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DISTILLATION CASCADES BASED ON DIFFERENTIAL VAPORIZATION AND DIFFERENTIAL CONDENSATION STAGES. I. TWO-STAGE DCDV CASCADES

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ABSTRACT

Distillation cascades based on differential condensation (DC) and differential vaporization (DV) stages are considered as an alternative to ordinary (equilibrium stage) distillation for the separation of binary mixtures. In the differential condensation–differential vaporization (DCDV) cascades, stage separation factors can be much higher than for cascades based on equilibrium stages, and as a result, significantly fewer stages and, possibly, lower recycle ratios may be required compared with conventional distillation. This paper presents the theory behind the DV and DC stages and explores the use of the two-stage cascade design (a DV reboiler and DC condenser), to recover oxygen for use in hypersonic flight. Part II will consider *N*-stage cascades that could possibly be used to separate most systems now separated by ordinary distillation.

Key Words: Countercurrent differential distillation cascades; Air separation; Hypersonic flight

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INTRODUCTION AND BACKGROUND

This study was motivated by some well-known facts concerning conventional "equilibrium stage" distillation.

- The "equilibrium stage" process used nowadays is basically the same as it was when first developed in the 1800s, and there have been no major changes in distillation that have significantly increased its energy efficiency since then.
- Distillation is very energy intensive.
    - The U.S. Department of Energy, (DOE) estimates that distillation columns operated in the United States use the equivalent of a continuous power consumption of about 91 GW of energy, equivalent to about 3.6 times the total annual energy consumption of Switzerland.
    - The thermodynamic efficiency of conventional distillation is less than 10%.
- Fewer stages and lower "reflux ratios" are required for systems of higher relative volatility. Recycle (or reflux) ratio in the distillation cascade to a large extent determines the energy input required to carry out a specific separation by distillation.
- In conventional distillation the ideal stage separation factor is the same as the relative volatility. Distillation stage efficiency is usually measured in terms of approach to "equilibrium" conditions.
- If a distillation separation cascade can be designed, which results in stage separation factors that are greater than those for equilibrium stage distillation, fewer stages and lower recycle ratios may be required compared with equilibrium stage distillation. This is one motivation for development of extractive distillation processes, for example, and the motivation behind this study. Distillation stage separation factors based on differential vaporization (DV) and differential condensation (DC) can be significantly greater than for equilibrium stage distillation.
- The low efficiency of equilibrium stage distillation is due to the nature of the process. Although the efficiency of the process may be increased slightly by making it more reversible in a thermodynamic sense, and recently there have been innovative tray and packing designs that increase tray efficiency and height equivalent to a transfer unit, (HETU), significant increase in overall efficiency can only happen by developing a radically different process. One possibility is to use the concept of differential condensation–differential vaporization (DCDV) which could circumvent some of the limitations of the equilibrium stage process, provided means can be developed to carry out (or at least approach) the DV and DC processes.

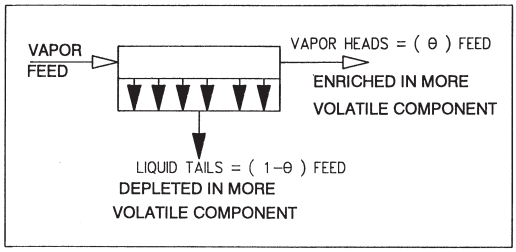
This paper discusses the theoretical aspects of separation cascades based on DV and DC as an alternative to ordinary equilibrium stage distillation starting with the two-stage process based on a DV reboiler and DC condenser. This is strictly a theoretical study since, to our knowledge, process systems hardware have not been developed to date to carry out the differential vapor–liquid equilibrium processes. Some preliminary thoughts on possible means of achieving DC and DV are discussed in a later section.

Many of the developments that follow are based on separation cascade theory as presented by Benedict and others.[1–4]

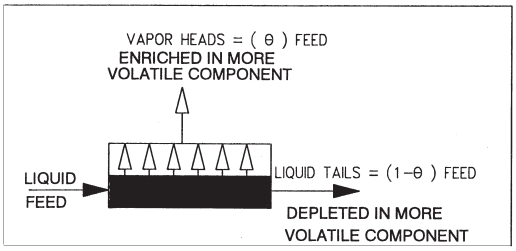
DIFFERENTIAL STAGE SEPARATIONS

In a differential stage separations, successive small portions of a feed stream are removed without mixing from the residual (feed) stream. The residual stream changes progressively in composition. The DC and DV stage separations are shown in Fig. 1.

In developments that follow, the *heads* stream designated by H is the one enriched in the more volatile component, the *tails* stream designated by T is the one depleted in the more volatile component (enriched in the least volatile



(a) DIFFERENTIAL CONDENSATION



(b) DIFFERENTIAL VAPORIZATION

Figure 1. The DC and DV stage separators.

component), while the feed stream is designated by Z, y, x, and z designate the respective mole fractions of the more volatile component, while $(1 - y)$, $(1 - x)$, $(1 - z)$, represent the compositions of the least volatile material. The *cut* or *stage cut* designated by θ is defined as the ratio of heads flow to feed flow. In distillation H is vapor, T is liquid, and Z may be either vapor or liquid.

In the DC stage, the liquid tails stream being removed in small portions is *depleted* in the more volatile component, while the remaining vapor heads stream becomes progressively enriched in this component.

In the DV stage the vapor heads stream being removed in small portions is *enriched* in the more volatile component, while the remaining liquid tails stream becomes progressively depleted in this component.

Partial condensation of a vapor feed stream, carried out in the "right" way, would approximate the DC process. The DV process would be approximated when partial vaporization is carried out in the "right" way. Some conceptual design considerations will be discussed in a later section.

Equations relating flow rates and compositions for the DC process shown schematically in Fig. 2 are derived below.

A material balance on the total flow is:

$$dT^* = -dH^* \tag{1}$$

A material balance on the more volatile component is:

$$x^*\, dT^* = -d(y^* H^*) = -y^*\, dH^* - H^*\, dy^* \tag{2}$$

Elimination of dT^* gives:

$$\frac{dH^*}{H^*} = \frac{dy^*}{x^* - y^*} \tag{3}$$

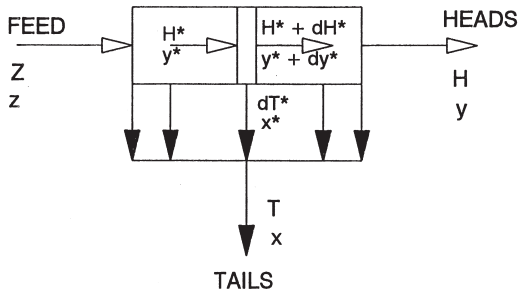


Figure 2. Schematic diagram of a DC separation stage.

It is assumed that, at any point, x^* is related to y^* by the local (or "point") separation factor which is defined as:

$$\alpha^* = \frac{y^*/(1-y^*)}{x^*/(1-x^*)}, \quad x^* = \frac{y^*}{y^* + \alpha^*(1-y^*)} \tag{4}$$

where y^* and x^* are the compositions of vapor and liquid phases in thermodynamic equilibrium and α^* is the (local) relative volatility of conventional distillation.

Substitution of Eq. (4) and integration of Eq. (3) yield

$$\ln\frac{H}{Z} = \int_z^y \frac{dy^*}{x^* - y^*} = \frac{\alpha^*}{\alpha^* - 1}\ln\left(\frac{z}{y}\right) + \frac{1}{\alpha^* - 1}\ln\left(\frac{1-y}{1-z}\right) \tag{5}$$

which assumes $\alpha^* =$ constant.

By definition:

$$\theta \equiv \frac{H}{Z} \tag{6}$$

With this substitution Eq. (5) can easily be transformed to:

$$\left(\frac{1-y}{1-z}\right)\theta = \left[\left(\frac{y}{z}\right)\theta\right]^{\alpha^*} \tag{7}$$

A similar derivation for the DV process yields

$$\frac{dT^*}{T^*} = \frac{dx^*}{y^* - x^*} \tag{8}$$

which, on substitution, integration, and transformation gives:

$$\left(\frac{x}{z}\right)(1-\theta) = \left[\left(\frac{1-x}{1-z}\right)(1-\theta)\right]^{\alpha^*} \tag{9}$$

Note that the DC equation relates the residual vapor (heads stream) composition (y), resulting from a DC of a vapor feed, to the stage feed composition, and cut, i.e., the ratio of residual vapor to feed, while the equation for DV relates the residual liquid (tails stream) composition (x), resulting from a DV of a liquid feed, to the feed composition, and $(1-\theta)$. These are the equations for the so-called Rayleigh distillation and condensation.

These "equilibrium" equations, together with material balance equations, can be used to determine the separation characteristics of either type of process; or a combination of the two processes, i.e., cascades composed of DCDV stages.

Separation in a Single Differential Stage

Cascade theory applies to systems in which stage separation factors are characterized by

$$\alpha_S = \frac{y_S(1 - x_S)}{x_S(1 - y_S)} \tag{10}$$

where x_S and y_S are the compositions of the two (separated) streams leaving the stage.

Figure 3 shows the stage separation factor for the DC and DV processes as a function for θ or $(1 - \theta)$ for different values of the "point" separation factor, or relative volatility which is given by Eq. (4).

As can be seen from Fig. 3, very large stage separation factors can result from DC as more and more condensation takes place $[(1 - \theta) \rightarrow 1]$ because the residual vapor composition $(1 - y_S) \rightarrow 0$. However, under these conditions, $x_S \rightarrow z_F$, and, although α_S is large, the recovery of the more volatile component in the residual vapor is small.

It is also seen that high values of α_S are obtained from DV as $\theta \rightarrow 1$ due to $x_S \rightarrow 0$, although, under these conditions, $y_S \rightarrow z_F$. However, under these

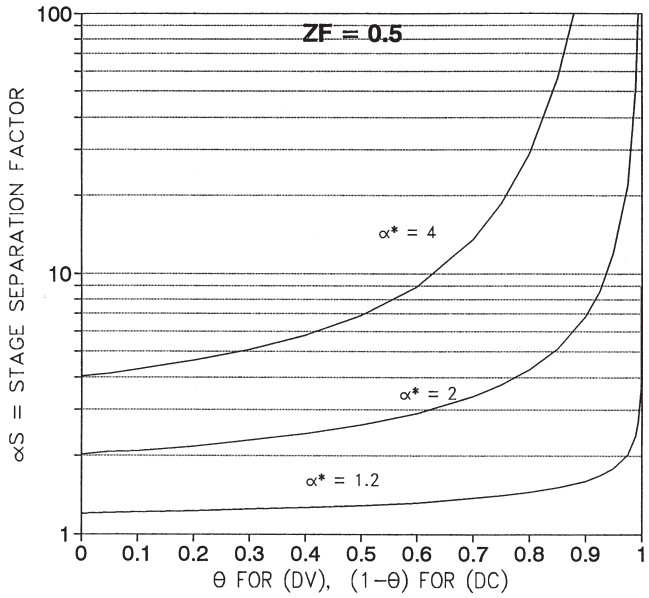


Figure 3. Stage separation factor as a function of θ or $(1 - \theta)$ for the DC and DV processes for different values of α^*. Feed composition $z_F = 0.5$.

conditions, the recovery of the less volatile material in the tails stream is small, although the concentration may be high.

In both cases higher relative volatility (α^*) results in better separations at lower values of $(1 - \theta)$ or θ.

Thus, although high enrichment of the more volatile material can be obtained by DC, and high enrichment of the less volatile material can be obtained by DV, these alone, in general, would not be practical separation devices, because of the poor recovery, although DC followed by DV may be useful for certain applications provided α^* is high enough (vide infra). However, a recycle cascade containing a combination of the two processes can theoretically result in an overall separation with high enrichment and recovery. The two-stage DCDV process will be considered in this paper, while *N*-stage cascades will be considered in Part II.

PRELIMINARY CASCADE MODELS FOR DCDV DISTILLATION

Just as with ordinary distillation, to carry out a specified separation, many cascade designs are possible based on the DCDV processes, with the cascades ranging from "constant recycle" (constant molal overflow), to "no-mix" cascades in which mixing of streams of different compositions does not occur in the stage, to an "ideal" cascade which requires the minimum interstage flow to make a desired separation. Some of the no-mix and ideal cascade requirements have been discussed by Herbst and McCandless.[4]

To date, experience shows that calculation procedures for the DCDV cascades are considerably more complicated than for ordinary distillation column design, and so the "easier" cascades are considered first. First, the two-stage process is considered which contains a DV stage as the "reboiler" and a DC stage as a (partial) condenser. Since, depending on α^*, very high recycle ratios are required to result in the high α_S required for to make separations with only two stages, models for *N*-stage cascades will be developed in Part II.

Two-Stage DCDV Process

Some separations can be carried out theoretically in the two-stage DCDV process with reasonable recycle ratios provided α^* is large enough. The two-stage cascade with liquid feed is shown schematically in Fig. 4.

In this process, the liquid feed is mixed with the tails stream from the DC stage to form the feed to the DV (reboiler) stage, while the combined vapor from the DV stage is the feed to the DC (partial condenser) stage. At least two 2-stage

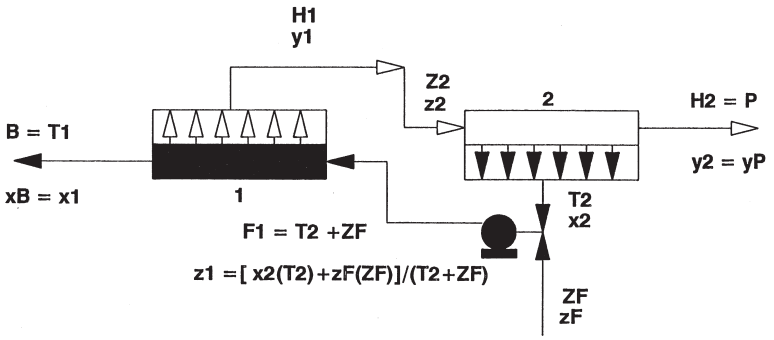


Figure 4. The two-stage DCDV process with partial DV reboiler and partial DC condenser, liquid feed, and liquid recycle.

arrangements are possible: one in which the recycle ratio, (RR) is specified, and one in which the combined tails stream from the DC stage has exactly the same composition as the feed.

In the no-mix cascade design the DC condenser produces T_2 with composition $x_2 = z_F$, while the DV reboiler produces T_1 at specified x_B ($x_1 = x_B$). This "no-mix" design requires a specific RR. The other stream flows and compositions are fixed by the "equilibrium" and material balance equations that are solved simultaneously to yield B, P, y_P, etc.

Here the recycle ratio is defined as

$$\mathrm{RR} = T_2/P$$

to be consistent with ordinary distillation.

As will be shown, high enrichment and recovery can be obtained provided α^* is large enough, which, even for the no-mix case, can require a large recycle ratio. Of course, for high α^*, separations are easily carried out for conventional distillation, but the two-stage DCDV process may be useful for certain applications since overall process "size" may be smaller than for conventional distillation (vide infra).

Calculations were made for the no-mix recycle DCDV process to produce $(1 - x_B)$ between 0.90 and 0.99, for $\alpha^* = 1.5$, 2, 3, 4, and 5.75 assuming $z_F = 0.5$. The results of these calculations are presented in Fig. 5 which presents the required recycle ratio, the resulting α_S and y_P for the two-stage no-mix DCDV process.

As can be seen from Fig. 5, the two-stage no-mix design can produce good separations with reasonable RR provided α^* is high enough. The stage separation factors that result from these two-stage no-mix designs are equal and vary from about 10 to 100, depending on the specific design and α^*. These calculations

suggest that the two-stage DCDV cascade may be feasible for systems with $\alpha^* >\sim 3$.

The two-stage no-mix design is a special case of fixed RR design, requiring a specific RR to make the specified separation. Calculations were made for the two-stage DCDV cascades, to produce $(1 - x_B) = 0.90$ and 0.99, again with feed composition $z_F = 0.5$, using different RRs for systems with $\alpha^* = 1.5$, 2, 3, 4, and 5.75. The results of these calculations are presented in Fig. 6, which presents the y_P and resulting stage separation factors (α_{S1} and α_{S2}) as function of RR.

As can be seen from Fig. 6, high RRs are required for good separation with $\alpha^* < 2$, but with $\alpha^* > 2$, RR < 20 produces good separations in the two-stage DCDV cascade. For this design the stage separation factors, in general, are different and increase with cascades designed for larger RR. At a specific RR the two-stage separation factors are equal and at that RR, they are no-mix cascades. At RR less than the no-mix value $\alpha_{S1} > \alpha_{S2}$, but $\alpha_{S1} < \alpha_{S2}$ for RR greater than the no-mix case.

Although many separations could be carried out theoretically using only two stages, higher recycle ratios and hence greater energy input may be required compared with optimized conventional distillation, but as mentioned earlier, process size may be smaller. In addition, in contrast to conventional distillation in which each ideal stage is at constant composition and temperature (with decreasing stage temperatures from reboiler to condenser), on each differential stage there is a concentration gradient with concomitant temperature gradient. An example of temperature gradients necessary for the separation of O_2 and N_2 for the DC and DV processes without recycle is presented in Fig. 9b below.

A possible application for the two-stage DCDV process is discussed in the following section.

A POSSIBLE APPLICATION FOR THE TWO-STAGE DCDV PROCESS: THE RECOVERY OF OXYGEN FROM AIR FOR USE IN HYPERSONIC FLIGHT

Various separation technologies were evaluated for air collection and enrichment systems (ACES) to be used on air-breathing space boosters.[5] The idea is to collect oxygen en route for rocket use beyond the air-breathing envelope.[6]

The goal for an ACES is to provide for an oxygen-enriched liquid stream of at least 90% O_2, with a recovery of at least 50% of the O_2 in the inlet feed air. Process equipment weight and volume restrictions require a specific weight of less than 10 kg-sec/kg and a specific volume of less than 0.5 ft^3-sec/lb. Liquid H_2, which is available on the flight vehicle at 25°R and 20 psia, may be used for process refrigeration. Only the separation requirements are addressed in this study.

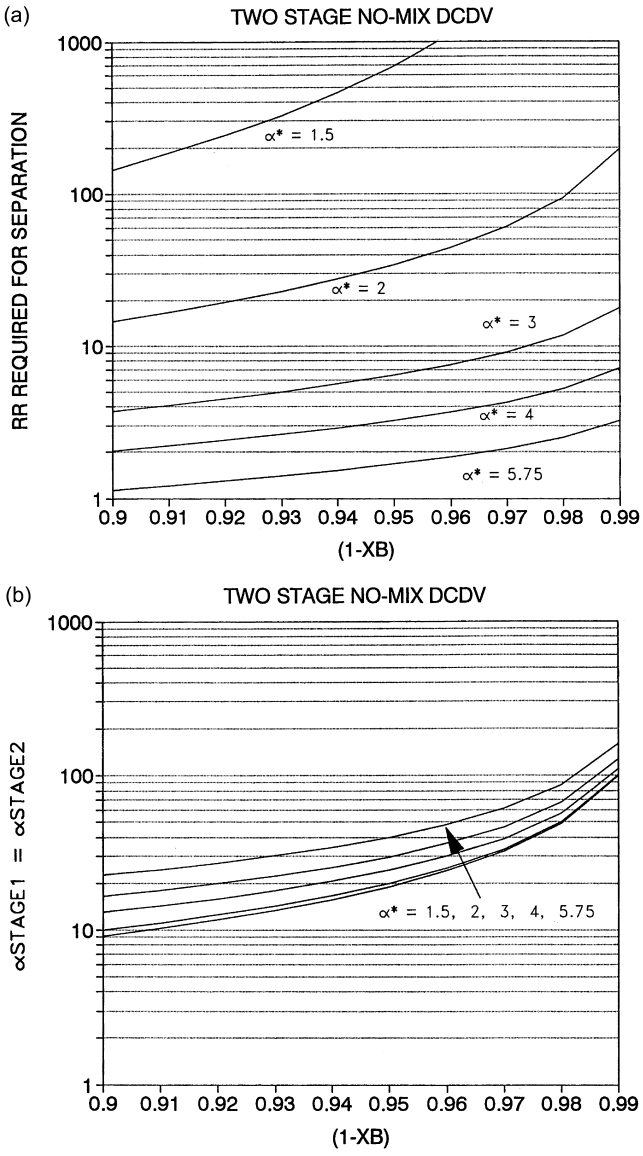


Figure 5. (a) Required recycle ratio for the indicated separation for a two-stage no-mix DCDV process for various α^*; (b) the resulting α_S for the no-mix design; (c) the resulting y_P.

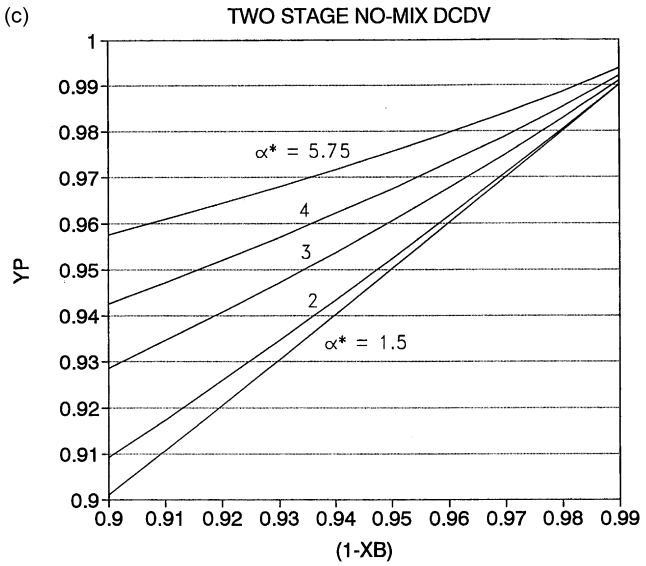


Figure 5. Continued.

Calculations using Eqs. (7) and (9) and appropriate material balance equations were made to determine the separation that would be obtained when the differential stage condensation and differential stage vaporizations are carried out for air, assuming constant relative volatility, $\alpha^* = 4$ and 5.75, assuming air to be composed only of O_2 and N_2 with composition $y_{N_2} = 0.79$. Nitrogen is the more volatile species, and, although relative volatility varies somewhat with composition, $\alpha^* = 4$ and 5.75 are the average values calculated by Raoult's law for about 1 and 0.2 atm pressure, respectively. CHEMCAD® III (Chemstations, Inc., Houston, TX, USA) calculations predict average $\alpha^* = 4.07$ and 6.09 at these pressures.[5] Raoult's law values were used for this preliminary study. Theoretical calculations indicate that a combination of DC and DV could achieve the desired enrichment/recovery of O_2 for the ACES for hypersonic flight applications. With $\alpha^* = 5.75$, the DCDV process without recycle can produce the desired minimum separation, while the two-stage recycle process with vapor feed can result in very high enrichment and recovery with both $\alpha^* = 4$ and 5.75.

Figure 7 shows the DCDV process without recycle. In calculations that follow the recovery of O_2 is defined as:

$$R^{O_2} = \frac{T_2(1 - x_B)}{Z(1 - z_F)} \tag{11}$$

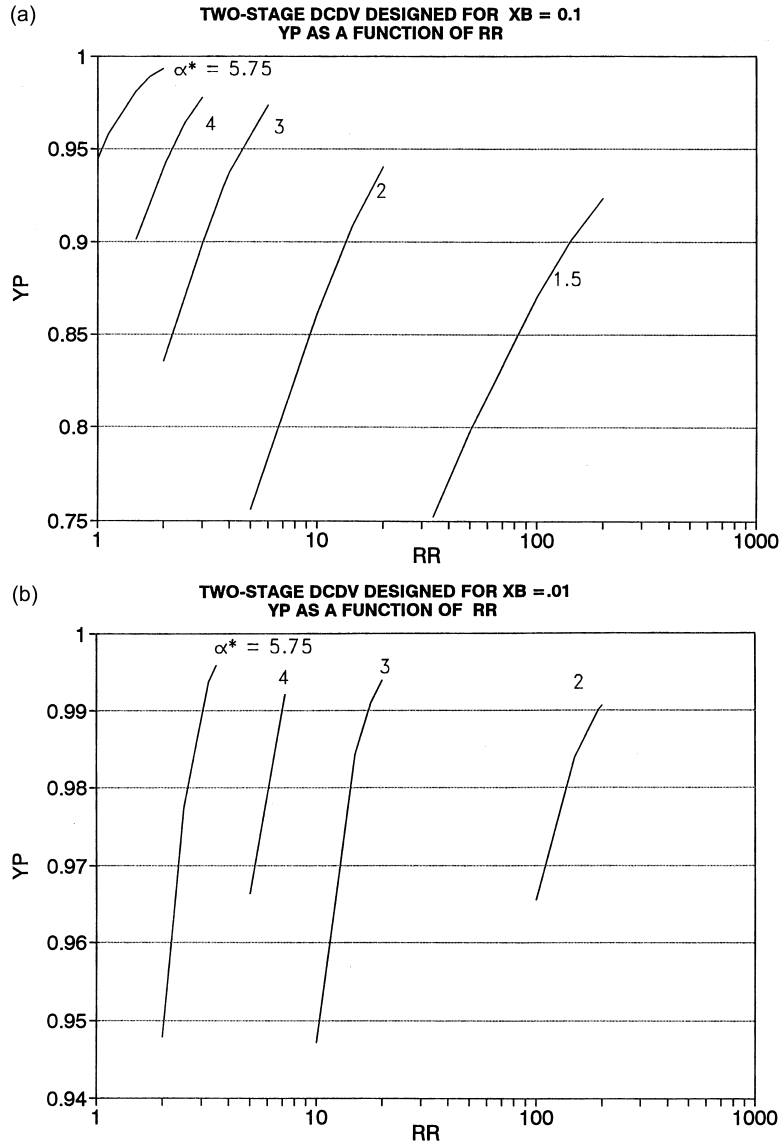


Figure 6. Separation characteristics of two-stage fixed recycle DCDV cascades designed to produce $(1 - x_B) = 0.90$ and 0.99 for $\alpha^* = 1.5$, 2, 3, 4, and 5.75. y_P as a function of RR, and α_{S1} and α_{S2} as a function of RR.

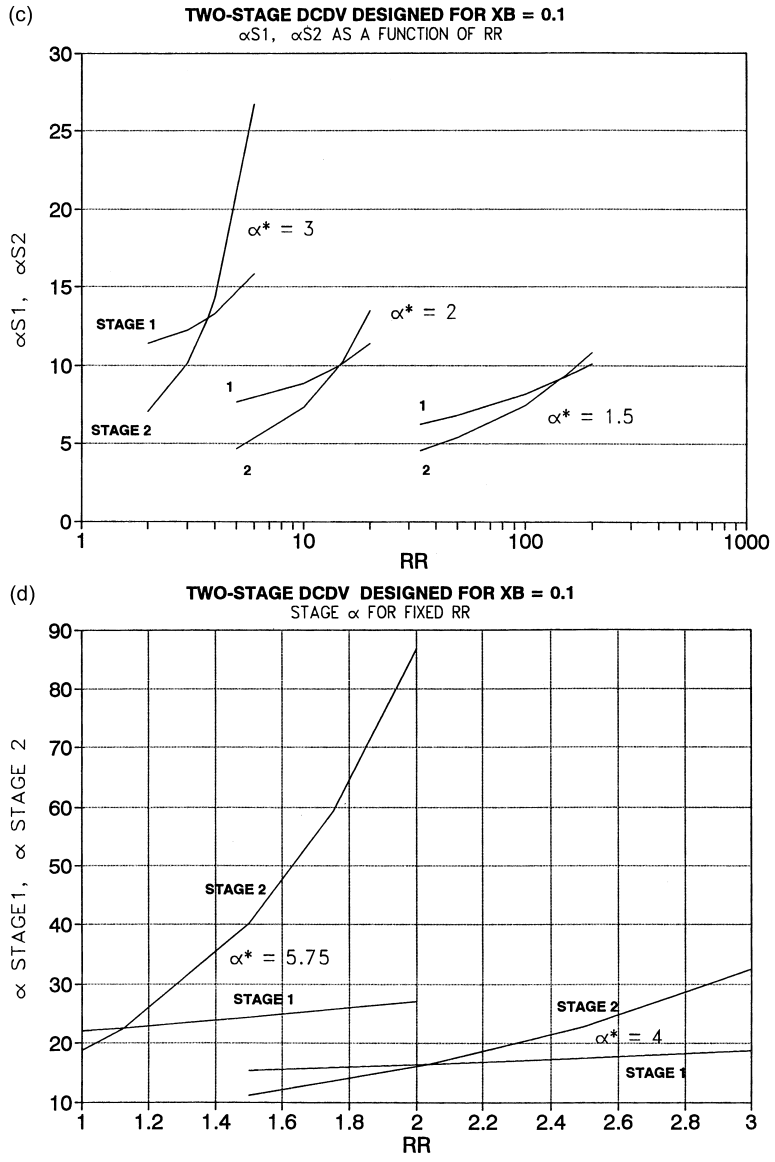


Figure 6. Continued.

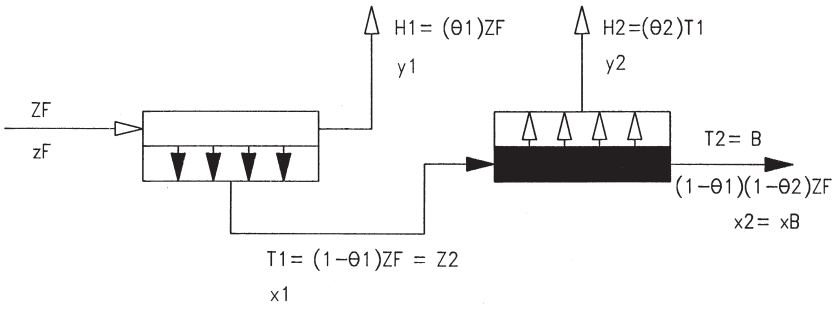


Figure 7. The DCDV process without recycle.

The possibility of producing a product with $(1 - x_B) = 0.9$ with $R^{O_2} > 0.5$ by the scheme in Fig. 7 was investigated. Calculations were made for $(1 - \theta_1)$ between 0.1 and 0.9, followed by θ_2 to produce $(1 - x_B) = 0.9$. Figure 8 shows the recovery and θ_2, resulting from these calculations. As can be seen, for $\alpha^* = 4$, $x_{O_2} = 0.9$, $R^{O_2} \geq 0.5$, cannot be attained with the simple DCDV process, but with $\alpha^* = 5.75$, the desired separation can be attained over a fairly wide range of $1 - \theta_1$ between about 0.4 and 0.85. The maximum recovery, $R^{O_2} = 0.5395$, is at

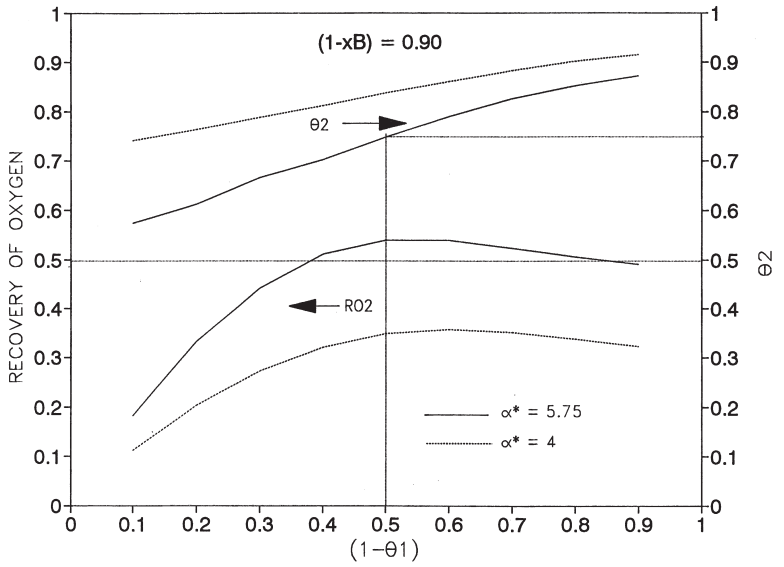


Figure 8. Recovery and θ_2 as a function of $(1 - \theta_1)$ for the no-recycle DCDV process to produce $(1 - x_B) = 0.90$ from $z_F = 0.79$.

about $(1 - \theta_1) = 0.5$, $\theta_2 = 0.7483$. This separation process is shown in Fig. 9a, which presents the liquid composition as a function of the recovery as the vapor feed is differentially condensed and the combined condensate is differentially vaporized under these process conditions. Figure 9b shows the temperature gradients (dew points and bubble points) for the DC and DV processes, calculated by Raoult's law.

This analysis shows that the desired separation could be obtained with the simple DCDV process, provided α^* is large enough, which only happens at lower pressures for the O_2–N_2 system. In addition, as shown below, the recovery-enrichment can be increased if a recycle DCDV process is used.

DCDV Recycle Process for ACES

A DCDV separation process with vapor feed and liquid recycle is shown schematically in Fig. 10.

In this process, the vapor feed is mixed with the heads stream from the DV stage to form the feed to the DC stage, while the combined condensate from the DC stage is the feed to the DV stage. The heads from the DC process is the product enriched in N_2, while the tails stream from the DV process is enriched in O_2. As will be shown, high enrichment and recovery can be obtained provided the process is designed for a large enough recycle ratio.

Calculations were made for the recycle DCDV process to produce $(1 - x_B) = 0.90$, 0.95, and 0.99 utilizing various RRs for $\alpha^* = 4$ and 5.75, again assuming $z_F = 0.79$. The results of these calculations are shown in Fig. 11.

As can be seen, high enrichment and recovery of oxygen is possible when the DCDV process is designed for recycle. The process designed for RR ≈ 0.337 would be required to produce the minimum desired separation with $\alpha^* = 5.75$, while a design with RR ≈ 0.558 would be required if $\alpha^* = 4$. Higher RR would result in higher enrichment and/or recovery. Enrichment up to 99% oxygen at 50% recovery could be obtained with RR about 0.98 and 0.50 with $\alpha^* = 4$ and 5.75, respectively.

For a design with RR $\rightarrow 0$, in the limit, there would be no separation, since there would be no vaporization or condensation occurring under these conditions.

It is interesting to compare the amount of condensation and vaporization required to produce the minimum desired separation for the simple and recycle DCDV processes, $\alpha^* = 5.75$. For this case, the recovery goes through a maximum (~ 0.54 at about $\theta_1 = 0.5$) as θ_1 is increased ($x_{O_2} = 0.9$), and, as a result, $R^{O_2} = 0.5$ at about $(1 - \theta_1) = 0.38$ and 0.835, as shown in Fig. 3. The required condensation and vaporization rates (based on a feed rate of 100) to accomplish this separation with the simple and recycle DCDV processes are shown in Table 1.

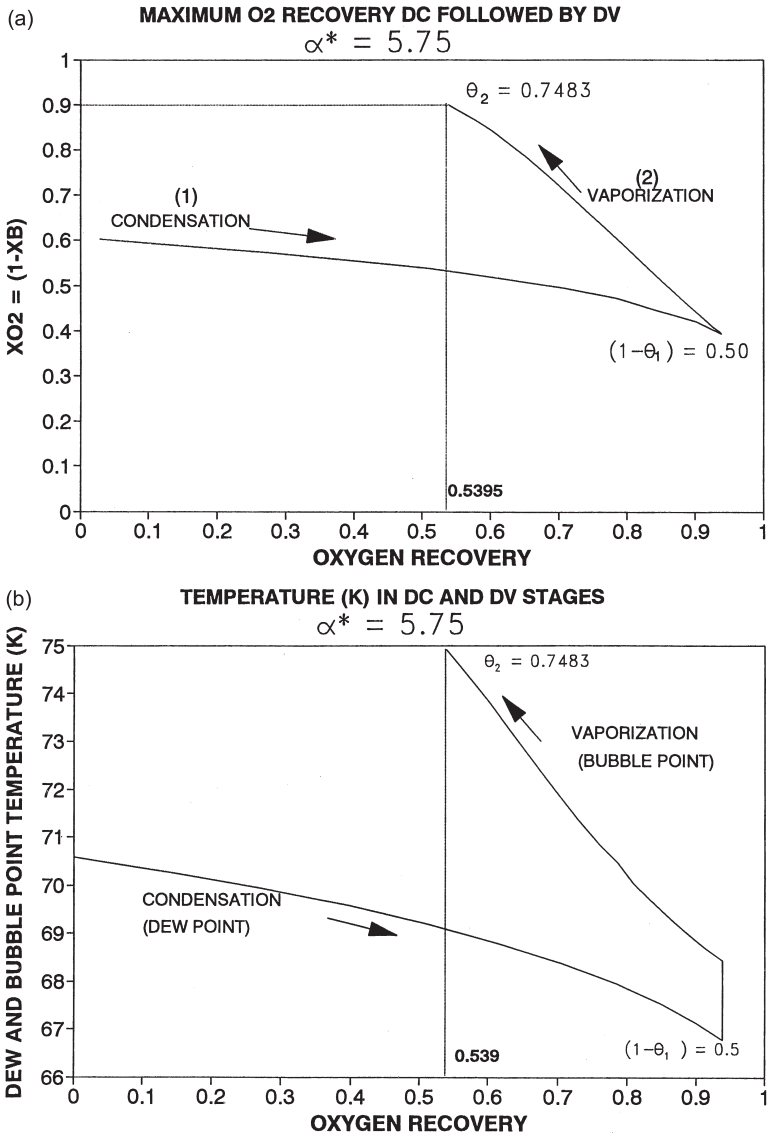


Figure 9. (a) Liquid phase composition as a function of % recovery and (b) temperature profile during condensation (dew point) and vaporization (bubble point) for the no-recycle DCDV process when vapor feed is: (1) condensed to $(1 - \theta_1) = 0.50$ and (2) vaporized to $\theta_2 = 0.748$.

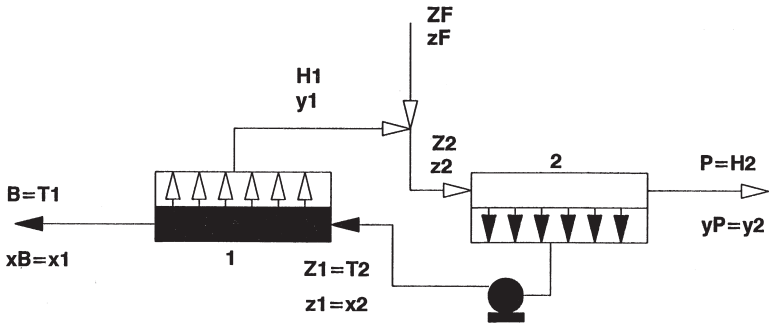


Figure 10. DCDV separation with vapor feed and liquid recycle.

Significantly, the recycle DCDV design requires less total condensation and vaporization than the simple (no recycle) DCDV process, to accomplish the desired minimum separation.

Comparison of Recycle DCDV with Ordinary Distillation

In ordinary distillation, the individual stage separation factors are limited to α^* as a maximum, but the stage separation factors in the DCDV process can be much higher than α^*. This is shown in Fig. 12 which shows the stage separation factor, α_i as a function of recycle ratio for the recycle DCDV process to produce $(1 - x_B) = 0.9$, when $\alpha^* = 4$ and 5.75.

As can be seen, for all RR > 0, both of the stage separation factors are greater than α^* and increases with increasing RR. As a result, the separation requires fewer stages than ordinary distillation to make the same separation.

Table 1. Comparison of the Simple and Recycle DCDV Processes to Produce the Minimum Desired Separation $(1 - x_B) = 0.9$, $R^{O_2} = 0.5$, $\alpha^* = 5.75$. Stream Rates: Relative to Feed = 100

	Simple		
Process	$(1 - \theta_1) = 0.38$	$(1 - \theta_1) = 0.835$	Recycle
Condensation rate	38	83.5	29.8
Vaporization rate	26.3	71.8	18.1

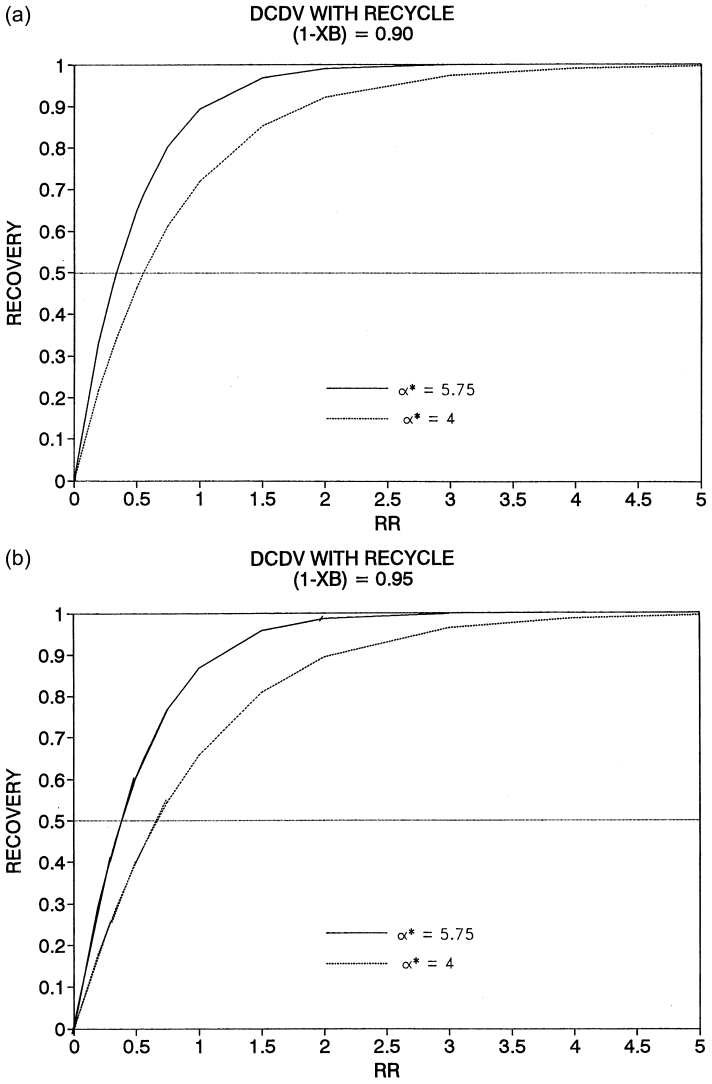


Figure 11. Recovery as a function of recycle ratio for the recycle DCDV process to produce: (a) $(1 - x_B) = 0.90$, (b) $(1 - x_B) = 0.95$, and (c) $(1 - x_B) = 0.99$ for $\alpha^* = 4$ and 5.75. (d) RR required to obtain 50% recovery at these product concentrations, for $\alpha^* = 4$ and 5.75.

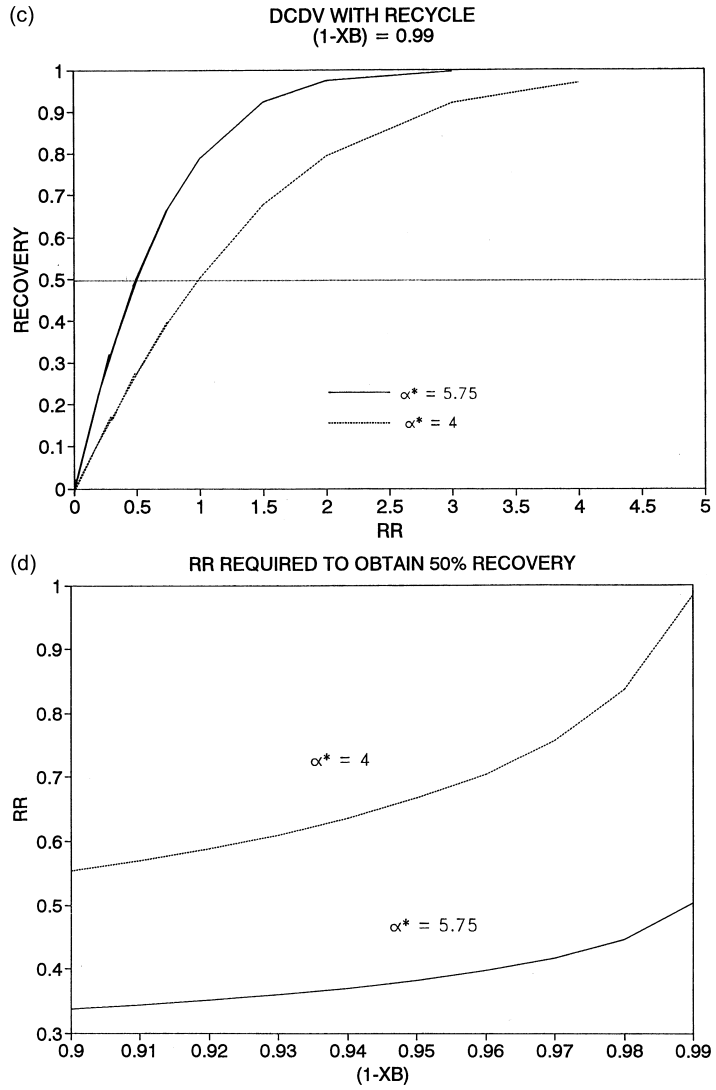


Figure 11. Continued.

A direct comparison of the DCDV process with ordinary distillation is complicated because many distillation designs are possible which will yield the desired separation, each requiring a different number of stages and RR (and pressure). For ordinary distillation the minimum number of stages (infinite RR)

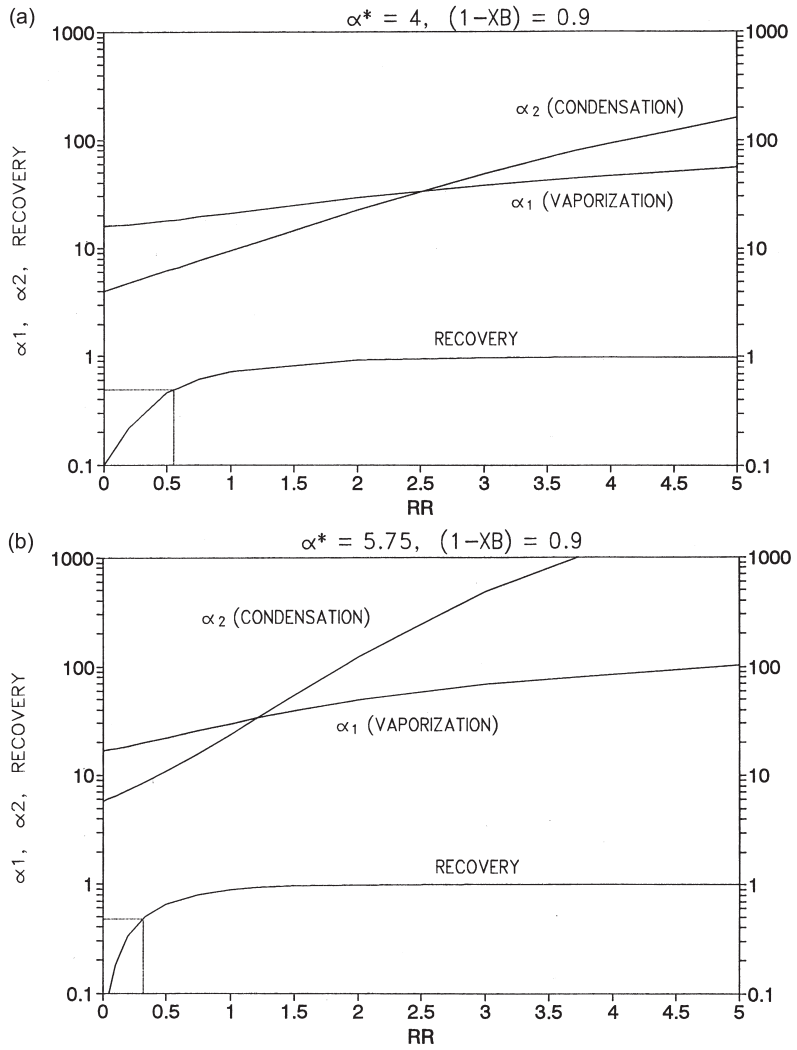


Figure 12. Stage separation factor and recovery as a function of RR to produce $(1 - x_B) = 0.9$, for the recycle DCDV process, when (a) $\alpha^* = 4$ and (b) $\alpha^* = 5.75$.

can be calculated using the Fenske–Underwood equation:

$$N_{\min} = \frac{\ln[y_P(1 - x_B)/x_B(1 - y_P)]}{\ln \alpha^*} \tag{12}$$

The results of calculations of $N_{\min}$ for 50% recovery and $(1 - x_B) = 0.90$, 0.95, and 0.99 are presented in Table 2. As a result of these $N_{\min}$, a two-stage ordinary distillation recycle cascade cannot effect even the desired minimum separation.

The actual number of theoretical stages required to make the specified separation can vary from $N_{\min}$ to very large values as designs are made for $RR \rightarrow RR_{\min}$. The actual design is usually dictated by economic factors, which is beyond the scope of the present study. However, calculations were made for distillation cascades to make some of the separations of interest, assuming constant relative volatilities of 4 and 5.75, and using what is thought to be "reasonable" RRs to illustrate the differences between ordinary distillation and the DCDV process.

Calculations were made for ordinary distillation to produce 0.90, 0.95, and 0.99 bottoms product with $R^{O_2} \geq 0.50$, assuming a vapor feed with "no-mix" at the feed stage, that is, $y_{F-1} = z_F = 0.79$. This design also assumes that stage N is a partial condenser, i.e., $H_N = P$, with composition y_P. A schematic diagram of this distillation cascade is shown in Fig. 13.

The designs were limited to an integral number of stages, and as a result, the recovery varied from about 0.56 to 0.82. The results of these calculations are summarized in Table 3.

With $\alpha^* = 4$, ordinary distillation would require from seven to nine ideal stages with RR = 0.45, 0.50, and 1.17 to produce $(1 - x_B) = 0.90$, 0.95, and 0.99, respectively. The total interstage heads rate (TISHR) (vide infra) varied from about 395 to 580 (based on a feed rate of 100), and ratio of reboiler boil-up rate to bottoms product varied from about 1.4 to 4.7.

With $\alpha^* = 5.75$, all of the desired separations could be accomplished with five theoretical stages with ordinary distillation, with RR between about 0.356 and 0.710. The designs investigated would require TISHR of about 144–260 (feed rate = 100) and a ratio of reboiler boil-up rate/bottoms product rate between about 1.1 and 3.1.

The comparable process variables for the DCDV process are:

Table 2. Minimum Number of Stages for 50% Oxygen Recovery for Ordinary Distillation of Air, $\alpha^* = 4$ and 5.75

	$N_{\min}$	
Bottoms Composition $(1 - x_B)$	$\alpha^* = 4$	$\alpha^* = 5.75$
0.90	3.03	2.40
0.95	3.57	2.83
0.99	4.77	3.78

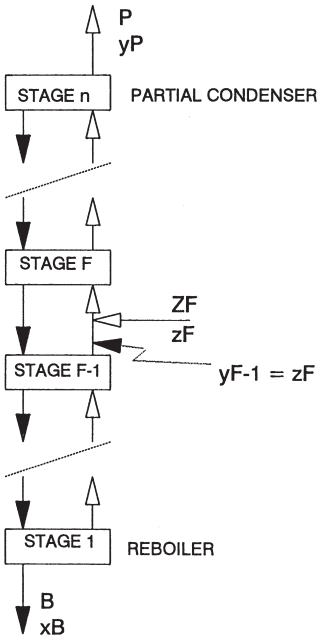


Figure 13. Schematic diagram of an ordinary distillation cascade with no-mix at the feed stage and a partial condenser.

(a) $\alpha^* = 4$, TISHR varies from 138 to 178, while "reboiler boil-up rate"/bottoms product rate, i.e., vaporization rate/bottoms product rate, varies from about 3.2 to 7.3.
(b) $\alpha^* = 5.75$, TISHR varies from about 118 to 134, while the ratio of vaporization rate/bottoms rate varies from about 1.6 to 3.3.

Table 3. Comparison of the Recycle DCDV Process with Ordinary Distillation to Produce $0.99 \geq (1 - x_B) \geq 0.90$ with $R^{O_2} \geq 0.5$

	DCDV	Ordinary Distillation
$\alpha^* = 4$		
Number of stages	2	7–9
TISHR	138–178	395–580
Boil-up rate/B	3.2–7.3	1.4–4.7
$\alpha^* = 5.75$		
Number of stages	2	5
TISHR	118–134	144–260
Boil-up rate/B	1.6–3.3	1.1–3.1

Total interstage flow rates of heads or tails is a measure of the "size" of the process,[2] while the ratio of vaporization (boil-up) rate to bottoms product rate is a measure of the relative amount of energy input required to make the separation.

Thus, for $\alpha^* = 4$, the DCDV "process size" would be smaller than for ordinary distillation, since the required TISHR would be only about 30–35% of that required for ordinary distillation. However, the DCDV process would require from about 1.5 to 2.3 times more energy input as shown by a comparison of the vaporization to bottoms ratio. The differences would be smaller if $\alpha^* = 5.75$, since the required TISHR for DCDV is from about 68 to 82% of that required for ordinary distillation, while the vaporization to bottoms ratio for DCDV is from about 1.1 to 1.43 times that for distillation.

A detailed comparison of process equipment size with different α^* (i.e., pressure) would require the calculation of actual volumetric flow rates at process conditions, which is beyond the scope of the present study.

Possible Means of Experimentally Achieving DC and DV

Simple cascades containing an infinite number of stages are equivalent to differential stage separations, and in some cases, a relatively few simple cascade stages can approach the behavior of the differential process.[2] Single-stage batch (Rayleigh) distillation approaches the DV process. In addition, stage separations by gas diffusion where the flow through porous barriers occurs by Knudsen flow is akin to Rayleigh distillation since the high pressure stream is continuously depleted in the faster diffusing compound, and the low pressure gas (which is removed through the barrier) at any point is related through K, which is analogous to α^*.[7] The countercurrent version of differential partial condensation is probably approached in condensers where the vapor flows upward condensing on tube walls while the condensate flows downward on the tube walls.[8] Although experimental evidence is lacking to the author, the process occurring in some recent dephlegmator designs may approximate the DC process. To our knowledge, the DC and DV processes have not been applied to continuous flow distillation type separations and the design of equipment to carry out the two processes represents a real challenge. Conceptual designs for the two processes are presented in Fig. 14. These view the processes as a number for simple flash vaporization and condensation stages in series, and as mentioned earlier, simple cascades containing "enough" equilibrium flash vessels in series could approach the performance of either the DC or DV processes.

These conceptual designs and their possible application in N-stage DCDV cascades will be discussed in more detail in Part II.

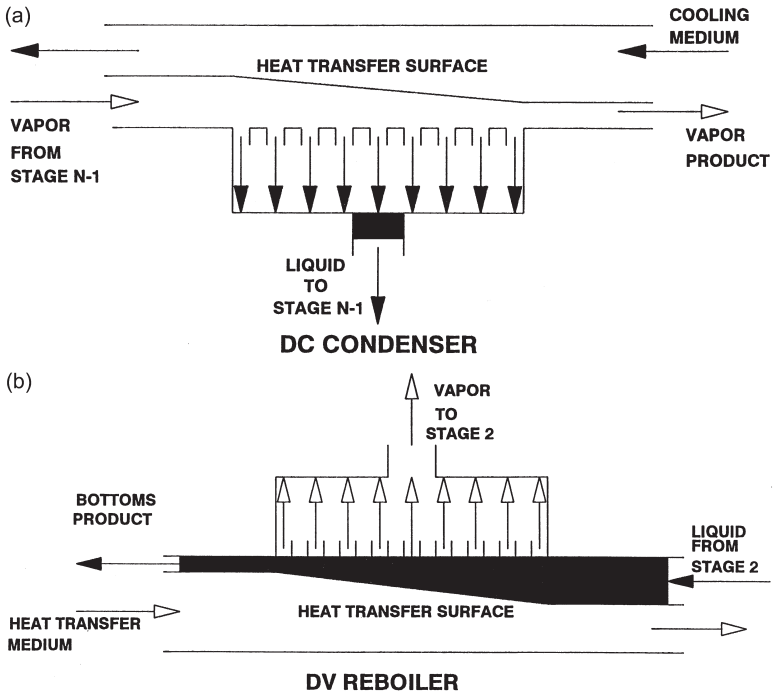


Figure 14. Schematic diagrams showing the concept for (a) DC and (b) DV cascade stages.

GENERAL DISCUSSION AND CONCLUSIONS

Ordinary distillation with stage (recycle) flow characterized by a countercurrent recycle cascade can produce any desired separation, provided $\alpha^* > 1$, and provided the cascade is designed with enough stages and with a high enough recycle rate to each stage. Theoretical calculations show that stage separation factors for the DCDV cascades can be significantly higher than for ordinary (equilibrium stage) distillation and thus fewer stages are required to carry out a specified separation. The two-stage DCDV recycle cascade can theoretically make any desired separation of a binary mixture with $\alpha^* > 1$, but very high recycle ratios are required for α^* less than about 3. If α^* is high enough, the two-stage DCDV cascade may be practical for certain applications, and theoretical calculations suggest that the process may be feasible for the recovery of oxygen from air for use in hypersonic flight. Indeed, at $P = 0.2$ atm (where $\alpha^* > 5.75$) air could be separated to produce 90% O_2 with oxygen recovery greater than 50% by DC followed by DV without recycle. Higher

enrichment and recovery can be achieved using the recycle two-stage DCDV cascade, but more energy (higher recycle ratio) may be required than for *N*-stage (5–9 stages) ordinary distillation at optimum recycle ratio.

The desired minimum separation for ACES cannot be achieved by two-stage ordinary distillation at any RR.

The two-stage DCDV requires a higher RR than the 5–9 stage ordinary distillation cascade, but the advantage of using the two-stage DCDV process is that process "size" as measured by TISHR could be significantly smaller than for ordinary distillation, an important consideration for the ACES application.

Depending on the magnitude of α^*, high RR may be required to achieve high stage separation factors in the DC and DV stages, and it will be interesting to compare RR, TISHR, energy input, etc., requirements for *N*-stage DCDV cascades compared with equilibrium cascades designed to make the same separation. The results of this study will be detailed in Part II.

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