adjacent
19 baffles. Liquid flows radially outwards due to the centrifugal force, passing as fine droplets
20 through the liquid-hole zones on all baffles. The liquid is then collected on the casing wall and
21 leaves the rotating bed through the liquid outlet.

22 The crossflow concentric-baffle rotating bed has lower shaft power requirements and little
23 backmixing compared to the rotating zigzag bed. However, the stage efficiency of the CRB is
24 one-third as much as that of the RZB, or even lower. The stage efficiency is here defined as
25 the ratio between the number of theoretical stages achieved in the rotating bed and the number
26 of contacting stages (annular spaces between adjacent rotational baffles).

27 28 **3.8 Other HiGee equipment**

29 *Multi-stage spraying rotating packed bed*

30 A multi-staged spraying rotating bed is similar to the crossflow RPB in flow mode, the rotor
31 of which comprises of multiple concentric packing rings. In the “spraying zone”, the liquid
32 was sprayed into fine droplets with a large interfacial area due to centrifugal force. Its
33 disadvantage is potential liquid entrained by gas flow despite lower gas frictional resistance.⁴⁰

1 *Rotating packed-bed with wave-form disk packing*

2 To effectively reduce gas flow resistance, a waveform disk rotating bed was developed, in
3 which the rotor contains a series of concentric waveform disks. This rotating bed can utilize
4 the extended interfacial area generated by atomization besides the surface area of disk.⁴⁰

5 *Helical rotating bed*

6 A helical rotating bed features four spiral blades installed on a rotating disk. Due to the small
7 contact area, its mass transfer performance is still lower compared to a rotating packing bed in
8 spite of the longer flow channel and residence time of gas and liquid. For a helical rotating
9 bed, long liquid residence time in the rotor is offset by the small interfacial area, which results
10 in low volumetric mass transfer coefficients.⁴⁰

11

12 Table 1 shows a list of the gas-liquid HiGee contactors that have been described in this
13 section, along with a summary of their advantages and disadvantages, as well as their
14 applications reported in the open literature. Table 2 summarizes the results of relevant
15 experimental studies of HiGee distillation reported in the literature.

16 It should be noted that, contrary to what is expected, the superficial gas and liquid velocities
17 often employed in HiGee are only a fraction of those used in packed columns. Such flow rates
18 lead to very low HTU and HETP values, but they may not be useful for design. Overall, the
19 HETP values shown in Table 2 vary widely between 1.02 cm and 15.0 cm and their variation
20 depends on the rotor design, the separation system and the operating conditions during the
21 experiments. However, the incomplete or no specification of the liquid distributor and/or the
22 packings in many HiGee studies, added to the aforementioned low superficial velocities
23 offsets the usefulness of the mass transfer data reported and makes it difficult to fairly
24 compare different HiGee configurations. For instance, the K_{Ga} data reported by Nascimento
25 et al.⁶⁶ is several orders of magnitude higher than the data reported by Ramshaw and
26 Mallinson¹. Since the former does not report gas flows, it is not possible to explain this
27 differences based on gas Reynolds numbers. In the end, the most efficient design is the one
28 with the highest mass transfer to power input ratio, where the power input is a function of the
29 pressure drop, gas velocity and shaft power.

30 While stripping, absorption and distillation have been studied in most of the aforementioned
31 HiGee contactors and some related industrial applications have been implemented (see section
32 6), limited research has been conducted on solid-catalyzed reactions⁶⁷⁻⁷¹ and/or catalytic
33 distillation⁷². This may be due to the typically low liquid holdups of rotating beds²⁸, which do

1 not only affect their fractional recovery of solute but also the attainable conversions.

2

3 **4. Modeling and simulation**

4 When it comes to modeling and simulation, the main differences between a conventional
5 packed column and a HiGee unit are: (1) A packed column is a straight bed while a HiGee
6 may be seen as a tapered bed, in which the cross-sectional area for flow varies along the radial
7 direction, leading to varying gas and liquid velocities. As a result, both gas-side and liquid-
8 side transfer coefficients vary along the radius, unlike in straight beds. (2) In a packed bed
9 mass transfer occurs only along the packing while in HiGee an additional mass transfer zone
10 exists between the rotor and the casing. These two differences are, unfortunately, not
11 adequately appreciated in the literature.

12 A small number of models describing mass-transfer in HiGee gas-liquid contacting processes
13 are available in the literature. Many of them have focused on modeling of absorption and
14 stripping, while just a few on distillation and solid catalyzed stripping. For absorption and
15 stripping, most of the models available are developed based on first principles whereas
16 distillation models usually involve the use of commercial process simulators (e.g. Aspen Plus)
17 and discretization tricks to adapt the available distillation modules to include the effect of
18 centrifugal force and the different flow geometry (radial flow instead of axial flow).

19 Most HiGee models are developed following a non-equilibrium (NEQ) modelling approach
20 analogous to the one explained in detail by Taylor and Krishna for distillation⁷³ and reactive
21 distillation⁷⁴ in conventional columns. In contrast with equilibrium models, non-equilibrium
22 models do not assume that the streams leaving a separation stage are in thermodynamic
23 equilibrium. Instead, NEQ models consider phase equilibrium only at the gas-liquid
24 interphase and therefore take into account that the mass transfer of components from one
25 phase to the other occurs at a certain rate, which is proportional to the concentration gradient.
26 Since phase equilibrium is only assumed to exist at the interphase, separate balance equations
27 are written for all components and for each phase. The resulting balance equations are often
28 called MERSHQ equations (where M = material balances, E = energy balances, R = mass-
29 and heat-transfer rate equations, S = summation equations, H = hydraulic equations for the
30 pressure drop, and Q = equilibrium equations).⁷³

31 When modeling HiGee contactors, the MERSHQ equations are used to write balances over a
32 differential element with radial length (dR) and surface area ($2\pi Rh$). Three different theories
33 are often used to describe mass transfer, they are: film theory^{75,76}, penetration theory⁷⁷ and
34 surface renewal theory⁷⁸. In case of reactive systems, a reaction term is included in the

1 component material balances^{74,79}. If a solid catalyst is used, the possible effect of intraparticle
 2 diffusional limitations is accounted for by introducing effectiveness factors when calculating
 3 the actual reaction rate⁷⁹. For heterogeneous reactions, such as solid catalyzed reactions, the
 4 overall rate of reaction can be limited by mass transfer of the reactants from their respective
 5 phases to the active sites of the catalyst. The overall resistance to mass transfer and resistance
 6 can be written as the sum of mass transfer resistances in series⁸⁰. For instance, for a solid-
 7 catalyzed first order reaction in which gas phase reactant A dissolves in a liquid phase and
 8 then diffuses through the liquid and solid catalyst, where it reacts, the overall rate of reaction
 9 can be written as:

$$10 \quad -r_A = \left(\frac{RT}{Hk_G a_{GL}} + \frac{l}{k_L a_{GL}} + \frac{l}{k_S a_{LS}} + \frac{l}{\eta k_r C_{Cat}} \right)^{-1} \frac{RTC_{A,G}}{H} \quad (2)$$

11 The first three terms in equation (2) account for the resistances to mass transfer of A from the
 12 bulk of the gas phase to the liquid-solid interface, and the last term accounts for the resistance
 13 to diffusion through the porous catalyst towards the active sites, where reaction occurs. The
 14 decrease in the reaction rate due to internal diffusional limitations in the porous catalyst is
 15 incorporated by the effectiveness factor.

16 Depending on the flow arrangement and on whether or not a steady-state assumption is valid,
 17 the model can result in a in a few ordinary differential equations or in a system of partial
 18 differential equations that must be solved simultaneously to yield the concentrations as a
 19 function of the radius and other important parameters. The development of HiGee models for
 20 configurations with counter-current flow is usually simpler than for those with cross-current
 21 flow. In the former, the flow is mainly in the radial direction and the variation of gas and
 22 liquid compositions in the axial direction can be neglected. In the latter, the liquid flows in the
 23 radial direction while the gas flows in the axial direction⁵⁵, therefore concentrations change in
 24 both directions and the model equations become more complex.

25 As Table 3 shows, different assumptions with respect to the liquid flow pattern are made
 26 when modeling HiGee contactors. The flow pattern assumed has a direct effect on the
 27 calculation of the effective surface area available for mass transfer⁸¹ and therefore on the
 28 model estimate for the mass transfer coefficients. Earlier mass-transfer models, i.e. the one by
 29 Munjal et al.⁸², considered that the area available for mass transfer in an RPB was provided by
 30 a thin film flowing over some or all of the packing. Burns and Ramshaw⁸³ performed a visual
 31 study of the liquid flow in an RPB and reported that for rotational speeds between 300 and
 32 600 rpm liquid flow occurs in the pores in the form of radial rivulets whereas for rotational

1 speeds above 600-800 rpm droplet flow and film flow predominate, with a higher proportion
2 of droplet flow as the rotational speed increases. Based on these observations, more recent
3 models assume droplet flow or a combination of droplet flow and film flow over the packing.
4 Guo et al.⁵⁶ developed a model to describe three types of mass transfer processes (a gas-side
5 mass transfer controlled process, a liquid-side mass transfer controlled process and gas-side
6 mass transfer controlled process with reaction) in a cross-flow RPB. In their work, Guo et al.
7 presented experimental correlations to estimate the droplet diameter and the film thickness as
8 a function of the centrifugal force and the specific area of the packing. Whether or not these
9 correlations represent the complex droplet-droplet and droplet-packing collisions inside the
10 packings for a given system needs to be proven on a case by case basis.

11

12 *First-principle mass-transfer models*

13 One of the earliest models for mass transfer in a rotating packed bed based on first principles
14 was developed by Munjal et al.⁸². They used penetration theory and the complete convection-
15 diffusion model to obtain correlations for the estimation of gas-liquid (k_L) and liquid-solid
16 (k_s) mass-transfer coefficients in rotating packed beds. The authors approximated the flow in
17 a high gravity packed bed by liquid-film flow along the flat vertical surface of a rotating blade
18 and liquid-film flow along the horizontal surface of a rotating disk. The expressions
19 developed for gas-liquid and liquid-solid mass transfer coefficients on these idealized surfaces
20 were then extended to the correlations for rotating packed beds.

21 Chen et al.⁸⁴ presented a rigorous dynamic model for the ozonation of a pollutant (o-Cresol).
22 The model considers the simultaneous ozone and oxygen mass transfer, the chemical
23 reactions of ozone self-decomposition, and pollutant ozonation and the effect of chemical
24 reactions on gas-liquid mass transfer. The resulting system of partial differential equations is
25 solved using the finite difference method based on the Taylor series. The model predicts the
26 dynamic variations of ozone, o-cresol and oxygen concentration profiles in RPB.

27 Sun et al.⁸⁵ developed a model to describe the simultaneous absorption of two gases with a
28 pseudo-first-order reaction between them at the liquid surface in a rotating packed bed. The
29 model was used to estimate the overall volumetric mass-transfer coefficients and it was
30 validated with the simultaneous absorption of CO₂ and NH₃ into water. They neglected end
31 effects and pressure drop and assumed the liquid flow in the rotor to be in the form of droplet
32 flow in the void and film flow on the packing surface. As a result, the total gas-liquid
33 interfacial area consisted of the surface area of the packing plus the surface area of all of the
34 droplets. They then developed expressions for the liquid side mass transfer coefficients both

1 in the droplets and the liquid film in terms of the droplet size and the film thickness,
 2 respectively. The model took into account the effect of rotational speed on the mass transfer
 3 area of the droplets and the liquid film, but assumed these parameters to remain constant
 4 along the radial direction. They then used the correlations for k_L , together with Onda's
 5 correlation⁶⁰, equation (1), for the gas-side mass transfer coefficient, to calculate the overall
 6 mass transfer coefficient using equation (3). The $K_G a$ values estimated by the model were
 7 found to agree well with the experimental results at various liquid volumetric flow rates, gas
 8 volumetric flow rates, rotational speeds, and NH_3/CO_2 molar ratios.

$$9 \quad \frac{1}{K_G a} = \frac{1}{k_G a_{GL}} + \frac{H}{k_L a_{GL}} = \frac{1}{k_G a_{GL}} + \frac{H}{k_{L_1} a_d + k_{L_2} a_f} \quad (3)$$

10 Here, the area of mass transfer is composed of the surface area of the droplets (a_d) and the
 11 surface area of the liquid film over the packing (a_f).

12 Yi et al.⁸⁶ modeled the gas-liquid mass transfer with reactions for the absorption of CO_2 by a
 13 Benfield solution in a rotating packed bed. They assumed liquid flow to be in the form of
 14 spherical droplets, neglecting laminar film flow at high rotating speeds. In contrast to
 15 previous models^{56,87}, theirs takes into account the variation in droplet size within the rotor
 16 along the radial direction. They went further to divide the rotor into two zones, the end zone
 17 and the bulk packing zone, and used correlations developed based on the droplet size
 18 measurements done by Zhang⁸⁸ to calculate the droplet size in each of the zones as a function
 19 of centrifugal acceleration. Most of the calculated mole fractions of the CO_2 in the outlet gas
 20 agreed well with the experimental data with a deviation within 10%. Furthermore, they
 21 presented a $K_G a$ profile along the radial direction of the packing.

22 Quian et al.^{89,90} developed a reaction-equilibrium-mass transfer model based on penetration
 23 theory to describe the selective H_2S absorption process in methyldiethanolamine (MDEA).
 24 Their model is based on three main assumptions: (1) Liquid flow in the rotating packed bed is
 25 laminar film flow only, (2) surface area of packing regarded as gas-liquid effective interfacial
 26 area, (3) the rotor in the rotating packed bed consists of a given number of layers and the film
 27 is renewed once every time it passes through one layer of packing. They validated the model
 28 using industrial scale experimental data obtained in a refinery in China. Solid-catalyzed
 29 reactive stripping for the production of octyl-hexanoate with simultaneous water removal
 30 from the reaction zone has been modeled by Gudena et al.⁷⁹ Their work included a
 31 mathematical model derived from first principles in order to study the diffusional mass
 32 transfer within a porous catalyst in an RPB by using the effectiveness factor as a measure of

1 diffusional resistance. They analyzed the influence of the centrifugal field on the variation of
 2 the catalyst effectiveness factor and the selectivity in an esterification reaction. Table 3 shows
 3 an overview of some of the first-principle modeling work done on rotating packed beds for
 4 gas-liquid processes.

5 *RPB simulations with the aid of process simulators*

6 Besides the mass-transfer models mentioned so far, there are also those developed with the
 7 aid of commercial process simulators. However, the fact that these simulators do not include a
 8 built-in module to model and simulate HiGee processes is a hurdle. This is especially true
 9 when modeling more complex processes such as distillation or reactive systems for which
 10 first-principle models can be both difficult and tedious. For this reason, most of the modeling
 11 and simulation studies of rotating packed beds reported in the literature⁹¹⁻⁹⁶ have opted to
 12 modify existing units (e.g. RADFRAC module in Aspen Plus) so that the special
 13 characteristics of HiGee contactors, such as the effect of the centrifugal field on mass transfer
 14 coefficients and pressure drop and the variation of flow area with radial length, can be taken
 15 into account. The rate-based functionality of Aspen Plus was used in all these cases, allowing
 16 for rigorous simulations based on non-equilibrium models, which use rate equations and
 17 experimentally obtained correlations for mass transfer, pressure drop and rates of reaction, in
 18 the case of reactive systems. It is worth mentioning that most of the mass transfer studies in
 19 HiGee have used rotors with inner and outer radii of about 2 and 8 cm, respectively, and have
 20 reported average mass transfer coefficients. The use of these data for the simulation of an
 21 industrial size HiGee of 0.5 m radius involves a huge and risky extrapolation due to the
 22 substantial variation of the flow area as the radius increases. Local mass transfer coefficients
 23 should be used instead in such cases, when the variation coefficient is very large. To the best
 24 of our knowledge, only Reddy et al.⁶² have reported local coefficients for their split-packing
 25 design which are not based on a broad range of parameters. The correlations developed by
 26 Chen et al.⁹⁷ and Chen⁹⁸ (equations (4) and (5)) can be used to estimate liquid-side and gas-
 27 side mass transfer coefficients since they have been shown to account for end-effects, packing
 28 characteristics and the size of HiGee contactors. Their application to any industrial large size
 29 HiGee should be done with caution.

30
$$\frac{k_L a_{GL} d_p}{D_L a_t} \left(1 - 0.93 \frac{V_o}{V_t} - 1.13 \frac{V_i}{V_t} \right) = 0.35 Sc_L^{0.5} Re_L^{0.17} Gr_L^{0.3} We_L^{0.3} \left(\frac{a_t}{a'_p} \right)^{-0.5} \left(\frac{\sigma_c}{\sigma_w} \right)^{0.14} \quad (4)$$

31
$$\frac{k_G a_{GL}}{D_G a_t^2} \left(1 - 0.9 \frac{V_o}{V_t} \right) = 0.023 Re_G^{1.13} Re_L^{0.14} Gr_G^{0.31} We_L^{0.07} \left(\frac{a_t}{a'_p} \right)^{1.4} \quad (5)$$

1 In order to solve the MERSHQ equations for a rotating packed bed using the non-equilibrium
2 approach in Aspen Plus with a RADFRAC unit, the system is discretized in such a way that it
3 takes into account that the flow, mass and heat transfer occur in the radial direction, and not in
4 the axial direction as the process simulator assumes for a conventional packed column.
5 Different discretization methods have been applied. For instance, Gudena et al.⁹⁴ divided a
6 coaxially oriented horizontal RPB into radial segments, as shown in Figure 11, and used a
7 variable transformation based on the conservation of material flux and volume for differential
8 segments within the RPB to convert the differential annular rings into a series of sequentially
9 attached vertical cylinders. For a detailed explanation of the variable transformation employed
10 by Gudena et al., the reader is remitted to their publication, where the methodology is
11 illustrated with the production of methyl acetate by reactive distillation in a RPB.⁹⁴ The same
12 methodology was also used in their subsequent articles about multiple-objective optimization
13 of an RPB for VOC stripping⁹³, modeling and optimization of HiGee stripper-membrane
14 systems for bioethanol recovery and purification⁹² and for methyl lactate hydrolysis⁹⁵. These
15 multi-objective optimization problems were formulated to maximize solute recovery while
16 minimizing total annual cost.

17 Prada et al.⁹¹ also performed a computational study using the distillation of the ethanol-water
18 system. The simulation was done in Aspen Plus V7 using the rate-based model of the
19 RADFRAC module, with a subroutine in Fortran 11.0 to replace the correlations used by
20 Aspen Plus to calculate the individual mass transfer coefficients with correlations previously
21 developed in the literature for rotating packed beds³⁷. However, it is not clear if they modified
22 the model to account for the fact that transport phenomena do not occur in the same direction
23 in a rotating packed bed than in a conventional packed column. The height equivalent of a
24 theoretical plate in the HiGee was calculated to be 0.0055 m while for the conventional
25 column it was 0.3246 m for the same separation.

26 Joel et al.⁹⁶ followed a similar approach to the one taken by Prada et al. but they used it to
27 model and simulate a HiGee reactive absorber for post-combustion CO₂ capture in
28 monoethanolamine (MEA) instead of distillation. To do so, they modified the rate-based
29 absorber model in Aspen Plus by replacing the default correlations with new ones suitable for
30 RPBs. These correlations were written in Visual Fortran as subroutines and were dynamically
31 linked with the Aspen Plus rate-based absorber model. The model was able to predict the
32 experimental data obtained by Jassim et al.⁹⁹ with a relative error of less than 8% for almost
33 all the variables assessed. The validated model was then used for process analysis of the
34 HiGee absorber in order to gain insights for process design and operation. From their results,

1 Joel et al. predicted a 12-fold size reduction when using a rotating packed bed absorber
2 instead of the conventional packed bed absorber.

3

4 **5. Process design and control**

5 The design of a counter-current HiGee unit differs from that of a conventional packed column
6 in three main aspects: (1) in a HiGee contactor, flow occurs in the radial direction while in a
7 conventional column it occurs in the axial direction, (2) a HiGee contactor has an additional
8 degree of freedom (the rotational speed) and (3) special considerations must be taken into
9 account for proper design of liquid and gas inlets and outlets in HiGee in view of their higher
10 gas-liquid throughputs per unit area¹⁰⁰.

11 Contrary to conventional packed columns, the flow in a HiGee contactor takes place in the
12 radial direction and not in the axial direction. For this reason, separation extent is determined
13 by the radial length of the rotor (the distance between the inner and outer radii) while
14 hydraulic capacity is given by the cross-sectional area at the inner periphery of the rotor
15 (Figure 12).⁵⁰

16 Certain constraints need to be taken into account in order to produce feasible designs. For
17 compactness, the inner radius should be as small as possible, provided that it results in an
18 acceptable exit gas velocity so that the liquid jets emanating from the distributor do not get
19 carried away¹⁰⁰. In addition to that, it should provide enough space to accommodate the liquid
20 distributor while allowing gas withdrawal from the eye of the rotor without excessive pressure
21 drop¹⁰⁰. The axial length, on the other hand, should be such that the unit is operated near but
22 below flooding conditions. Too high of an axial length will result in unwetted packing, and
23 therefore in a rotating packed bed that is bulkier than necessary.¹⁰⁰ Both outer radius and axial
24 length are constrained by mechanical considerations such as bearing loads, vibration moments
25 and by the strength of the packing material and the support basket used to contain the
26 packing.¹⁰¹ Sudhoff et al.¹⁰² provide a list of suggested constraints for rotational speed, inner
27 and outer radii, axial length, number of rotors, superficial gas velocity and pressure drop that
28 can be used during the design of rotating packed beds for distillation.

29 Most of the HiGee contactors reported in the literature have been designed for specific cases
30 and only one systematic design procedure has been reported by Agarwal et al.¹⁰⁰. Sudhoff et
31 al.¹⁰² recently complemented the procedure by including equations to calculate power
32 consumption, required equipment space and investment and operating costs for an RPB for
33 distillation, which is useful for analysis during conceptual process design. A simplified
34 workflow of the complete design methodology is shown in their paper.

1 In the systematic design procedure for HiGee absorption/distillation systems suggested by
 2 Agarwal et al.¹⁰⁰ the authors explain how to calculate the basic design parameters for a
 3 rotating packed bed, which are the inner radius (r_i), the outer radius (r_o) and the axial height
 4 (h). The paper also contains guidelines for proper liquid distributor design, packing material
 5 selection, rotational speed and pressure drop considerations, casing design and power
 6 consumption.

7 The design procedure starts by assuming an actual operating reflux (rectification), boil-up
 8 (stripping) or liquid to gas (absorption) ratio between 1.2 to 1.5 times the minimum ratio that
 9 can be calculated using conventional equilibrium stage techniques (see, e.g., the classic text
 10 by Treybal¹⁰³), fixing this way the gas (vapor)-liquid load inside the RPB. The next step is to
 11 choose an RPB packing material (metal foam, wire mesh, or other) and a rotor type (single
 12 block, split packing) and based on this selection, impose constraints on the maximum
 13 permissible RPB height (h), and outer radius (r_o). After that, a reasonable operating rotational
 14 speed is selected and the inner radius is chosen as the lowest radius that can accommodate the
 15 liquid distributor while allowing gas withdrawal from the eye of the RPB without an
 16 excessive pressure drop and such that the liquid jet to exit gas kinetic energy ratio at the inner
 17 periphery of the rotor is above a suggested value of 3. Agarwal et al.¹⁰⁰ derived equation (6) to
 18 calculate the required inner radius:

$$19 \quad r_{i,min} = \left(\frac{G}{\pi v_{jet} (1 - f_d)} \right)^{\frac{1}{2}} \left(\frac{\rho_g P}{\rho_l} \right)^{\frac{1}{4}} \quad (6)$$

20 After the inner radius has been calculated, the axial length, h , of the rotating packed bed is
 21 then chosen such that process operation at design conditions is only slightly below flooding
 22 for the chosen rotational speed. The superficial gas velocity at an approach to flooding
 23 between 0.70 and 0.90 is calculated with Wallis correlation, whose rearranged version is
 24 shown in equation (7), and is then used to find the RPB axial length as a function of the
 25 volumetric gas flow rate with equation (8).

$$26 \quad U_{G,i} = \left(\frac{\beta N_g^a a_i^b \mu^c (\rho_l - \rho_g)^{\frac{1}{4}}}{\rho_g^{\frac{1}{4}} + \lambda \left(\frac{L}{\alpha G} \right)^{\frac{1}{2}} \rho_l^{\frac{1}{4}}} \right)^2 \quad (7)$$

$$27 \quad h = \frac{G}{2\pi r_i U_{G,i}} \quad (8)$$

28 Where the coefficients and powers (β , a , b , c , λ) in equation (7) vary from system to system.

1 Finally, the outer radius, r_o , is determined by the desired degree of separation. This can be
2 done by performing a material balance over a differential annular shell of the RPB for the
3 primary component and integrating it from the inner radius until the desired degree of
4 separation is achieved. Having now calculated the three most important design parameters,
5 the mechanical robustness of the RPB should be checked. If the maximum outer radius and
6 axial length constraint(s) is (are) violated, the gas and liquid feeds should be split into half
7 and the design is repeated for each half. The liquid distributor is designed for each RPB, the
8 total pressure drop is calculated and the compressor/blower are sized accordingly. The casing
9 liquid sump is sized for a liquid hold up of 1-2 min and the motor(s) power consumption for
10 rotating the RPB(s) is determined. For a more detailed explanation please refer to the original
11 publication by Agarwal et al.¹⁰⁰, where they demonstrate their procedure with four case
12 studies: n-butane/isobutane distillation, benzene-cumene distillation, natural gas dehydration
13 using TEG and CO₂ absorption in DEA, which they compare with their corresponding
14 conventional column distillation processes, estimating total volume reduction factors of 13,
15 10, 68 and 7, respectively¹⁰⁰. It should be noted that the rotating packed beds designed in
16 these case studies had split packing and so the authors used the corresponding hydraulic and
17 mass-transfer coefficient correlations in their procedure.

18 More recently, Sudhoff et al.^{102,104} presented an integrated design methodology for distillation
19 in RPBs, which is based on the procedure presented by Agarwal et al. The method can be
20 used as a tool for feasibility studies for the application of rotating packed beds for distillation.
21 Their integrated design method enables the selection of an appropriate range of operating and
22 design variables to design a highly flexible RPB that can handle a range of feed compositions.
23 For this flexibility analysis, Sudhoff et al. developed a graphical method that uses flexibility
24 maps previously used in other fields but new for chemical processes. They define the degree
25 of flexibility of an RPB as the measure of the range of fluctuations in the feed composition
26 that can still be compensated by varying the rotational speed of the RPB without changing the
27 product specifications. In their methodology, the design parameters (h , r_i , r_o) of an RPB are
28 calculated following a similar approach to the one developed by Agarwal et al.¹⁰⁰ and
29 previously described. However, the design parameters thus calculated are not taken as the
30 optimal design values but rather as initial values, since they do not necessarily offer the
31 biggest flexibility. The effect of variations (both positive and negative) of these initially
32 calculated design parameters on the flexibility of the RPB is studied independently to find
33 new values for h , r_i and r_o that lead to a highly flexible design. All of this, off course, taking

1 into account that the RPB flexibility changes with its dimensions, which have an effect on
2 both investment and operating costs and are finally reflected in the cost per ton of product.
3 Concerning the process control of HiGee, the literature is practically absent on this topic.
4 Because of the high throughput and low inventory, HiGee contactors respond much faster to
5 feed composition variations, and feed flow variations. The control strategy for HiGee
6 distillation should depend on the analysis frequency: if a fast inline measurement of
7 composition is possible (direct by RI/UV-VIS, NMR) then the reboiler and condenser volume
8 should be as low as possible to have a fast response. If a slow “off-line” measurement is done
9 (GC/HPLC), then the condenser and reboiler volume should be higher, to dampen fluctuations
10 and allow for timely control. The first case is obviously desired since the whole idea of the
11 RPB is to reduce volumes and inventories.

12 In the same way, scarce information about process economics of HiGee distillation is
13 available in the literature. Lower investment and operating costs are reported for some of the
14 industrial applications discussed in section 6 of this paper. However, those figures depend not
15 only on the dimensions of the rotating bed, reboiler and condenser, but also on undisclosed
16 variables affecting each specific case. Therefore they cannot always be used for cost estimates
17 of other processes. For a calculation basis for investment and operating costs of rotating
18 packed beds based on similarities, the reader is also referred to Sudhoff et al.¹⁰².

19

20 **6. Industrial applications**

21 To the best of our knowledge, the only reported industrial applications of HiGee for gas-
22 liquid contacting processes are: seawater de-aeration, reactive stripping of hypochlorous acid,
23 SO₂ removal, selective absorption of H₂S, water de-aeration for softdrink bottling and
24 distillation. These applications are summarized in Table 4 and described hereafter.

25

26 **6.1. Seawater de-aeration**

27 The first commercial application of HiGee to be reported was the seawater de-aeration at the
28 Shengli Oil Field of China Petrochemical Corporation¹⁰⁵, where rotating strippers with a 1.4
29 m diameter replaced a 32 m high vacuum tower system. In this plant, two HiGee units with a
30 water throughput per unit of 250 t/h were used to reduce dissolved oxygen in the seawater
31 from 6-12 ppm to less than 50 ppb. The HiGee units replaced a high vacuum tower system
32 that had a lower oxygen removal efficiency and therefore required additional chemical
33 treatment of the water to achieve the desired oxygen levels.

34

6.2. Reactive stripping of hypochlorous acid

In 1999, Dow Chemicals successfully introduced one of the first commercial applications of rotating packed beds, the production of hypochlorous acid (HOCl)^{32,106}. In this reaction, chlorine is absorbed into an aqueous solution of sodium hydroxide and reacts instantaneously to produce sodium chloride and HOCl, the desired product. In the presence of sodium chloride, the HOCl quickly decomposes to sodium chlorate (NaClO₃), an undesired by-product, reducing the product yield. Due to the fast kinetics of both HOCl formation and decomposition reactions, the chlorine absorption-reaction step is liquid-side mass transfer limited while desorption of HOCl is gas-side mass transfer limited. Taking into account the short residence time and high mass transfer rates offered by rotating packed bed technology, Trent and Tirtowidjojo³² designed a process for the production of HOCl through reactive stripping in a rotating packed bed. In this process, the aqueous solution of sodium hydroxide is introduced at the eye of the rotor and then moves radially outwards while contacting the chloride gas stream countercurrently. The HOCl produced is quickly stripped into the gas phase and removed from the reaction zone before it decomposes to chlorate. Thanks to the intensified mass transfer and the low residence time, chlorine absorption-reaction and HOCl desorption both take place quickly, reducing HOCl decomposition and leading to HOCl yields of more than 10% higher than those achieved with the conventional process, which was unable to reach a yield of 80%. Additionally, less than half the amount of stripping gas is necessary while a 40-fold reduction in equipment size is achieved compared with the conventional spray tower technology. In 2003, Trent and Tirtowidjojo³² reported that after two and a half years of operation the rotating packed bed had consistently maintained these and even better yields, while proving to be mechanically reliable. They also stated that start up and shut down was easy and with little maintenance required.

6.3. SO₂ removal

SO₂ is a major pollutant emitted from the combustion of fossil fuels, which is hazardous to human health and contributes to the formation of acid rain. In the ammonia-based wet scrubbing process, which is one of the desulfurization methods most widely applied in China, the absorbent solution containing ammonium sulfite reacts with the SO₂ absorbed, removing it from the acid gas¹⁰⁷. However, due to poor mass transfer efficiency, this process requires large packed columns or spray towers, leading to high capital and operating costs¹⁰⁸. Rotating packed beds can then be used to intensify mass transfer and reduce the size of the columns.

1 In 1999, a rotating packed bed absorber with a capacity of 3000 m³/h of gas was installed at
2 the Zibo Sulphuric acid plant (Shandong province, China) in parallel to the existing tower
3 system for tail gas cleaning of SO₂¹⁰⁹. During the absorption tests, SO₂ concentrations in the
4 tail gas of less than 300 ppm (or even as low as 50ppm)²⁶ were achieved, while capital
5 investment, volume and energy consumption substantially decreased in comparison to the
6 conventional tower system (see Table 4).

7 8 **6.4. Selective absorption of H₂S**

9 H₂S needs to be removed during the gas treating process of refinery gas, syngas or natural
10 gas. However, these gas streams normally contain big amounts of CO₂ that, if co-absorbed,
11 affect the performance of desulfurization process by increasing the solvent circulation rate
12 and the energy load of the solvent regeneration system. Moreover, the CO₂ present in the
13 desorbed acid gas dilutes the H₂S in the feed stream going into the Claus unit, hindering the
14 efficiency of the sulfur recovery system. For these reasons, selective H₂S removal is a subject
15 of high interest for the oil and gas industry⁸⁹.

16 The selective desulfurization process most widely used in refineries is based on the chemical
17 absorption of H₂S in methyldiethanolamine (MDEA)⁸⁹. MDEA is thermodynamically
18 selective towards CO₂ but kinetically selective towards H₂S and, as a result, long gas-liquid
19 contact times in conventional packed columns promote CO₂ co-absorption. Therefore, the
20 combination of low residence time and high mass transfer rates offered by rotating packed
21 beds favor the selective absorption of H₂S⁸⁹. Fujian Petroleum Refinery Co. (China) installed
22 a rotating packed bed to replace a packed bed column for the selective absorption of H₂S over
23 CO₂ using MDEA as a solvent. As a result, the CO₂ co-absorption was reduced from 79.9% to
24 8.9% while the equipment volume drastically reduced from 36 m³ to 3.4 m³. Details are
25 shown in Table 4.

26 27 **6.5. Water de-aeration for soft drink bottling**

28 In 2006, GasTran installed their first RPB vacuum de-aeration system at PepsiAmericas to
29 remove dissolved oxygen from water and thus reduce foaming and increase bottling line
30 speed and product quality in their carbonated soft drink bottling process¹¹⁰. In the GasTran
31 vacuum de-aeration system water is fed through the center of the rotor, in which very small
32 water droplets with a large specific surface area are produced and exposed to the vacuum,
33 allowing gas desorption and removal to occur. The de-aerated water is collected and exits
34 through the bottom of the casing while the dissolved gases are desorbed and exit through the

1 top of the bed towards a vacuum pump. No stripping gas is required to achieve dissolved
2 oxygen levels in the 200 to 500 ppb DO range. GasTran reported increased filling speeds
3 ranging from 10% to 40%, improvement in the distribution of carbonation levels and
4 increased fill accuracy resulting in less variation in net contents of the final product and in the
5 reduction of reject rates due to low fills. PepsiAmericas purchased their second GasTran
6 Vacuum Deaeration System in early 2008, confirming thus the success of the first system.

7 8 **6.6. HiGee distillation**

9 Besides the aforementioned applications, Wang et al.²⁷ reported the commercialization of
10 about 200 rotating zigzag units up to 2011, for the separation of alcohol/water, acetone/water,
11 DMSO/water, DMF/water, ethyl acetate/Water, methanol/tert-butanol, dichloromethane/silyl-
12 ethers, methanol/formaldehyde/water, methanol/toluene/water, ethyl acetate/toluene/water,
13 methanol/methylal/water, methanol/DMF/water. Table 5 shows a list of commercial suppliers
14 of HiGee technology for distillation, absorption and other processes.

15
16 It must be highlighted that no industrial applications involving heterogeneous catalysis, i.e.,
17 solid-catalyzed reactions or catalytic distillation in HiGee, have been reported so far. This is
18 not surprising since most experimental work has focused on gas-liquid mass transfer while
19 very few⁶⁷⁻⁷¹ on effect of high gravity on liquid-solid mass transfer.

20 This list of applications just described is very limited considering the large amount of studies
21 (see Table 1) that have been conducted to demonstrate the functionality of HiGee contactors
22 for stripping, distillation, absorption, homogeneous gas-liquid reactions and even solid-
23 catalyzed gas-liquid reactions. The main reason for the slow deployment of these technologies
24 in the industry still seems to be the concerns about the energy use and reliability of these
25 rotating machines, even though the rotating speeds at which they operate is closer to that of
26 pumps and fans than that of high speed centrifuges¹¹¹, all of which are widely used in the
27 industry. The main drawbacks of HiGee technology include: rotating equipment, additional
28 energy requirements (electricity), problems related to the reliability and longevity due to the
29 moving parts, mechanical stability issues, complicated hydrodynamics, seals and bearings.

30 31 **7. Conclusions**

32 This review paper provided an overview of the state-of-the-art in the field of HiGee
33 contactors used for gas-liquid mass transfer processes. Different HiGee contactors have been
34 discussed with respect to their advantages and disadvantages, rotor configuration, working

1 principles, modeling and simulation, design procedures and practical applications. The RZB
2 seems to have the best performance for continuous distillation when compared to other
3 rotating beds. This is mainly due to its higher gas-liquid contact time and to the possibility of
4 increasing the number of separation stages by installing multiple rotors within one casing.
5 However, the single-block RPB has a much simpler rotor and provides a high surface area
6 that can be used for catalyzed reactive systems.

7 The literature available confirms that HiGee contactors have been used for several gas-liquid
8 processes, such as absorption, stripping and distillation. Besides, while only few studies have
9 incorporated solid catalysts into the packing⁶⁷⁻⁷¹, they have reported favorable results in
10 comparison to conventional processes. This shows the potential that HiGee has for solid-
11 catalyzed gas-liquid reactions⁶⁷⁻⁷¹, not only due to the intensified gas-liquid mass transfer
12 rates but also to the potentially good catalyst wettings at high centrifugal forces. Most of these
13 studies⁶⁷⁻⁷¹ have addressed specific applications but little work has been done to gain
14 fundamental knowledge about solid-catalyzed reactions under high centrifugal forces. One of
15 the main issues that has to be addressed during future research is the very low liquid holdup
16 of HiGee contactors, which reduces the residence time in reactive systems.

17 Another deficiency in the field is the lack of clear scale-up rules. The variation in flow area
18 and centrifugal force in the radial direction of a HiGee make the average mass transfer
19 coefficients (usually reported in literature) not very useful for industrial scale-up. The use of
20 these data is even more complicated when important information about the type of distributor
21 and packing used and the operating conditions at which experiments were conducted is not
22 disclosed in research articles.

23 Despite the considerable amount of research carried out so far, most of it has not exploited the
24 full potential of HiGee. Operation at high superficial velocities at a laboratory scale requires
25 such small HiGee units that they can be far from representing industrial scale rotating beds. A
26 trade-off then exists between reaching the maximum capabilities of rotating beds and
27 obtaining scalable results. This trade-off could be eliminated by having more involvement
28 from the industry in HiGee research, making it possible to run experiments in production
29 sites, enabling operating conditions not reachable in a research laboratory.

30 There are also other issues and challenges ahead that need to be solved, such as: mechanical
31 complexity and reliability, uncertainty in design data, use of catalytic reactions, reliability at
32 high/low pressure/temperature, corrosion resistance of packing materials, dynamic balance of
33 large rotating packed beds, limitation in the number of stages, sensitivity towards initial liquid
34 distribution, limitations in scale of operation (i.e. throughput).

1 On the other hand, based on the amount of experimental work done and on the list of
2 industrial applications reported in the literature, HiGee technology seems to have already its
3 place in China while it has been relegated in Western countries, where it was first developed.
4 Concerns about the reliability of rotating packed beds may be one of the reasons for this lack
5 of interest, even though Dow Chemicals has reported the successful long-term operation of
6 their RPBs for the production of HOCl and the fast and simple startup of these machines to
7 perform preventive and corrective maintenance when required.

8 The lack of simulation modules in commercial process simulators is another matter that
9 deserves attention. Without these modules, it is difficult to assess the performance of HiGee
10 technology during conceptual design phases and feasibility studies. This may keep companies
11 from realizing potential benefits of HiGee technology for their processes, resulting in new
12 plants being designed with the same conventional units that have already proven to work for
13 decades instead of taking risks with novel technology.

14 The process industry could benefit from HiGee in several ways. For instance, by having
15 inherently safer designs (with reduced inventories) and by gaining flexibility (due to much
16 lower start-up times that make HiGee technology convenient for production campaigns), as
17 well as switching from batch to continuous processes (without significant production loss).
18 Collaboration between the private sector and academia is then required to develop this
19 promising technology and prove its benefits and long-term reliability in order to overcome the
20 obstacles for its deployment in the industry.

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24 AkzoNobel and the Department of Chemical Engineering and Chemistry from Eindhoven
25 University of Technology – where 12 PhD students will work on various aspects and
26 applications of HiGee technologies.

27 28 29 **Nomenclature**

30 31 Symbols

32 a	Total specific area of mass transfer, $m_i^2 m_R^{-3}$
33 a_{GL}	Gas-liquid specific interfacial area, $m_i^2 m_R^{-3}$
34 a_{LS}	Liquid-solid specific interfacial area, $m_i^2 m_R^{-3}$

1	a_d	Specific area of the droplets, $m_i^2 m_R^{-3}$
2	a_f	Specific (wetted) area of the packing, $m_i^2 m_R^{-3}$
3	a'_p	Surface area per unit volume of the 2 mm diameter bead, $m^2 m^{-3}$
4	a_t	Specific area of the packing, $m^2 m^{-3}$
5	ATU	Area of a transfer unit, m^2
6	C	Constant in Onda equation
7	C_{cat}	Catalyst concentration, $mol_{cat} m^{-3}$
8	D_G	Gas diffusivity, $m^2 s^{-1}$
9	d_p	Diameter or characteristic dimension of packing, m
10	f_d	Fraction of cross-sectional area of RPB eye occupied by distributor(s)
11	$F - factor$	Vapor kinetic energy term, defined by $U_G \rho_G^{1/2}$, $m/s (kg/m^3)^{1/2}$
12	G	Volumetric gas flow rate, m^3/s
13	Gr_G	Gas Grashof number, defined by $Gr_G = \frac{d_p^3 \rho_G^2 a_c}{\mu_G^2}$
14	Gr_L	Liquid Grashof number, defined by $Gr_L = \frac{d_p^3 \rho_L^2 a_c}{\mu_L^2}$
15	h	Axial height of the bed packing, m
16	H	Henry constant of a gas in a liquid, $Pa m^3 mol^{-1}$
17	HETP	Height equivalent to a theoretical plate, cm
18	k_G	Gas-side (gas-liquid) mass-transfer coefficient, $m_G^3 m_i^{-2} s^{-1}$
19	$k_G a_{GL}$	Volumetric gas-side mass transfer coefficient, $m_G^3 m_R^{-3} s^{-1}$
20	k_L	Liquid-side (gas-liquid) mass-transfer coefficient, $m_L^3 m_i^{-2} s^{-1}$
21	$k_L a_{GL}$	Volumetric liquid-side mass transfer coefficient, $m_L^3 m_R^{-3} s^{-1}$
22	k_{L_1}	Mass-transfer coefficient in the droplet, $m_L^3 m_i^{-2} s^{-1}$
23	k_{L_2}	Mass-transfer coefficient in the film, $m_L^3 m_i^{-2} s^{-1}$
24	k_S	Liquid-solid mass-transfer coefficient, $m_L^3 m_i^{-2} s^{-1}$
25	$k_S a_{LS}$	Liquid-solid mass-transfer coefficient, $m_L^3 m_R^{-3} s^{-1}$
26	$K_G a$	Overall volumetric mass-transfer coefficient, $m_G^3 m_R^{-3} s^{-1}$
27	k_r	Reaction rate constant, $m^3 mol_{cat}^{-1} s^{-1}$

1	L	Liquid flow rate, $\text{m}^3 \text{s}^{-1}$
2	N_g	Ratio of centrifugal to gravitational acceleration, $\frac{\omega^2 r_i}{g}$
3	p	Ratio of liquid jet to exit gas kinetic energy, -
4	R	Gas constant, $\text{m}^3 \text{Pa mol}^{-1} \text{K}^{-1}$
5	Re_L	Gas Reynolds number, defined by $\frac{L}{a_i \mu_L}$
6	Re_G	Gas Reynolds number, defined by $\frac{U_G \rho_G}{a_i \mu_G}$
7	r_i	Inner radius of the bed packing, m
8	r_o	Outer radius of the bed packing, m
9	Sc_L	Liquid Schmidt number, defined by $\frac{\mu_L}{\rho_L D_L}$
10	Sc_G	Gas Schmidt number, defined by $\frac{\mu_G}{\rho_G D_G}$
11	U_G	Gas superficial velocity, $\text{m}_G^3 \text{m}_R^{-2} \text{s}^{-1}$
12	$U_{G,i}$	Gas superficial velocity at RPB inner radius, $\text{m}_G^3 \text{m}_R^{-2} \text{s}^{-1}$
13	v_{jet}	Liquid distributor jet velocity, m s^{-1}
14	V_i	Volume inside the inner radius of the bed, m^3
15	V_o	Volume between the outer radius of the bed and the stationary housing, m^3
16	V_t	Total volume of the RPB, m^3
17	We	Webber number, defined by $\frac{L^2}{\rho_L a_i \sigma}$
18	<i>Abbreviations</i>	
19	BP-RPB:	Blade-packing rotating packed bed
20	CD:	Corrugated disk
21	CM:	Cross meshwork
22	CRB:	Cross-flow concentric-baffle rotating bed
23	CFCR-RB:	Cross-flow concentric ring rotating bed
24	DEA:	Diethanolamine
25	HiGee:	High-gravity
26	MDEA:	Methyldiethanolamine

1	MEA:	Monoethanolamine
2	MERSHQ:	Material balances, energy balances, summation equations, hydraulic equations
3		and equilibrium equations
4	NEQ:	Non-equilibrium
5	ppm:	Parts per million
6	RPB:	Rotating packed bed
7	RZB:	Rotating zigzag bed
8	SP-RPB:	Rotating packed bed with split packing
9	SS:	Stainless steel
10	TEG:	Triethylene glycol
11	TSCC-RPB:	Two-stage counter-current rotating packed bed
12	VOC:	Volatile organic compounds
13	WM:	Wire mesh
14	WT:	Wave thread
15		
16	<i>Greek letters</i>	
17	α, β, λ	RPB flooding correlation fitting parameters
18	ϵ_p	Porosity of packing, -
19	η	Effectiveness factor, -
20	ρ	Density, kg m^{-3}
21	μ	Fluid viscosity, $\text{kg m}^{-1} \text{s}^{-1}$
22	σ	Surface tension, kg s^{-2}
23	σ_c	Critical surface tension of packing, kg s^{-2}
24	σ_w	Surface tension of water, kg s^{-2}
25	ω	RPB rotational speed, rad s^{-1}
26		
27	<i>Subscripts</i>	
28	a, b, c	RPB flooding correlation fitting parameters
29	G	Gas
30	L	Liquid

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29

1 Tables

2 **Table 1.** Comparison of HiGee gas-liquid contactors found in the literature

Equipment	Advantages	Disadvantages	Application
Single block Rotating Packed Bed (Counter-current flow)	Simple single-block rotor design with a higher mechanical strength than other rotors. Higher mass transfer performance than conventional packed bed columns. Single-block rotor offers a high specific surface area for gas-liquid contact.	Little or no increase in the gas-side mass-transfer coefficient with respect to fixed bed column ⁵⁹ . Coaxial multirotor structurally complicated ⁴⁰ . Middle-feed streams difficult to be inserted. Multiple RPBs needed for continuous distillation. Very short liquid residence time ⁴⁰ . Requires liquid distributors. ^{63,83}	Stripping ^{1,39,52,112-119} Distillation ^{1,120,66,121,122} Absorption ^{1,39,53,59,85,86,99,108,123-129} Homogenous gas-liquid reactions ^{84,130-134} Solid catalyzed gas-liquid reactions ⁶⁷⁻⁷¹
Single block Rotating Packed Bed (Cross-current flow)	Simple single-block rotor. Capable of handling higher gas flow rates than counter-current flow RPB due relaxed flooding limitations ⁵⁵ . Lower pressure drop than counter-current RPB ^{57,58} Mass transfer performance comparable to that of an RPB ⁵⁷	Same disadvantages of counter-current flow RPB	Absorption ^{55,57,135,136}
Multi-stage spraying rotating packed bed	Liquid is sprayed into fine droplets with a large interfacial area due to centrifugal force ⁴⁰ .	Difficult to coaxially install multiple rotors in one casing ⁴⁰ Potential liquid entrained by gas flow despite lower gas frictional resistance ⁴⁰	N/A
Rotating packed-bed with wave-form disk packing	Lower gas flow resistance than RPB ⁴⁰	Difficult to coaxially install multiple rotors in one casing ⁴⁰	N/A
Helical rotating bed	Longer residence times for both gas and liquid than in an RPB ⁴⁰	Mass transfer performance lower than that of an RPB due to the small interfacial area ⁴⁰ Difficult to coaxially install multiple rotors in one casing ⁴⁰	Absorption ⁴⁰
Rotating packed bed with split packing	Higher tangential slip velocities which may result in higher gas-side mass transfer coefficients ^{61, 100} . Flooding limits comparable to other Hige devices while a_e and k_{La_e} are higher ³⁷ .	More complex design than single block RPB with two rotors driven by two independent motors ³⁹ . Maximum rotor size limited by mechanical strength of packing ³⁹ . Middle-feed streams difficult to be inserted ^{62,61} . Coaxial multirotor structurally complicated. Requires liquid distributor ⁶¹	Stripping ^{39,61,62} Distillation ³⁸ Absorption ^{37,39,62,137}
Rotating zigzag bed	Mid-feed streams can be inserted ^{27,40,41} . Multiple rotor configuration in one casing without dynamic seals ^{27,40,41}	Higher pressure drop than RPB. Higher pressure drop compared to the conventional RPB and power consumption than RPB ^{40,41,64} . Lower surface area for the gas-	Distillation ^{40,41} Absorption ⁴⁰ Stripping ^{138, 139} Reaction ⁴⁰

	Higher liquid-gas contact time than RPB ⁴¹ . No liquid distributors needed ^{27,40,41} . Equivalent mass transfer performance to RPB. Higher turndown ratio than a RPB ⁴⁰ .	liquid contacting than RPB ^{41,40,30} .	
Two-stage counter-current rotating packed bed	Can be filled with packing while admitting intermediate feeds for continuous distillation ³⁰ . Mass transfer performance comparable to that of the RPB ³⁰ . More compact device than two combined RPBs ³⁰ . Higher number of theoretical plates per meter than in the RZB and the packed column ³⁰ . Lower pressure drop than RZB ³¹ .	Complex rotor structure with a combination of rotational packing and static rings ³⁶ . Lower number of theoretical plates per meter under some operation conditions compared with two combined RPBs ³⁰ .	Distillation ^{30,31} Absorption ³⁶ Reactive distillation ⁷²
Blade packing rotating packed bed	Lower gas pressure drop than RPBs ⁴⁵ . HTU values (1-3cm) comparable to those of RPBs ⁴⁵ .	Low specific surface area ⁶⁵ . Not suitable for solid-catalyzed reactions.	Stripping ^{42,43,65,140,141} Absorption ⁴⁶
Rotating bed with packing and blades	Higher mass transfer performance than RPB due to artificially created end-zones ⁴⁴ . Higher specific surface area than other rotating beds with blades.	Structure of rotor is more complex than conventional RPB rotor ⁴⁴ . Lower specific surface area than RPB and other rotating beds without blades. Middle-feed streams difficult to be inserted.	Distillation ⁴⁷ Absorption ⁴⁴
Rotating bed with blade packing and baffles	Higher slip velocity than RPB ⁴⁵ . Lower pressure drop than in an RPB and a rotating bed with blade packings ⁴⁵ . Higher volumetric gas-side mass transfer coefficients than in a rotating bed with blade packings ⁴⁵ .	Lower specific surface area than RPB and other rotating beds without blades. Middle-feed streams difficult to be inserted.	Absorption ^{45,142} Stripping ¹⁴²
Counter-flow concentric-ring rotating bed	Higher gas-liquid throughputs than RZB (5.6 times) ⁴⁸ . Lower pressure drop compared with RZB due to the elimination of the stationary baffles ⁴⁸ . Higher gas-side mass transfer coefficient compared to RPB ⁴⁸ . Middle-feed streams can be inserted. Multirotors can be coaxially installed in one casing.	Lower mass-transfer performance compared with RZB ⁴⁸ . Much lower gas-liquid effective interfacial area compared to RPB ⁴⁸ .	Distillation ⁴⁸
Cross-flow concentric-baffle rotating bed (CRB)	Lower pressure drop and lower shaft power than RZB ⁴⁹ . Little backmixing.	Stage efficiency of the CRB one-third as much as that of the RZB, or even lower ⁴⁹ .	Distillation ⁴⁹

1 **Table 2.** Distillation studies in HiGee contactors

System/Type of bed	Dimensions (r _i , r _o , h) (m)	Liquid flow, gas flow	Packing type, <i>a</i> (m ² /m ³), ε _p	Rotor speed (rpm)	Mass transfer performance (HETP, cm), (ATU, m ²), mass transfer coeffs.	Reference
Methanol/Ethanol (RPB/ Tot. reflux)	(0.06, 0.09,-)	-, 8.42x10 ⁻³ - 8.6x10 ⁻³ mol/m ² s	SS gauze, 1650, -	1600	$K_G = 5.4 \times 10^{-5} - 44 \times 10^{-5}$ mol/m ² s $K_G a = 0.034 - 0.72$ mol/m ³ s	(Ramshaw and Mallinson, 1981) ¹
Ethanol/isopropanol (RPB)	(0.4, -, -)	-, -	-, -, -	1500-3000	HETP=1.75 - 2.0	(Short, 1983) ¹⁴³
Cyclohexane/n-heptane (RPB/ Tot. reflux)	(0.0875, 0.30, 0.15)	3.75 L/s, -	Metal sponge-like, 2500, 0.92 Rectangular packing, 524, 0.533	400-1200	HETP=3.50 - 7.50 ATU=0.04 - 0.13	(Kelleher and Fair, 1996) ¹²⁰
Methanol/Ethanol (RPB/Tot. reflux)	(0.0305, 0.074, (0.05,0.095))	-	SS WM, 982, 0.971	600-1600	HETP=3.0 - 9.0	(Lin et al., 2002) ¹²¹
Ethanol/water (RZB/cont. distill.)	(0.2, 0.63, 0.08)	-	Concentric baffles, -, -	600-1400	HETP=2.9 - 5.4	(Wang et al., 2008b) ⁴¹
Methanol/water (RZB/cont. distill.)	(0.1, 0.52, 0.078)	-	Concentric baffles, -, -	800-2000	HETP=4.0 - 5.0	(Wang et al., 2008a) ⁴⁰
Ethanol/water (RPB/cont. distill.)	(0.03, 0.055, 0.063)	0 - 30 L/h,-	SS CD, 400, 0.82 SS CM, 1750, 0.86 SS WT, 350, 0.95	0-1830	HETP=1.34 - 2.54 HETP=1.46 - 2.50 HETP=1.02 - 2.36	(Li et al., 2008) ¹²²
n-hexane/n-heptane (RPB/ Tot. reflux)	(0.022, 0.08, 0.04)	5.7 - 29.0 cm ³ /s,- 5.7 - 29.0 cm ³ /s,- 2.0 - 18.5 cm ³ /s,-	Raschig rings, 627, 0.62 Raschig rings, 765, 0.55 SS WM, 2100, 0.74	300-2500	ATU=0.013 - 0.027 ATU=0.012 - 0.026 ATU=0.012 - 0.042 $K_G a = 340$ mol/m ³ s	(Nascimento et al., 2009) ⁶⁶
Acetone/water (TSCC-RPB/ cont. distill.)	(0.0725, 0.178, 0.046)	-	SS WM, 670, 0.96	400-1200	HETP=1.73 - 4.05	(Luo et al., 2012) ³⁰
Methanol/Ethanol (SP-RPB/ Tot. reflux)	(0.03, 0.155, 0.027)	-	SS WM, 280,-	600-1550	HETP=2.9 - 15.0 ATU=0.02 - 0.1 m ²	(Mondal et al., 2012) ³⁸

Methanol/water (TSCC-RPB/ cont. distill.)	(0.0725, 0.183, 0.058)	10 L/h feed	SS WM, 670, 0.96 Porous sheet: 4mm thickness Packing ring: 16mm thickness	400- 1000	HETP=3.12 - 4.72	(Chu et al., 2013) ³¹
Ethanol/water (CFR-RB/ Tot. reflux)	(0.07, 0.136, 0.015, 0.045)	-	Concentric baffles,-,-	800- 1400	HETP=3.0 - 6.5 $K_G a = 3.18 - 92.30s^{-1}$ $K_G = 0.1 - 0.6$ m/s $k_G = 0.1 - 1.1$ m/s $k_L = 0.001 - 0.002$ m/s	(Li et al., 2014) ⁴⁸
Ethanol/water (CRB/ Tot. reflux)	(0.06, 0.117, 0.08)	-	Concentric baffles,-,-	400- 1200	Stage efficiency 10 -15%	(Wang et al., 2014) ⁴⁹
Methanol/water (TSBP/ cont. distill.)	(0.064, 0.174, 0.040)	20 - 140 L/h, 1600 - 8000L/h	Packing rings (SS WM, 500 m^2/m^3 , 0.96) and blade rings.	600- 1400	HETP=1.9 - 10.0 $k_L = 0.0005 - 0.0019$ m/s $a = 93 - 337$ m^2/m^3	(Luo et al., 2016) ⁴⁷

1 **Table 3.** Overview of works published on RPB modeling with a first-principles approach

Application	RPB config.	Model description	G-L Mass transfer model	Liquid flow (assumed)	References
Chemical absorption of CO ₂ in NaOH and dissolution of naphthalene in water	Counter-flow	Gas-liquid and liquid-solid mass transfer	Penetration theory and convection-diffusion model	Film flow	Munjal et al. ^{82,123}
Selective H ₂ S absorption in MDEA	Counter-flow	Reaction-equilibrium-mass transfer	Penetration theory	Film flow	Quian et al. ^{89,90}
CO ₂ absorption in MDEA	Counter-flow	Diffusion-reaction mass-transfer	Penetration theory	Film flow	Zhang et al. ¹⁴⁴
Absorption of SO ₂ into aqueous NaOH and stripping of O ₂ from water	Counter-flow	Mass transfer coefficients	Two-film theory	N/A	Shivhare et al. ³⁹
CO ₂ absorption by Benfield solution	Counter-flow	Gas-liquid mass transfer with reaction	Two-film theory	Droplet flow	Yi et al. ⁸⁶
Simultaneous absorption of CO ₂ and NH ₃ into water	Counter-flow	Gas-liquid mass transfer	Two-film theory	Film and droplet flow	Sun et al. ⁸⁵
Ozonation of pollutant (o-Cresol)	Counter-flow	Gas-liquid mass transfer with reaction	Two-film theory	Film flow	Chen et al. ⁸⁴
Absorption of volatile organic compounds (VOCs) into water	Cross-flow	Gas-liquid mass transfer	Surface renewal theory	Film and droplet flow	Chen et al. ⁵⁵
Water deaeration, NH ₃ absorption and SO ₂ chemical absorption	Cross-flow	Gas-liquid mass transfer (reaction)	Surface renewal theory	Film and droplet flow	Guo et al. ⁵⁶
Solid catalyzed reactive stripping	Counter-flow	Gas-liquid mass transfer with solid catalyzed reaction	Two-film theory	Film flow	Gudena et al. ⁷⁹

1
2 **Table 4.** List of major industrial applications of HiGee gas-liquid contactors reported in the
3 literature

Company	Application	Capacity	Year	Remarks	Ref.
China Petrochemical Corporation/ Shengli Oil Field Co., China	Seawater deaeration	Two units of 250 t/h	1998	<ul style="list-style-type: none"> Investment cost 40% lower. Ground space reduced by 60%. Equipment weight reduced by 80%. Improved oxygen removal efficiency. 	105
The Dow Chemical Company, USA	Reactive stripping of HOCl	150 t/h.	1999	<ul style="list-style-type: none"> Yield 10% higher. Less than half stripping gas. Investment cost 70% lower. Operating cost 30% lower. Reactor volume 40% smaller. 	32,106,1 45
Zibo Sulphuric acid plant (Shandong province, P. R. China),	SO ₂ absorption from tail gas of SO ₃ absorber.	3000 m ³ /h of gas	1999	<ul style="list-style-type: none"> Degree of equilibrium absorption close to 100%. Compared with conventional tower, capital investment reduced by 35%. Volume reduced by 50%. Energy consumption reduced by 25%. 	26,146, 147
Fujian Petroleum Refinery Co., China	H ₂ S/CO ₂ selective absorption in MDEA	11 t/h (feed gas)		<ul style="list-style-type: none"> CO₂ co-absorption reduced from 79.9% to 8.9%. Equipment volume reduced from 36.1 m³ to 3.4 m³. Packing volume reduced from 14 m³ to 0.3 m³. Steady state reached in few minutes. Lower power requirement than packed bed. 	89,105
PepsiAmericas Plant, USA	Water deaeration for soft drink bottling	N/A	2006, 2008	<ul style="list-style-type: none"> Lower dissolved oxygen and CO₂ levels (from 8.1 ppm to 0.3-0.6 ppm, at 9-6°C) without stripping gases. Annual savings (Indianapolis plant) of (\$88,000). Filling speeds increased by 10%-40% Improvement in the distribution of carbonation levels. Increased fill accuracy. 	110,148

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1 **Table 5.** List of suppliers of HiGee equipment.

Company	Country	Description	Website/contact info
Hangzhou Huadong Chemical Equipment Industrial Co., Ltd.	China	Rotating zigzag beds for distillation.	www.hzglb.com
GasTran Systems LLC	USA	Rotating packed beds for water deaeration.	www.gastransfer.com
Wenzhou Jinzhou Group International Trading Corporation	China	Rotating packed beds for HiGee distillation.	www.jzmachinery.com
SolFirst Technologies India Private Limited	India	Single block and split packing RPBs and zigzag rotating beds for distillation.	www.solfirsttechnologies.com
Suzhou HiGee Environment & Energy Technology Co. Ltd. (In USA as HiGee Environment & Energy Technology Inc.)	China	Rotating packed beds for acid removal, off-shore gas processing, water deoxygenation, devolatilization of polymers, extraction, wastewater treatment, preparation of nanoparticles, polymerization reactions.	www.higeetech.com www.higeusa.com
Zhejiang Chuangxing Chemical Equipment Co., Ltd	China	Hige distillation equipment. Technical services, installation and tuning.	www.cx-hige.com

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1 **Figure captions**

2

3 Figure 1. Working principle of HiGee distillation in a rotating packed bed (RPB)

4

5 Figure 2. Schematic drawing of a single-block rotating packed bed: (top) Counter-current
6 flow RPB, (btm) cross-current flow

7

8 Figure 3. Schematic drawing of a split-packing rotating packed-bed

9

10 Figure 4. Schematic drawing of the rotor of the RZB

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12 Figure 5. Schematic drawing of the rotor (one stage) of a TSCC-RPB

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14 Figure 6. Arrangement of blade packings in RPB

15

16 Figure 7. Schematic drawing of RPB with packing and blades (top) Rotor design; (btm) main
17 structure of the RPB

18

19 Figure 8. Schematic drawing of blades and baffles rotating bed; (left) blade packings on the
20 rotational disk and baffles on the stationary disk; (right) structure of the RPB with blade
21 packings and baffles.

22

23 Figure 9. Schematic drawing of a counter-flow concentric-ring rotating bed

24

25 Figure 10. Schematic drawing of a crossflow concentric-baffle rotating bed (CRB)

26

27 Figure 11. Schematic drawing showing the method followed by Gudena et al.⁹⁵ to
28 approximate HiGee (left sketch) as a conventional column (right sketch). Continuous arrows:
29 vapor flow, dashed arrows: liquid flow

30

31 Figure 12. Design analogy between conventional and HiGee distillation

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Working principle of HiGee distillation in Rotating Packed Bed (RPB)

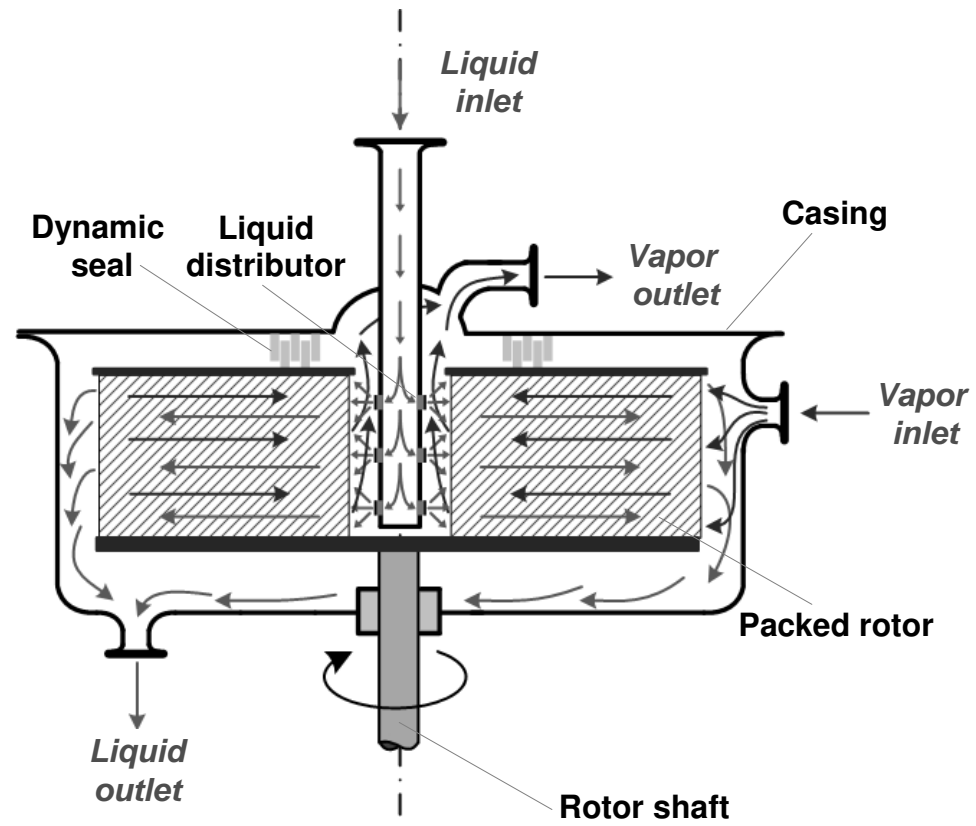


Figure 1

Schematic drawing of a single-block rotating packed bed:
(top) counter-flow RPB, (btm) cross-flow

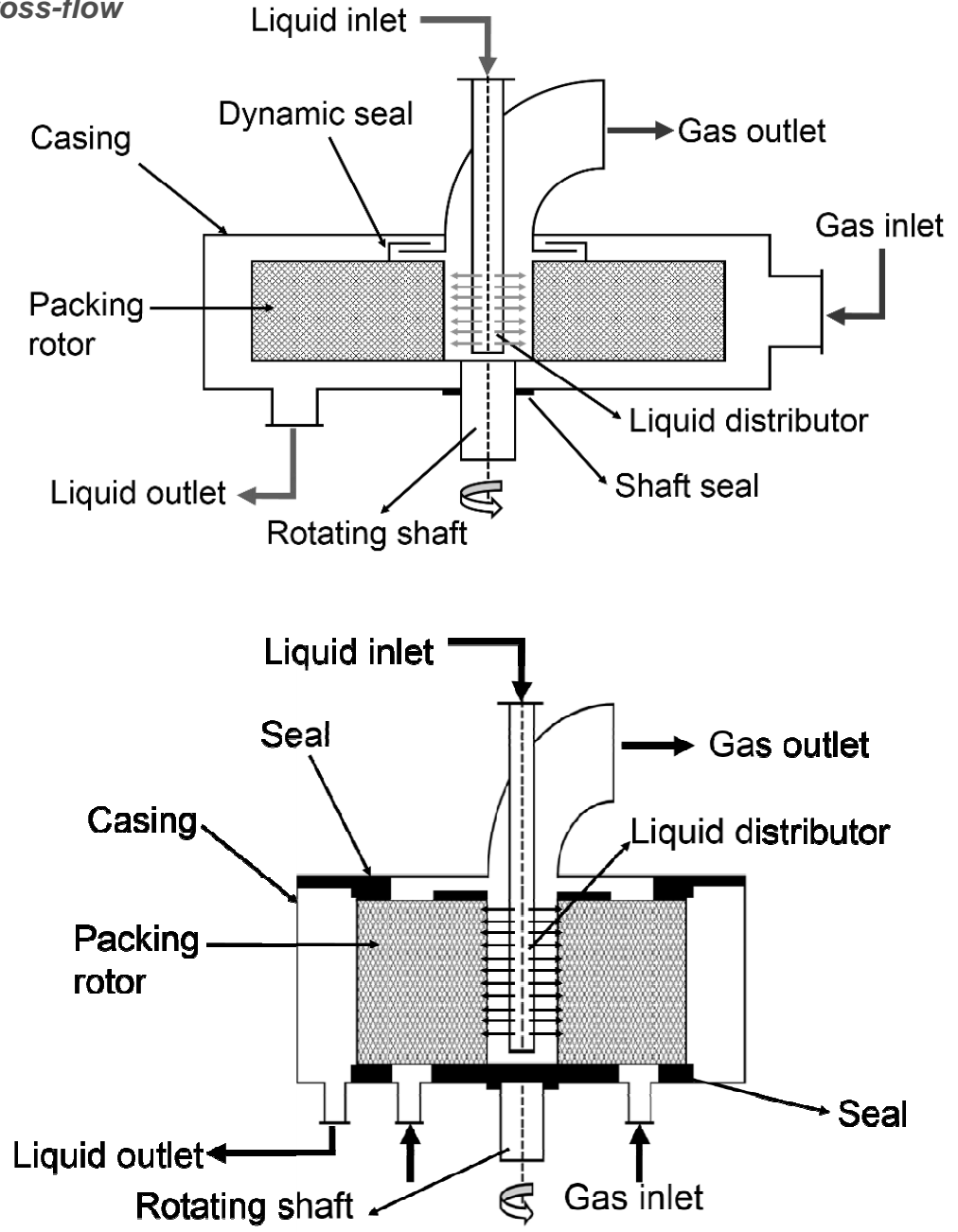


Figure 2

Schematic drawing of a split-packing rotating packed-bed

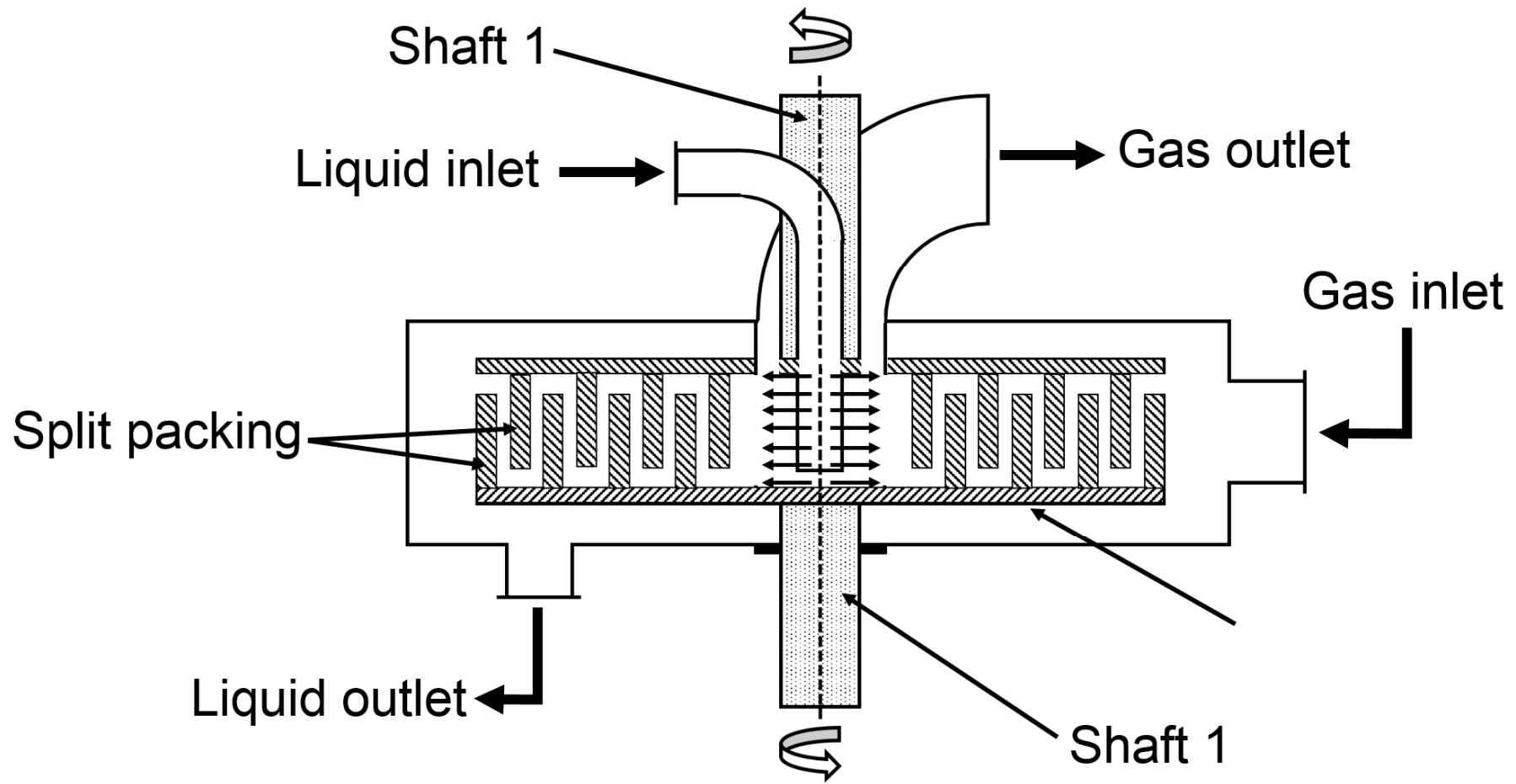


Figure 3

Schematic drawing of the rotor of the RZB

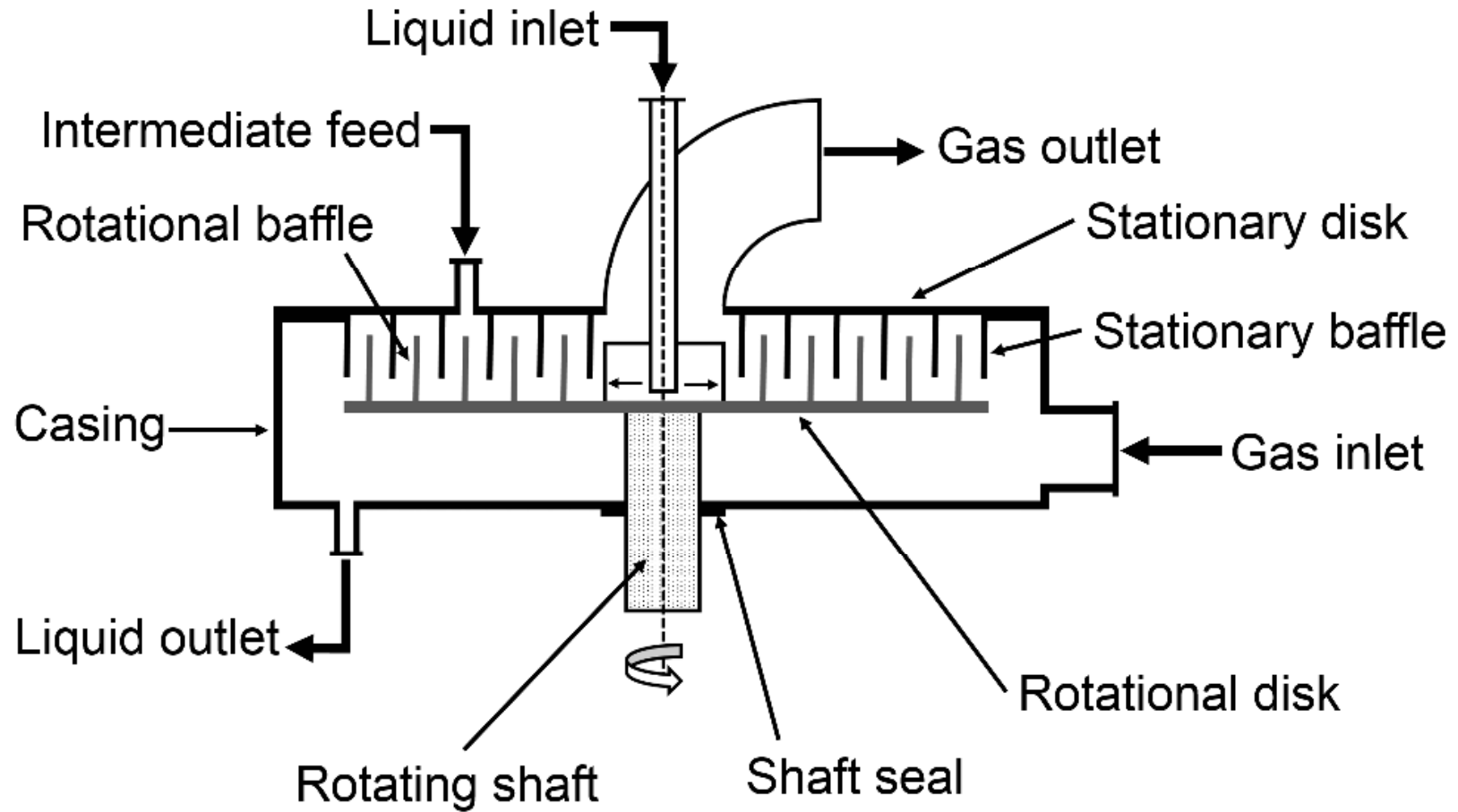


Figure 4

Schematic drawing of the rotor (one stage) of a TSCC-RPB

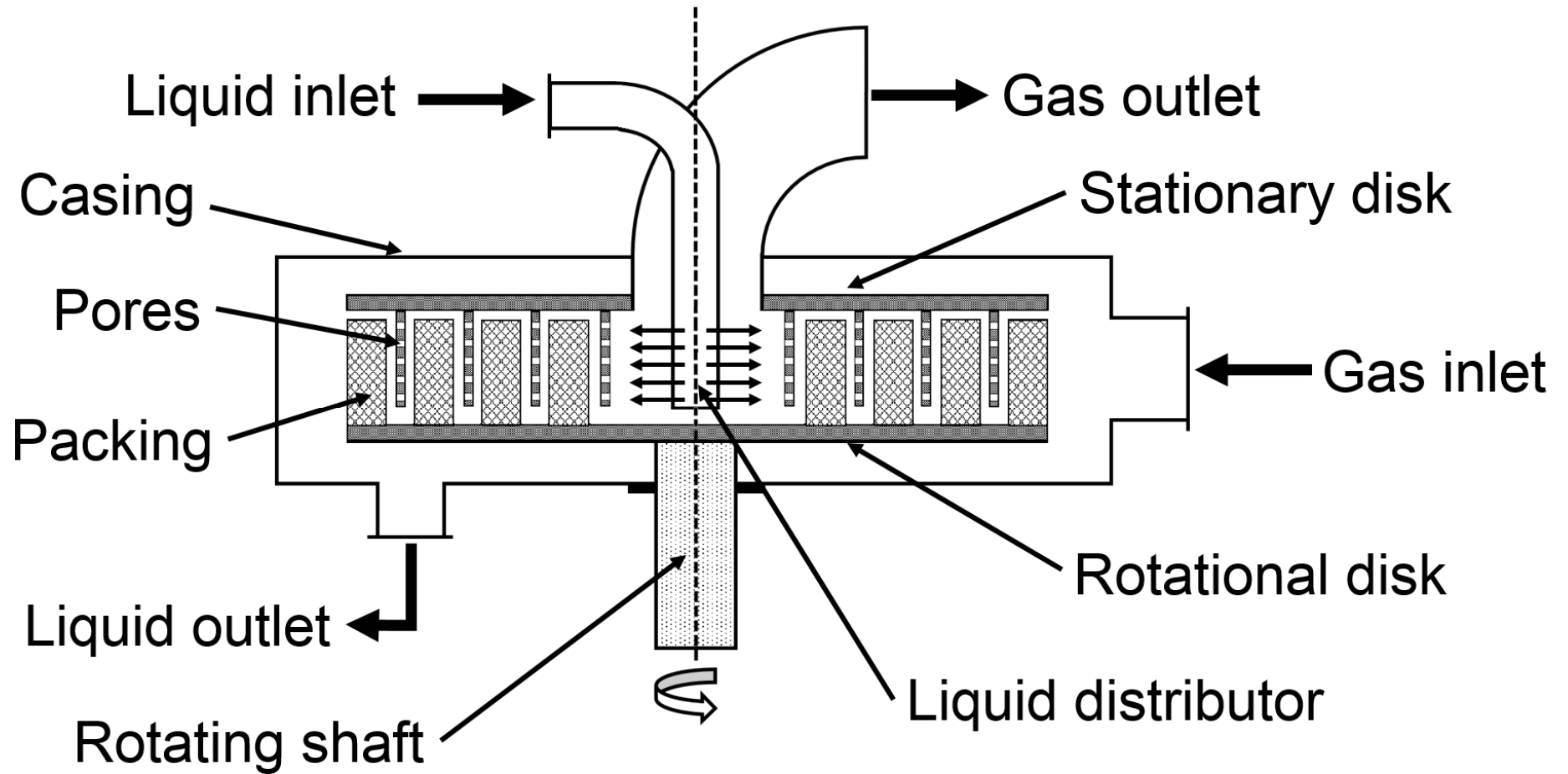


Figure 5

Arrangement of blade packings in RPB

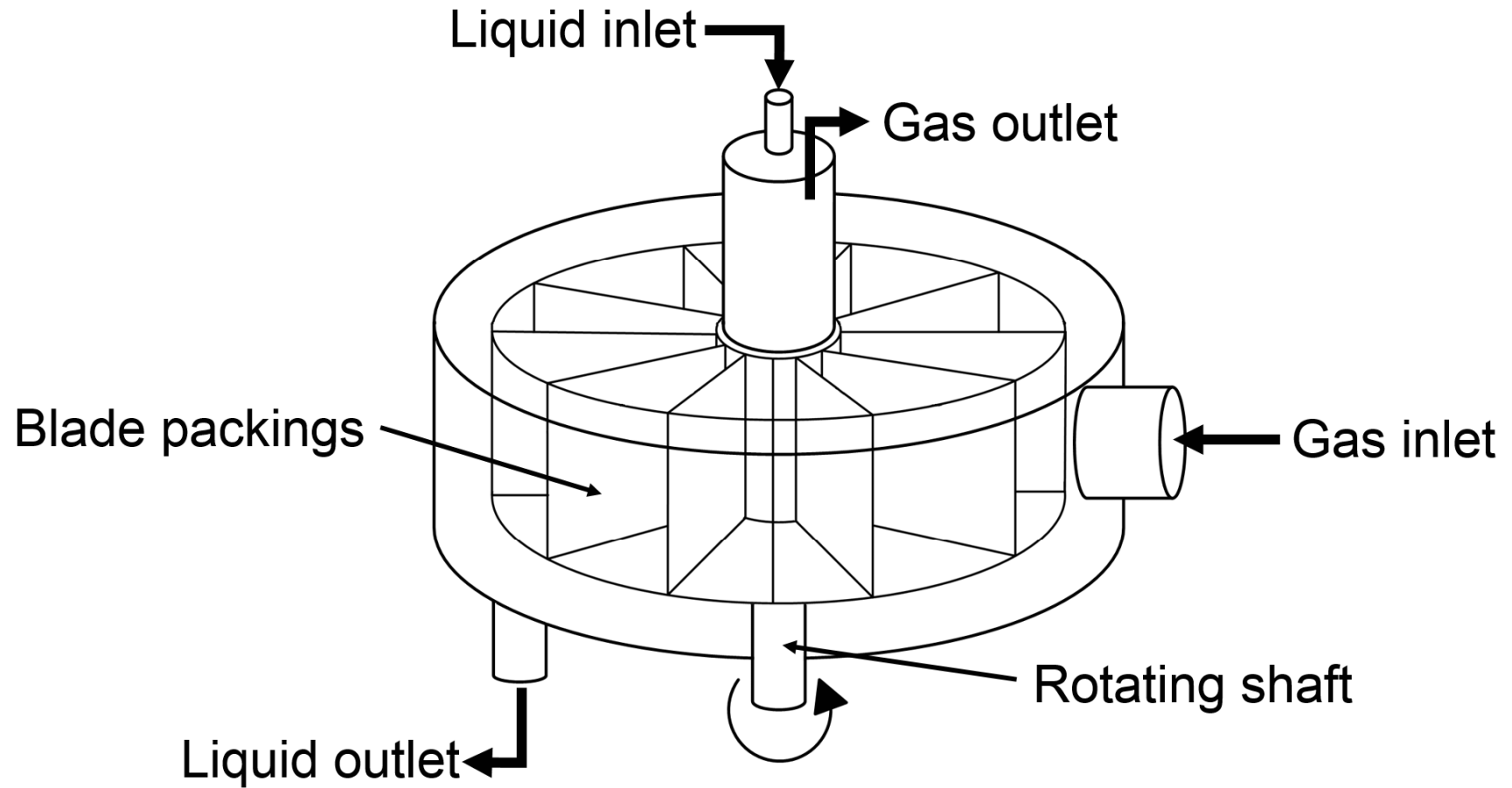


Figure 6

Schematic drawing of RPB with packing and blades (left) Rotor design; (right) main structure of the RPB

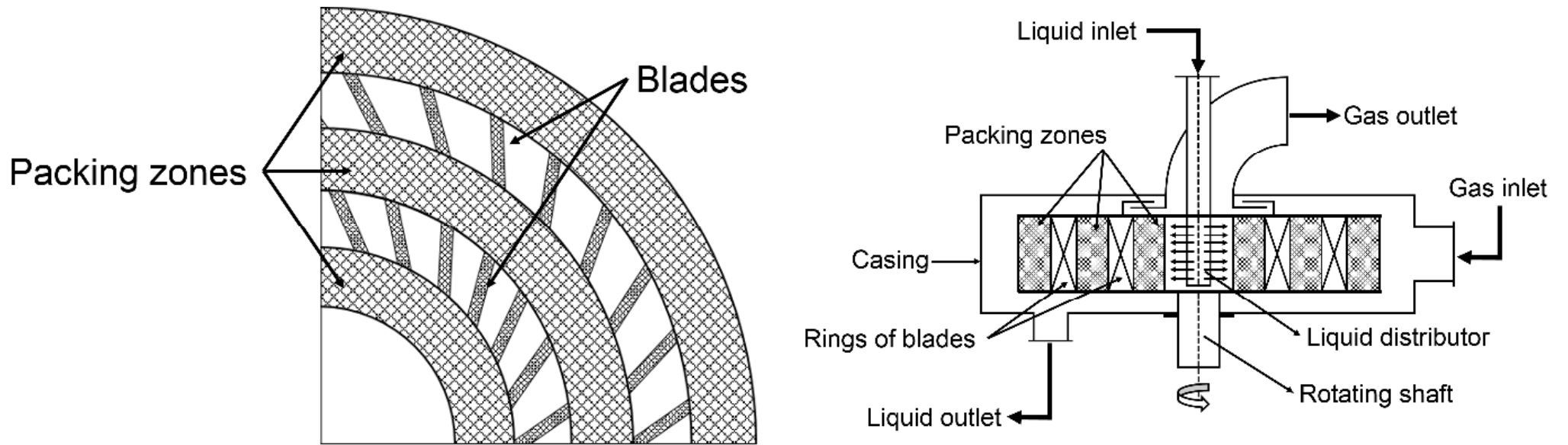


Figure 7

Schematic drawing of blades and baffles rotating bed:

*(left) blade packings on the rotational disk and baffles on the stationary disk
(right) structure of the RPB with blade packings and baffles*

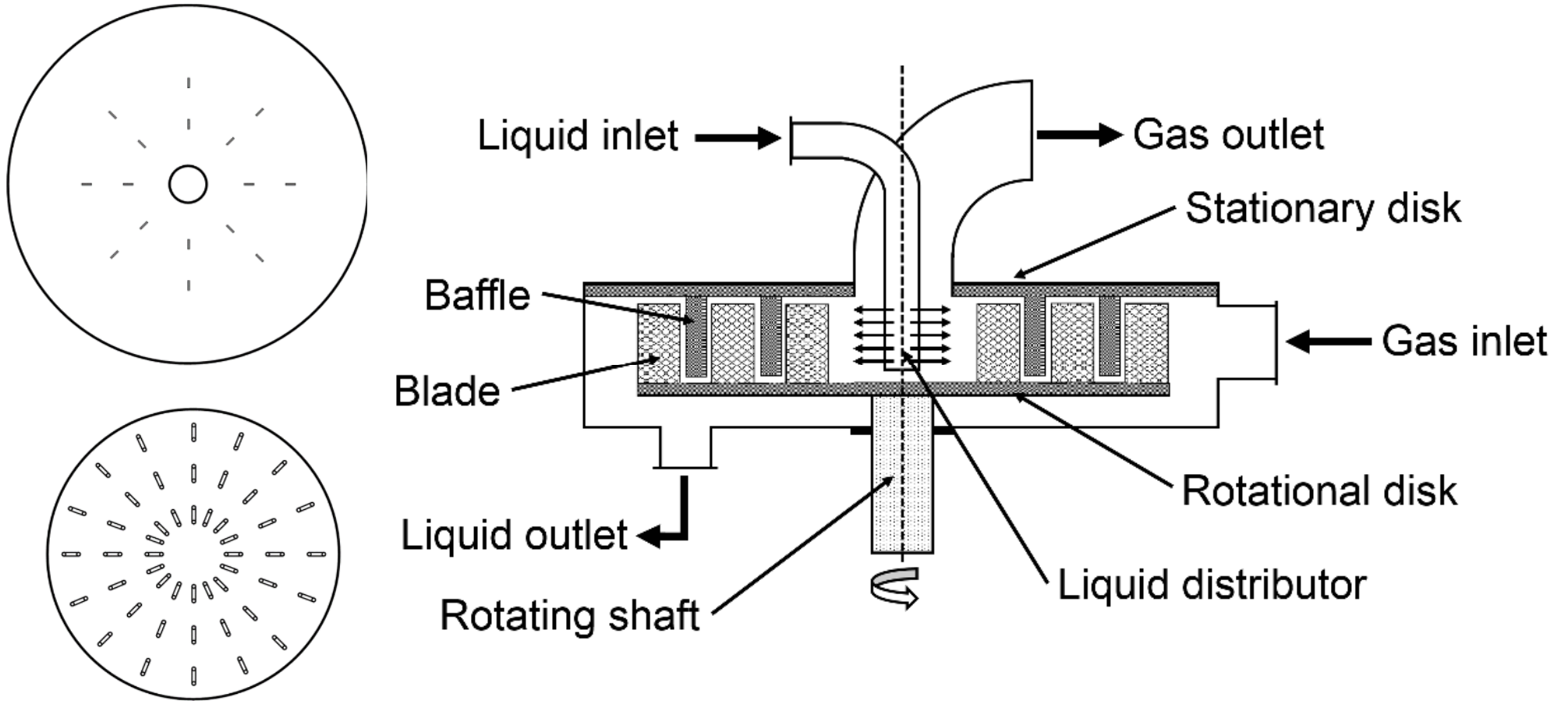


Figure 8

Schematic drawing of a counter-flow concentric-ring rotating bed

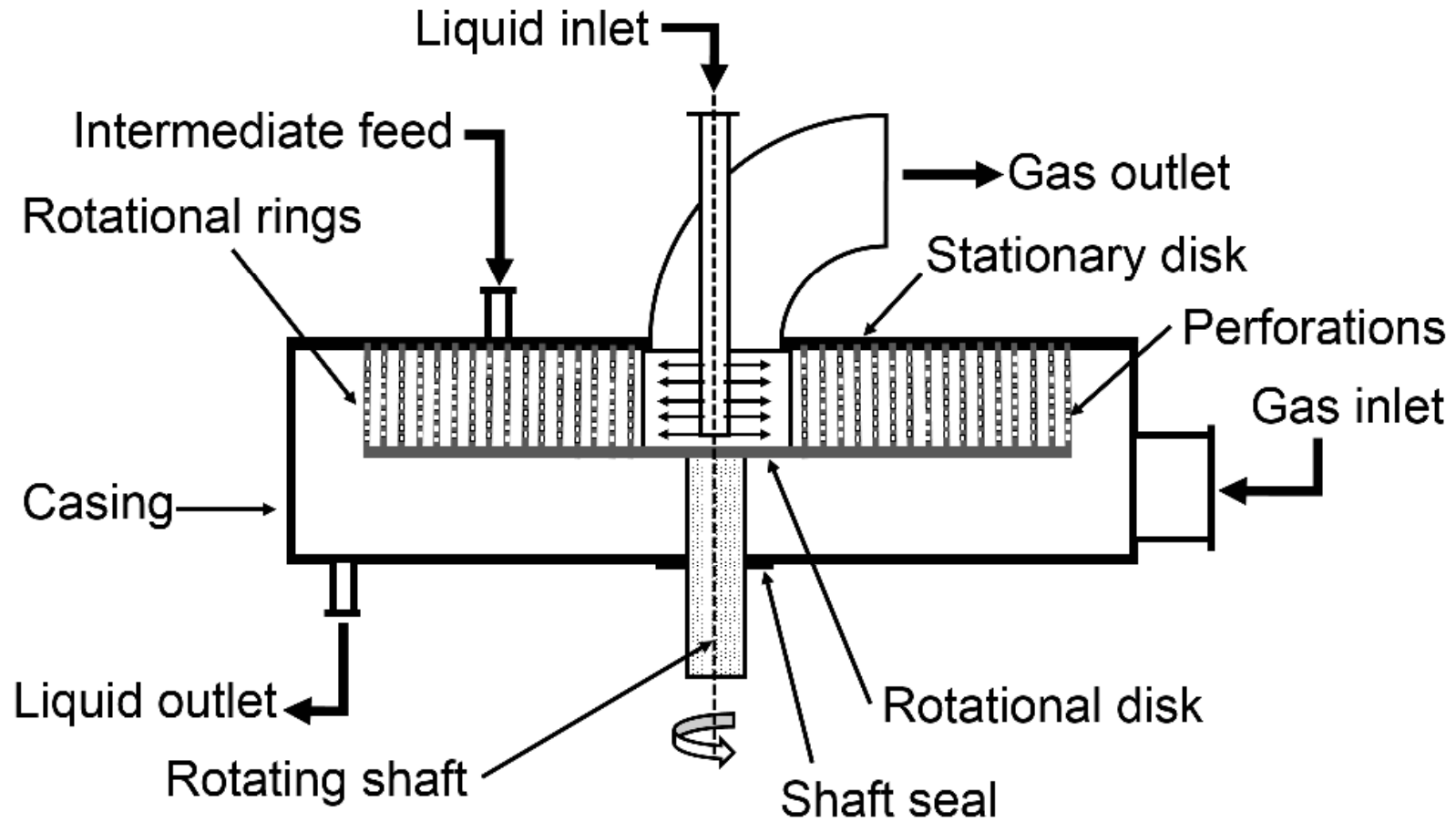


Figure 9

Schematic diagram of a crossflow concentric-baffle rotating bed (CRB)

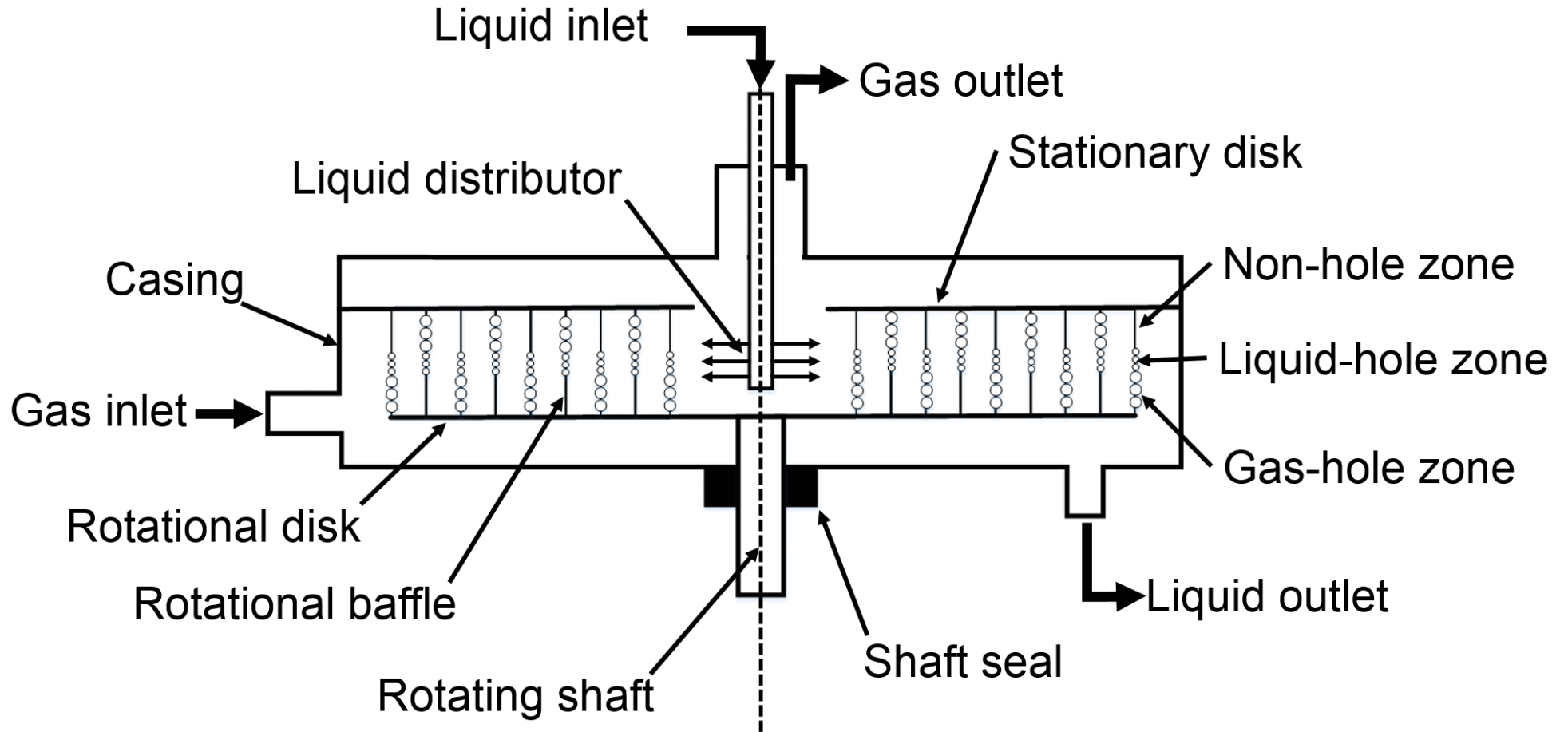


Figure 10

Schematic drawing showing the method followed by Gudena et al. (2012) to approximate HiGee (left sketch) as a conventional column (right sketch). Continuous arrows: vapor flow, dashed arrows: liquid flow

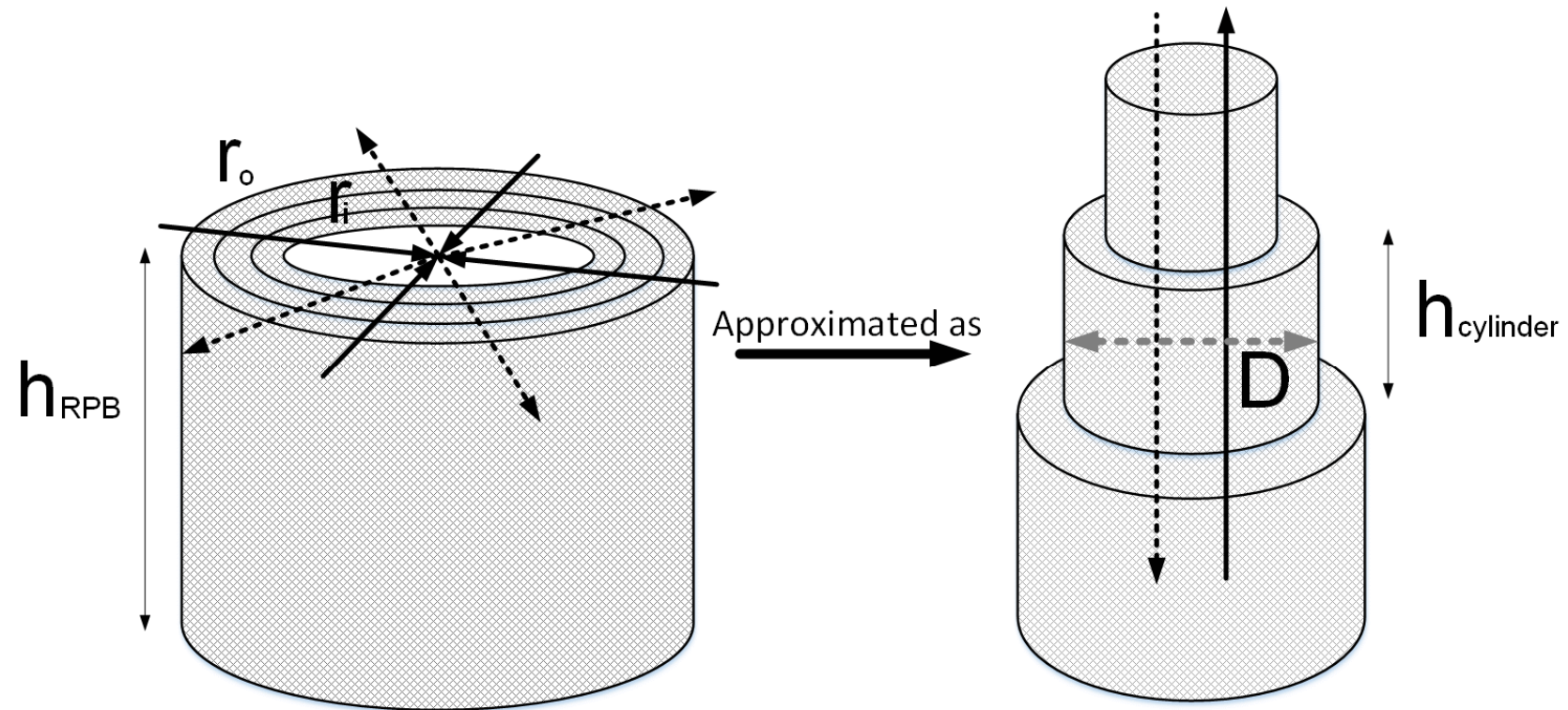


Figure 11

Design analogy between conventional and HiGee distillation

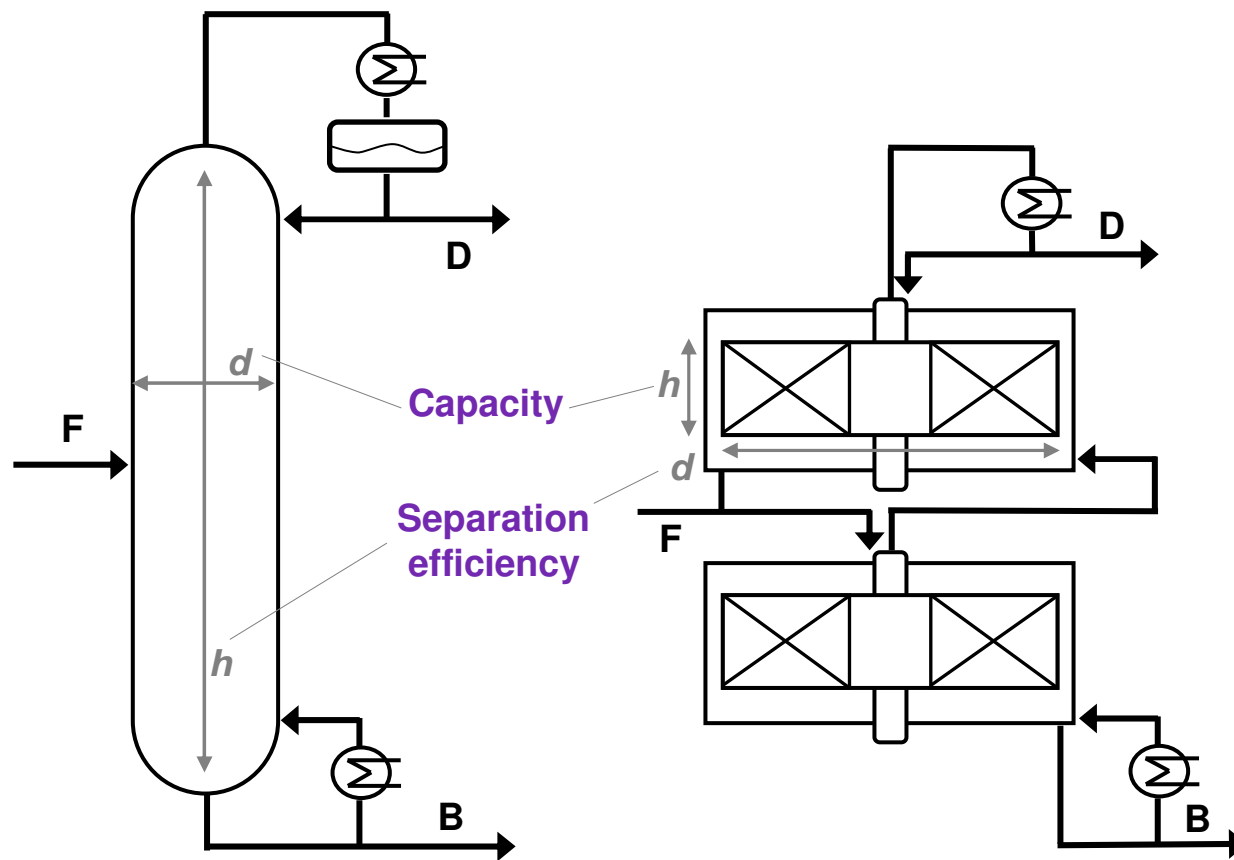


Figure 12