

## Design of mass transfer column

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### I. DEFINITION OF MASS TRANSFER COEFFICIENT

In most of industrial devices mass transfer is due by both convection and diffusion. The specific mass flux  $N = \dot{m}/A$ , [ $kg/m^2s$ ], transferred per unit area of the interface between two phases can be expressed in a form which is very similar to Fick law:

$$\text{Fick law} \quad N = -D \frac{dC}{dz} \quad (1)$$

$$\text{Mass transfer coefficient approach} \quad N = k\Delta C \quad (2)$$

where  $k$ , [ $m/s$ ] is the mass transfer coefficient (or the mass transfer velocity). From a physical view point, within each phase  $k = f(D, \delta, u)$ , where  $D$  is the species diffusion coefficient,  $\delta$  is a characteristic length over which significant concentration gradients may develop and  $u$  is a characteristic velocity which affects  $\delta$  and any convective transport of species. The mass transfer coefficient can be made dimensionless using any reference velocity for the system. Generally, two different dimensionless number can be defined, using either the convection velocity or the diffusion velocity as reference:

$$\text{Stanton Number} = St = \frac{\text{mass transfer velocity}}{\text{fluid velocity}} = \frac{k}{u} \quad (3)$$

$$\text{Sherwood Number} = Sh = \frac{\text{mass transfer velocity}}{\text{diffusion velocity}} = \frac{k\delta}{D} \quad (4)$$

These dimensionless numbers can be predicted as a function of two independent dimensionless parameters:

$$\text{Schmidt Number} = Sc = \frac{\text{diffusivity of momentum}}{\text{diffusivity of mass}} = \frac{\nu}{D} = \frac{\mu/\rho}{D} \quad (5)$$

$$\text{Reynolds Number} = Re = \frac{\text{inertial force}}{\text{viscous force}} = \frac{u\delta}{\nu} = \frac{\text{time for momentum diffusion}}{\text{time for convection}} = \frac{u/\delta}{\nu/\delta^2} \quad (6)$$

Using these dimensionless numbers, mass transfer correlations in the form:

$$Sh = a \cdot Sc^n \cdot Re^p \quad \text{or} \quad St = a \cdot Sc^{n'} \cdot Re^{p'} \quad (7)$$

have been developed experimentally to predict the value of  $k$  in a number of configurations of industrial interest (e.g. mass transfer coefficient for gas/liquid in a packed tower, gas bubbles in a stirred tank, liquid drops rising in stirred/unstirred solution, flow through packed beds). Correlations may differ in their analytical form depending on the specific problem configuration. Examples of correlations can be found in supplementary material (p. 226, Diffusion – Mass transfer in fluid systems, Cussler) and in Perry's Chemical Engineering Handbook.

### II. TWO FILMS THEORY: OVERALL MASS TRANSFER COEFFICIENT

Figure 1 shows the reference configuration of mass transfer across the interface (solid black line) between Phase 1 and 2 when a concentration gradient between the bulk of the two phases exists.

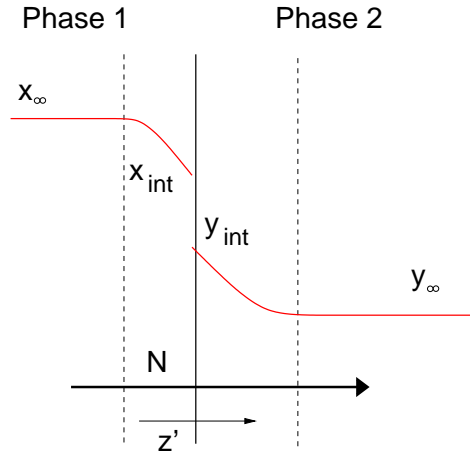


Figure 1. Sketch of two films configuration.

We use  $x$  and  $y$  to indicate the concentration of the species in phase 1 and 2. The footers  $\infty$  and  $int$  identify (i) a point far enough from the interface and (ii) a point at the interface. The qualitative variation of concentration in Phase 1 and 2 is represented by the thick red line. Vertical dashed lines identify the distance from the interface within which a concentration gradient develops in each of the two Phases in contact. Using the mass transfer coefficient defined for each Phase we can write the specific mass flux transferred between the Phases (which at steady state is independent of the position  $z'$  at which it is evaluated) as:

$$N = k_1(x_\infty - x_{int}) \quad (8)$$

or

$$N = k_2(y_{int} - y_\infty) \quad (9)$$

The concentrations at the interface in Phase 1 and 2 are linked by the equilibrium condition

$$y_{int} = m \cdot x_{int} \quad (10)$$

where  $m$  is the partition (or the distribution) coefficient (defined as the ratio of concentration of a species in Phase 1 and 2 when the two Phases are in dynamic equilibrium). For Liquid/Gas systems  $m = f(H)$  where  $H$  is the Henry constant. We can recombine these equations to obtain a different equation for  $N$  in which the driving force for the mass transfer (the difference of concentration) is given as a function of bulk values only (i.e.  $x_\infty$  and  $y_\infty$ ). Using Eq. 10 into Eq. 9 we get:

$$N = k_2(mx_{int} - y_\infty) \quad (11)$$

and using Eq. 8 to obtain  $x_{int} = x_\infty - N/k_1$  and substituting into Eq. 11 we get:

$$N = k_2(mx_\infty - mN/k_1 - y_\infty) \quad (12)$$

and rearranging to isolate  $N$  we get

$$N \left( 1 + \frac{k_2 m}{k_1} \right) = k_2 (mx_\infty - y_\infty) \quad (13)$$

and

$$N = \frac{k_2}{\left( 1 + \frac{mk_2}{k_1} \right)} (mx_\infty - y_\infty) = k_{tot,2} (mx_\infty - y_\infty) \quad (14)$$

$k_{tot,2}$  is the overall mass transfer coefficient representing the velocity of transfer when the driving force considered is the concentration gradient between the two phases expressed using species concentration as defined in Phase 2 ( $y$ ,  $m \cdot x$ ). It can be written as:

$$\frac{1}{k_{tot,2}} = \frac{1}{k_2} + \frac{m}{k_1} \quad (15)$$

Considering that  $1/k_i$  represents a resistance to mass transfer,  $1/k_{tot,2}$  can be considered as the sum of two resistances to mass transfer which are in series (the one across phase 1 and the one across phase 2). Whenever one of these two resistances is very large compared to the other (i.e.  $k_i$  is very small), the mass transfer in Phase i will be **controlling**. This means that most of the concentration gradient for the species transferred will be in Phase i, whereas the variation of concentration in the other Phase will be almost negligible. Whenever this condition is satisfied, the mass transfer problem can be solved focusing on one single Phase (the controlling phase) only.

### III. MASS TRANSFER WITH CHEMICAL REACTION

Chemical reaction occurring inside one phase can be used to enhance the mass transfer. A simple way to account for the effect of chemical reactions is to recall the semi-infinite slab problem which may be solved in closed form either under diffusion only or under diffusion plus reaction. We know that the reaction of the species once absorbed into the second phase (the slab) increases the local concentration gradient and therefore the flux across the interface. Using a mass transfer coefficient to describe the effect of reaction implies a change of the mass transfer rate from  $k$ , the mass transfer velocity without reaction, to  $k_{react}$ , the mass transfer velocity with reaction, which can be written as  $k_{react} = E \cdot k$  where  $E > 1$  is a dimensionless enhancement factor (which depends on the rate of reaction) responsible for the increase of the mass transfer velocity. The chemical reaction reduces the resistance to mass transfer in the medium in which the reaction occurs.

### IV. MASS TRANSFER FLUX USING BULK CONCENTRATION GRADIENTS

Whichever the Phases, in general the specific mass flux can be written as a function of concentration difference between the bulk of the two phases. The phase concentration selected for reference (y or x) determines the definition of overall mass transfer coefficient to use:

$$\text{if concentration in Phase 2, } y, \text{ is selected} \quad N = k_{tot,2}(mx_\infty - y_\infty) \quad (16)$$

$$\text{if concentration in Phase 1, } x, \text{ is selected} \quad N = k_{tot,1} \left( x_\infty - \frac{y_\infty}{m} \right) \quad (17)$$

where  $1/k_{tot,1} = 1/k_1 + 1/(mk_2) = 1/k_{tot,2} \cdot 1/m$ .

### V. DESIGN OF CONTINUOUS CONTACT MASS TRANSFER EQUIPMENTS

Equations 16-17 are useful to design distributed mass transfer systems. We will focus on a gas absorption column in which a gas containing a contaminant flows in contact (co-currently or counter-currently) with a liquid film. The contact between Phases allows to transfer the contaminant from the gas into the liquid medium. We refer to the sketch in Figure 2 to set up our design equations.

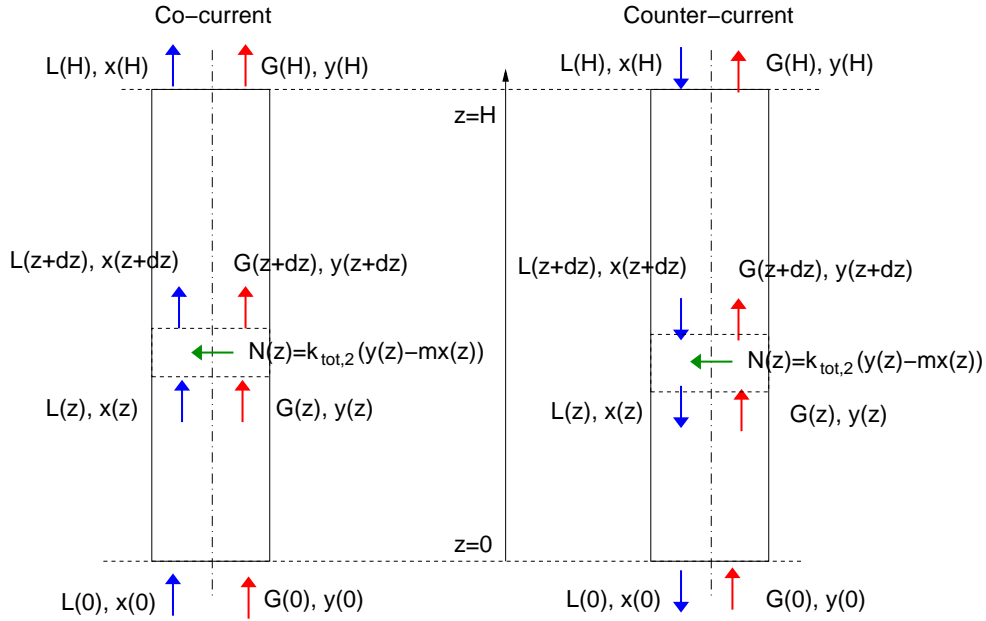


Figura 2. Sketch of absorption column.

We identify with  $L$  and  $G$  the flow rate (in  $m^3/s$  or *mole of fluid/s*) of Phase 1 (the absorbing liquid in our example) and 2 (the gas to be cleaned), and we use  $x$  and  $y$  to identify the concentration in Phase 1 and 2 of the species to be transferred (units will be  $kg/m^3$  or *mole of species/mole of fluid* depending on the units chosen for the flow rate).

In order to size the column, we need to fix the column cross section area,  $A$ , and the column height,  $H$ . The sizing of the section area is based on fluid mechanics considerations which are mainly related to the maximum pressure drop which can be accepted for the system (see supplementary material). Here we focus on the identification of the column height necessary to perform a given mass transfer task.

Typical design variables for the problem are:

- the inlet characteristics of Phase 2, the one to be treated,  $G(0)$  and  $y(0)$ ;
- the target to be achieved,  $y(H)$  (i.e. the outlet concentration allowed for Phase 2);

The characteristics of Phase 1, which is the receiving medium, may be either defined at the inlet section of Phase 2 ( $L(0)$  and  $x(0)$ , co-current configuration) or at the outlet section of Phase 2 ( $L(H)$ ,  $x(H)$ , counter-current configuration). The process variables can be fixed and changed at the design stage until “optimal” operating conditions are found.

To size the column we can write mass balance equations of the form:

$$\frac{dMass}{dt} = \left( \dot{Mass}_{in} - \dot{Mass}_{out} \right)_{convection} + \left( \dot{Mass}_{in} - \dot{Mass}_{out} \right)_{diffusion} \quad (18)$$

This equation can be written for each Phase and with reference to many different control volumes. We assume that the column will operate at steady state conditions: no accumulation of species in the system is therefore possible over time and the term on the left hand side in Equation 18 is zero. To transfer species from Phase 2 to Phase 1 we need to assume that the concentration of species in Phase 2 (the gas in our example) is larger than in Phase 1 (the absorbing liquid). The local value of specific mass flux can be written as:

$$N(z) = k_{tot,2}(y(z) - mx(z)) \quad (19)$$

where the driving force is referred to the bulk concentration of Phase 2.

### A. Co-current configuration

With reference to the variables indicated in Figure 2, for the co-current configuration we can write

1. Mass conservation for Phase 1 in a volume  $dV$  at a generic position  $z$  along the column

$$0 = -(Lx)(z + dz) + (Lx)(z) + N(z)A_{int} \quad (20)$$

where the interfacial area  $A_{int} = adV = aAdz$  is expressed as a function of the specific surface area  $a$ , [ $m^2/m^3$ ], and the reference control volume  $dV$ . The equation can be rearranged to give:

$$0 = -\frac{d(Lx)}{dz}dz + k_{tot,2}(y(z) - mx(z))Adza \quad (21)$$

and if  $L$  is constant with  $z$

$$\frac{dx}{dz} = \frac{k_{tot,2}a}{L/A}(y(z) - mx(z)) = \frac{k_{tot,2}a}{v_{s,L}}(y(z) - mx(z)) \quad (22)$$

where  $v_{s,L}$ , [ $m/s$ ] is the superficial velocity of Phase 1 in the column.

2. Mass conservation for Phase 2 in a volume  $dV$  at a generic position  $z$  along the column

$$0 = -(Gy)(z + dz) + (Gy)(z) - N(z)A_{int} \quad (23)$$

The equation can be rearranged to give:

$$0 = -\frac{d(Gy)}{dz}dz - k_{tot,2}(y(z) - mx(z))Adza \quad (24)$$

and if  $G$  is constant with  $z$

$$\frac{dy}{dz} = -\frac{k_{tot,2}a}{G/A}(y(z) - mx(z)) = -\frac{k_{tot,2}a}{v_{s,G}}(y(z) - mx(z)) \quad (25)$$

where  $v_{s,G}$ , [ $m/s$ ] is the superficial velocity of Phase 2 in the column.

3. Mass conservation for both Phases in a volume  $dV$  at a generic position along the column

$$0 = -(Lx)(z + dz) + (Lx)(z) - (Gy)(z + dz) + (Gy)(z) \quad (26)$$

which gives

$$\frac{d(Lx)}{dz} = -\frac{d(Gy)}{dz} \rightarrow \frac{dy}{dx} = -\frac{L}{G} \quad (27)$$

Equation 27 is the link between concentration in the bulk of Phase 1 and Phase 2, whichever the point  $z$  along the column and is called the operating line.

4. Combined mass transfer equation for Phase 1 and 2

Equations 22-25 can be combined into a single differential equation:

$$\frac{dy}{dz} - m\frac{dx}{dz} = -\frac{k_{tot,2}a}{v_{s,G}}(y(z) - mx(z)) - m\frac{k_{tot,2}a}{v_{s,L}}(y(z) - mx(z)) \quad (28)$$

which can be simplified into

$$\frac{d(y - mx)}{dz} = -k_{tot,2}a \left( \frac{1}{v_{s,G}} + \frac{m}{v_{s,L}} \right) (y(z) - mx(z)) \quad (29)$$

Defining the liquid to gas (Phase 1 to Phase 2) flow ratio as  $R = v_{s,L}/v_{s,G}$ , we get

$$\frac{d(y - mx)}{dz} = -\frac{k_{tot,2a}}{v_{s,G}} \left(1 + \frac{m}{R}\right) (y(z) - mx(z)) \quad (30)$$

which can be integrated by separation of variables:

$$\frac{d(y - mx)}{y(z) - mx(z)} = -\frac{k_{tot,2a}}{v_{s,G}} \left(\frac{R + m}{R}\right) dz \quad (31)$$

to obtain:

$$\ln \frac{(y(z) - mx(z))}{(y(0) - mx(0))} = -\frac{k_{tot,2a}}{v_{s,G}} \left(\frac{m + R}{R}\right) z \quad (32)$$

and finally

$$(y(z) - mx(z)) = (y(0) - mx(0)) \exp \left[ -\frac{k_{tot,2a}}{v_{s,G}} \left(\frac{R + m}{R}\right) z \right] \quad (33)$$

Equation 33 gives the variation of the driving force along the column. The sign of the exponential constant  $(R + m)/R$  is always positive: the difference in concentration between the two phase decreases exponentially along the column.

Equation 32 can be used to calculate how long should be the column to perform a given mass transfer task:

$$H = \left[ -\frac{R}{m + R} \ln \frac{(y(H) - mx(H))}{(y(0) - mx(0))} \right] \cdot \frac{v_{s,G}}{k_{tot,2a}} = NTU \cdot HTU \quad (34)$$

The the term in square brackets is dimensionless and is called NTU, number of transfer units; the second term,  $v_{s,G}/k_{tot,2a}$  has the dimension of a length and is called HTU, height of transfer unit. They measures the difficulty of the mass transfer task (NTU) and the efficiency of the device. The smaller is HTU, the more efficient is the unit; the larger in NTU, the more difficult is to achieve the target.

The number of transfer units is the product of two factors:  $R/(R + m)$  which depends on the value of the flux ratio and on the constant describing the equilibrium between the Phases, and the logarithmic term, which depends on the mass transfer task.

#### 5. Mass conservation for Phase 1 and 2 over the column

To calculate  $H$  we need information on the value of  $x(H)$  which can be obtained quantity writing the mass balance to the entire volume of the column:

$$0 = Lx(0) + Gy(0) - Lx(H) - Gy(H) \rightarrow x(H) = x(0) + \frac{G}{L}(y(0) - y(H)) = x(0) + \frac{(y(0) - y(H))}{R} \quad (35)$$

### B. Counter-current configuration

With reference to the variables indicated in Figure 2, for the counter-current configuration we can write

#### 1. Mass conservation for Phase 1 in a volume $dV$ at a generic position $z$ along the column

$$0 = +(Lx)(z + dz) - (Lx)(z) + N(z)A_{int} \quad (36)$$

The interfacial area  $A_{int} = adV = aAdz$  is expressed as a function of the specific surface area  $a$ , [ $m^2/m^3$ ], and the reference control volume  $dV$ . The equation can be rearranged to give:

$$0 = \frac{d(Lx)}{dz} dz + k_{tot,2}(y(z) - mx(z))Adza \quad (37)$$

and if  $L$  is constant with  $z$

$$\frac{dx}{dz} = -\frac{k_{tot,2a}}{L/A}(y(z) - mx(z)) = -\frac{k_{tot,2a}}{v_{s,L}}(y(z) - mx(z)) \quad (38)$$

where  $v_{s,L}$ , [ $m/s$ ] is the superficial velocity of Phase 1 in the column.

2. Mass conservation for Phase 2 in a volume  $dV$  at a generic position  $z$  along the column

$$0 = -(Gy)(z + dz) + (Gy)(z) - N(z)A_{int} \quad (39)$$

The equation can be rearranged to give:

$$0 = -\frac{d(Gy)}{dz}dz - k_{tot,2}(y(z) - mx(z))Adza \quad (40)$$

and if  $G$  is constant with  $z$

$$\frac{dy}{dz} = -\frac{k_{tot,2}a}{G/A}(y(z) - mx(z)) = -\frac{k_{tot,2}a}{v_{s,G}}(y(z) - mx(z)) \quad (41)$$

where  $v_{s,G}$ , [ $m/s$ ] is the superficial velocity of Phase 2 in the column.

3. Mass conservation for both Phases in a volume  $dV$  at a generic position along the column

$$0 = +(Lx)(z + dz) - (Lx)(z) - (Gy)(z + dz) + (Gy)(z) \quad (42)$$

which gives

$$\frac{d(Lx)}{dz} = \frac{d(Gy)}{dz} \rightarrow \frac{dy}{dx} = \frac{L}{G} \quad (43)$$

Equation 43 is the link between concentration in the bulk of Phase 1 and Phase 2, whichever the point  $z$  along the column and is called the operating line.

4. Combined mass transfer equation for Phase 1 and 2

Equations 38-41 can be combined into a single differential equation:

$$\frac{dy}{dz} - m \frac{dx}{dz} = -\frac{k_{tot,2}a}{v_{s,G}}(y(z) - mx(z)) + m \frac{k_{tot,2}a}{v_{s,L}}(y(z) - mx(z)) \quad (44)$$

which can be simplified into

$$\frac{d(y - mx)}{dz} = -k_{tot,2}a \left( \frac{1}{v_{s,G}} - \frac{m}{v_{s,L}} \right) (y(z) - mx(z)) \quad (45)$$

Using again the liquid to gas (Phase 1 to Phase 2) flow ratio  $R = v_{s,L}/v_{s,G}$ , we get

$$\frac{d(y - mx)}{dz} = -\frac{k_{tot,2}a}{v_{s,G}} \left( 1 - \frac{m}{R} \right) (y(z) - mx(z)) \quad (46)$$

Equations 30 and 46 differ for a single term  $(1 + m/R)$  versus  $(1 - m/R)$ . Both terms tend to 1 if  $R \gg m$ . Equation 46 can be integrated by separation of variables:

$$\frac{d(y - mx)}{y(z) - mx(z)} = -\frac{k_{tot,2}a}{v_{s,G}} \left( \frac{R - m}{R} \right) dz \quad (47)$$

to obtain:

$$\ln \frac{(y(z) - mx(z))}{(y(0) - mx(0))} = -\frac{k_{tot,2}a}{v_{s,G}} \left( \frac{R - m}{R} \right) z \quad (48)$$

$$(y(z) - mx(z)) = (y(0) - mx(0)) \exp \left[ -\frac{k_{tot,2}a}{v_{s,G}} \left( \frac{R - m}{R} \right) z \right] \quad (49)$$

Equation 49 gives the variation of the driving force along the column. Differently from before, the sign of the exponential constant  $(R - m)/R$  determines the variation of the driving force along the column: the concentration difference can either increase or decrease between phases moving along the column depending on the sign of  $(R - m)/R$ :

- if  $R > m$  (for high flow rate ratios), the driving force is decreasing from bottom to top;
- if  $R = m$  (for a precise flow rate ratio which depends on the equilibrium between phases), the driving force is constant all along the column;
- if  $R < m$  (for small flow rate ratios), the driving force is increasing from bottom to top (but very small everywhere);

Equation 48 can be used to calculate how long should be the column to perform a given mass transfer task:

$$H = -\frac{v_{s,G}}{k_{tot,2a}} \cdot \left[ \frac{R}{R-m} \ln \frac{(y(H) - mx(H))}{y(0) - mx(0)} \right] = HTU \cdot NTU \quad (50)$$

Also in this case, to calculate  $H$  we need information on the value of  $x(0)$ . We can obtain this quantity from the mass balance written for the entire volume of the column:

$$0 = -Lx(0) + Gy(0) + Lx(H) - Gy(H) \rightarrow x(0) = x(H) + \frac{G}{L}(y(0) - y(H)) = x(H) + \frac{(y(0) - y(H))}{R} \quad (51)$$

## VI. CO-CURRENT VS COUNTER-CURRENT CONFIGURATION

Equations 35-51 indicate that the change in concentration of Phase 1 between inlet and outlet section is the same whichever the flow configuration (co-current or counter-current). Nevertheless if we fix the inlet concentration for Phase 1, either at the bottom or at the top of the column, the concentration profiles expected along the column in the two alternative configurations will be very different, as sketched in Figure 3.

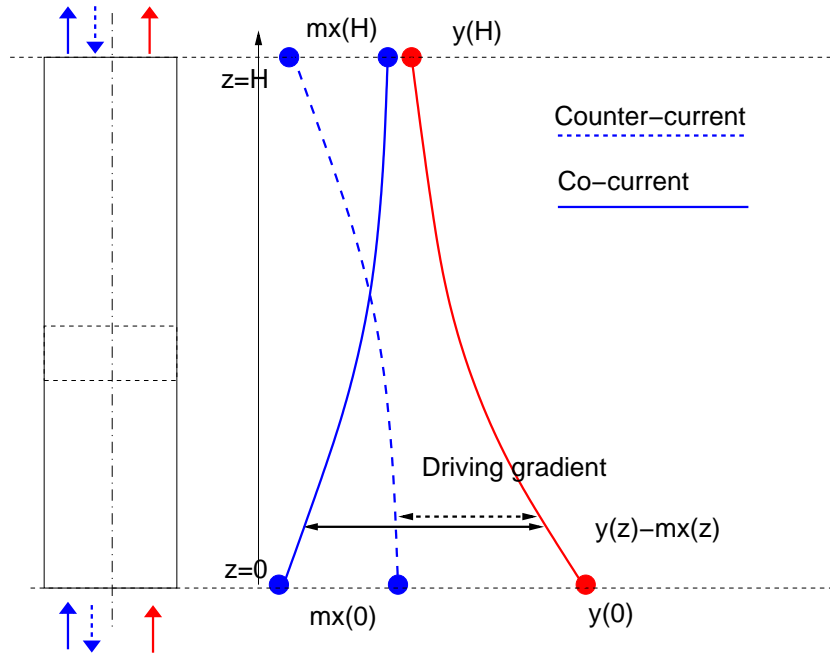


Figura 3. Profiles of driving force along the column in the co-current and counter-current configurations.

Considering that:

$$NTU_{co-current} = -\left[ \frac{R}{R+m} \ln \frac{(y(H) - mx(H))}{y(0) - mx(0)} \right] \simeq \ln \frac{\Delta C_{in}}{\Delta C_{out\ co-current}} \quad \text{if } R/m \gg \quad (52)$$

and

$$NTU_{counter-current} = -\left[ \frac{R}{R-m} \ln \frac{(y(H) - mx(H))}{y(0) - mx(0)} \right] \simeq \ln \frac{\Delta C_{in}}{\Delta C_{out\ counter-current}} \quad \text{if } R/m \gg \quad (53)$$



for a sufficiently large  $R$ , the NTU is found to be larger in the co-current case compared to the counter-current case. This means that a longer column is generally required to complete a target mass transfer task when a co-current configuration is used. Therefore, the counter-current configuration is generally preferred because it leads to more compact design of the mass transfer unit.

## VII. GRAPHICAL INTERPRETATION OF MASS BALANCE EQUATIONS

Figure 4 shows a graphical representations of the design equations found using  $(x, y)$  coordinates, i.e. the concentration of species in Phase 1 and 2. The black line is the Equilibrium line linking the concentration of species at the interface between phases. The slope of the curve is the partition coefficient  $m$ . The green lines represent operating lines, linking the different values of species concentrations in phase 1 and 2 along the column. The slope of the curve depends on the flow ratio  $R = L/G$ . Point A (at  $y = y(0)$ ) indicates the bottom of the column (inlet for Phase 2) whereas point B (at  $y = y(H)$ ) represents the top of the column (outlet for Phase 2). The red dashed line is the 'flux' line linking the concentration of species at the bulk and at the interface. The slope of the curve is the ratio between the mass transfer coefficients in the two phases,  $-k_1/k_2$ .

Consider first the graph on the left, representing a co-current configuration in which the concentration in both Phases is known at the bottom of the column (point A is fixed). The target (reducing  $y$  from  $y(0)$  to  $y(H)$ ) can be achieved selecting different values of  $R$ , which determine a change in species concentration of phase 1 at the outlet section,  $X(H)$  (different blue points). The limiting values of  $R$  which allows to reach the mass transfer target are defined by the two lines indicated in the graph, identified as  $min R$  and  $max R$  ( $max R = \infty$ ). For each position along the column (any point between A and B along the green line) a line parallel to the red dashed line can be used to link the concentration of species in the bulk to the concentration at the interface at the given position  $z$  along the column. The distance between the operating line and the equilibrium line is proportional to  $R - m$ .

Consider now the graph on the right, representing a counter-current configuration in which the concentration in Phase 1 is known at the top (point B is fixed). The target (reducing  $y$  to  $y(H)$ ) can again be achieved selecting different values of  $R$ . Nevertheless, in this case  $max R = \infty$  and  $min R$  depends on the choice of  $x(H)$ .

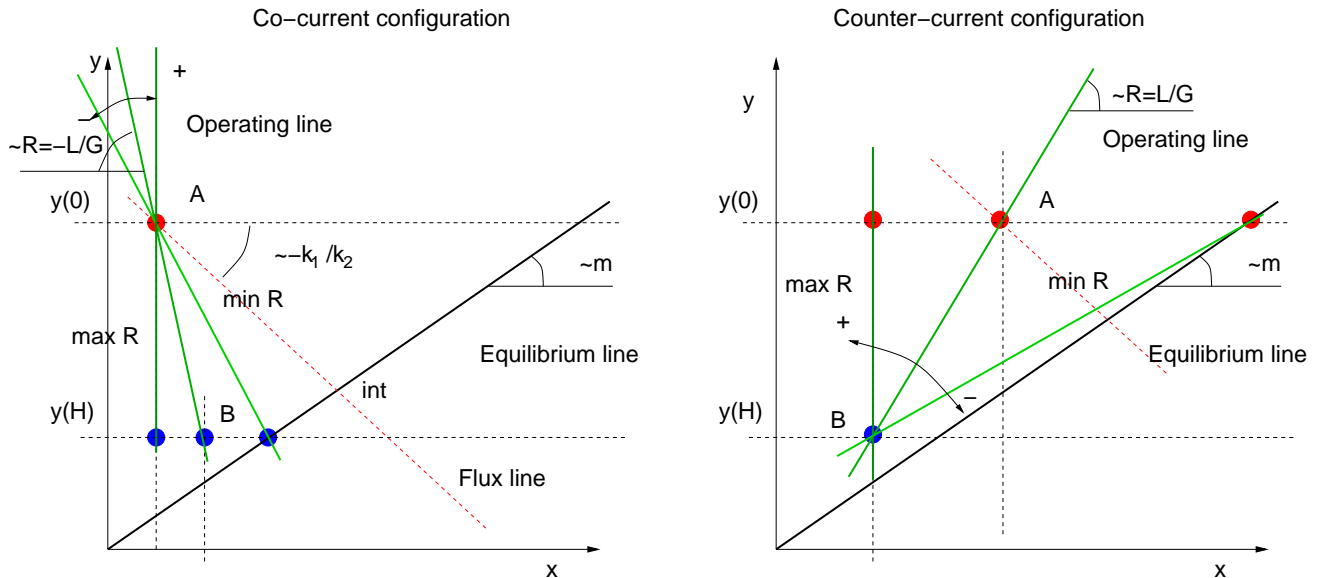


Figure 4. Equilibrium and operating line.