Separation Science and Technology

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

Process Modeling of Laser Photochemical Reactor

L. M. Gantayet[a]; K. P. Madhavan[b]
[a] MULTIDISCIPLINARY RESEARCH SCHEME BHABHA ATOMIC RESEARCH CENTRE, BOMBAY, INDIA [b] DEPARTMENT OF CHEMICAL ENGINEERING, INDIAN INSTITUTE OF TECHNOLOGY, BOMBAY, INDIA

To cite this Article Gantayet, L. M. and Madhavan, K. P.(1992) 'Process Modeling of Laser Photochemical Reactor', Separation Science and Technology, 27: 6, 793 — 811
To link to this Article: DOI: 10.1080/01496399208019725
URL: http://dx.doi.org/10.1080/01496399208019725

SEPARATION SCIENCE AND TECHNOLOGY, 27(6), pp. 793–811, 1992

Process Modeling of Laser Photochemical Reactor

L. M. GANTAYET
MULTIDISCIPLINARY RESEARCH SCHEME
BHABHA ATOMIC RESEARCH CENTRE
BOMBAY 400085, INDIA

K. P. MADHAVAN
DEPARTMENT OF CHEMICAL ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY
POWAI, BOMBAY 400076, INDIA

Abstract

The applicability of conventional process engineering terms like value function, separative work unit, etc. to the laser isotope separation process has been established through appropriate formulation of the problem. The value function developed in this paper has been shown to be useful in arriving at guidelines for optimal design and operation of an isotope separation plant. A direct and simple model has been developed for a single-phase single-component laser photoreactor stage. This model, incorporating the key features and constraints of the laser isotope separation process, can provide an analysis and design basis to the process engineer.

INTRODUCTION

In recent years the field of laser photoreactions has seen intensive research which has led to new methods of synthesis of chemical and biochemical compounds, novel techniques of gas-phase catalysis, trace impurity removal, and highly efficient methods of isotope separation. Due to the high cost of a unit mole of photons, industrialized application of laser photochemical and photophysical methods has been possible only in the field of separation of isotopes (*1*), which is one of the most difficult separation tasks.

Photochemical and photophysical reactions have a unique feature, i.e., their selectivity. The selectivity of photoreactions makes feasible the manufacture of products which would be thermodynamically impossible by thermal means or would be deemed practically forbidden by kinetic considerations (*2*). Although it would seem probable that an ordinary light source properly filtered and optically manipulated would suffice for photoreactions, only the advent of lasers has removed the limitations of ordinary light sources; namely, in the degree of monochromaticity, tunability, intensity, spatial and temporal coherence, and short pulse widths (*3*). Besides, multiphoton dissociation of molecules by IR laser light has introduced a new regime of photochemistry and a range of process applications including laser isotope separation (LIS).

DISTINCTIVE FEATURES OF THE LIS PROCESS

The conventional separation processes are classified into "equilibrium" and "nonequilibrium" processes. Chemical exchange and vapor–liquid exchange are examples of 2-phase equilibrium separation processes. The laser isotope separation process does not fall into the category of thermodynamic equilibrium processes. And yet, unlike the other rate processes, the energy demand for the process itself is extremely small, thanks to the high selectivity of photoreactions. The isotopic shift in the spectrum and the consequent selectivity of the photon are utilized in achieving the separation. In the laser excitation process the time scales in which the steady states are attained are extremely small and are within the lifetimes of the excited states. The excited species is either dissociated, reacted, or ionized irreversibly within the stated time scales. In fact, if the excited species were allowed to come back to thermodynamic equilibrium with its surroundings, the selectivity or the separation factor would be lost (*3*). Thus, in the case of laser isotope separation we are dealing with a rate process where the monochromatic photons produced in short pulses are one of the reagents.

The growing importance of this field, given the possibility of rapid industrial applications, makes it necessary for a process engineer to evolve a systematic approach for analysis and design of laser photochemical reactors. In this paper we develop a formal approach toward analysis of isotope separation by laser photoreaction using conventional process engineering terms, namely, value function, separation factor, number of transfer units, countercurrent stages, etc. We have examined the possibility of defining an ideal separation factor similar to the maximum separation factor $\alpha_{\max} = (M_1/M_2)$ of the gaseous diffusion process or a maximum radial separation factor for a gas centrifuge $\alpha_{\text{radial}} = \exp(\Delta M v^2/2RT)$. An attempt is also made to describe the separation behavior in the usual y vs x curve, which will enable the process engineer to make preliminary design decisions and identify optimal operating conditions. The approach presented here could be used for making *a priori* comparison with other isotope separation processes.

VALUE FUNCTION AND SEPARATION DUTY IN LIS

Historically, the concept of value function and separative work unit, SWU, have been applied to hitherto known commercial processes having high throughput and separation factors close to unity. Thus it is commonly believed (*4–6*) that the value function and SWU are applicable to processes with small enrichment factors ($\epsilon = \alpha - 1$) and where ϵ is virtually independent of concentration. Many authors (*4–7*) are of the opinion that the value function and SWU are not applicable to LIS because of its large separation factor and because it is not used in an ideal cascade. In the following analysis it is shown that with proper formulation, the concepts of value function can be applied to LIS processes.

By definition, the separation duty of the separator or the value added by the separator is given by

$$\delta U = Pv(y) + Wv(x) - Fv(z) \tag{1}$$

where $v(i)$, $i = x, y, z$, is the value of 1 kg of mass. Here x, y, and z are, respectively, the compositions of W (tail), P (head), and F (feed) in a separator. The derivation of the value function will be shown below for two broad classes of separation regimes, namely, a close separation case and a large separation case.

Close Separation Case

In this case the separation factor (*8*)

$$\beta = \frac{y(1 - z)}{(1 - y)z} \tag{2}$$

is close to unity and thus $\beta - 1$ may be approximated as

$$y - z = (\beta - 1)z(1 - z) \tag{3}$$

Following Cohen (*8*), we use Taylor's expansion of $v(y)$ and $v(x)$ about $v(z)$ in Eq. (1) and obtain for $\theta = P/F = 1/2$

$$\delta U = F\frac{(\beta - 1)^2}{2} \tag{4a}$$

$$\frac{d^2v(z)}{dz^2} = \frac{1}{z^2(1 - z)^2} \tag{4b}$$

We solve Eq. (4b) with the initial conditions $v(z_0) = 0$ and $v'(z_0) = 0$. It is advantageous to write this solution as $v(z;z_0)$ where z_0 is a reference parameter. The solution

$$v(z;z_0) = (1 - 2z) \ln \left(\frac{1 - z}{z} \frac{z_0}{1 - z_0}\right) + \frac{(z - z_0)(1 - 2z_0)}{z_0(1 - z_0)} \tag{5}$$

assumes the following form for $z_0 = 0.5$,

$$v(z;0.5) = (2z - 1) \ln \left(\frac{z}{1 - z}\right) = \phi(z) \tag{6}$$

where $\phi(z)$ is the elementary value function, which is the same as the elementary separative potential defined by Brigoli (*9*).

Large Separation Case

In the large separation regimes we consider two cascades, one without a stripper and the other with a stripper section. For a cascade without a stripper, the value of waste is taken as zero. We now define

$$Y = \frac{y}{1 - y}, \qquad X = \frac{x}{1 - x}, \qquad Z = \frac{z}{1 - z}, \qquad Z_0 = \frac{z_0}{1 - z_0} \tag{7a}$$

and

$$\beta_1 = Y/Z, \qquad \beta_2 = Z/X, \qquad \alpha = \beta_1\beta_2 \tag{7b}$$

For $\beta_1 = \beta_2$, without any loss of generality, we can represent the separative duty of a separator stage by Eq. (1).

We can write the following material balance and value balance relations:

$$\frac{P}{F} = \frac{(1 + \beta Z)}{(1 + Z)(\beta + 1)} = \theta \tag{8a}$$

$$\frac{W}{F} = \frac{\beta(1 + Z/\beta)}{(1 + Z)(\beta + 1)} = 1 - \theta \tag{8b}$$

$$\delta U = \frac{(1 + \beta Z)}{(1 + Z)(\beta + 1)} v(\beta Z) + \frac{\beta(1 + Z/\beta)}{(1 + Z)(\beta + 1)} v\left(\frac{Z}{\beta}\right) - v(Z) \tag{8c}$$

To derive the expression for $v(Z)$, we shall follow the method of Cohen (*8*) and Pratt (*10*), which is based on the difference equation for stage processes. The main steps are given below. Assume that the separative duty of the separator is independent of concentration, Z, and that β is a constant. Therefore,

$$\delta U/F = C \tag{9}$$

The composition Z at any stage n is expressed as

$$Z = \beta^n Z_0$$

$$n = \frac{\ln [Z/Z_0]}{\ln \beta} \tag{10}$$

where Z_0 is the concentration of the stream entering Stage 1. Now setting

$$(1 + Z)v(Z) = (1 + Z_0\beta^n)v(Z_0\beta^n) = G(n) \tag{11}$$

The difference equation is obtained as

$$G(n + 1) - (\beta + 1)G(n) + \beta G(n - 1) = C(\beta + 1)(1 + Z_0\beta^n) \tag{12}$$

whose general solution can be written in the form

$$G(n) = C\frac{(\beta + 1)}{(\beta - 1)}n(Z_0\beta^n - 1) + A\beta^n + B \tag{13}$$

With the initial conditions

$$v(Z_0) = v(Z_0/\beta) = 0 \tag{14a}$$

and taking

$$C = \frac{\delta U}{F} = \frac{(\beta - 1)}{(\beta + 1)} \ln \beta \tag{14b}$$

the constants A and B in Eq. (13) can be evaluated to yield

$$v(Z) = (2z - 1) \ln \frac{z(1 - z_0)}{(1 - z)z_0} + \frac{\ln \beta[\beta - z_0(\beta + 1)](z - z_0)}{(\beta - 1)z_0(1 - z_0)} \tag{15}$$

The above mathematical definition of the value function is structurally the same as that for the close separation case. Because of the β-dependent term on the RHS of Eq. (15), many authors have concluded that the value function has any real significance only in the case of β close to 1. Pratt (*10*) suggested that if the value of waste in the stripper section is included, then the value function can be made independent of the term β.

For a cascade with a stripper section, we derive the value function for large β by changing the initial conditions. In setting up the new initial conditions, we must recognize that while dealing with valuable and scarce resources where recovery factors are important, the assumption $v(Z_0/\beta) = 0$ may not be valid. The new conditions are

$$v(Z_0) = 0, \qquad v'(Z_0) = 0$$

Using these conditions in Eq. (13), we obtain

$$A + B = 0$$

$$A = \frac{C(\beta + 1)}{\ln \beta(\beta - 1)}(1 - Z_0)$$

Further, using Eqs. (7) and (14b) we have

$$v(Z;Z_0) = (2z - 1)\ln \frac{z(1 - z_0)}{(1 - z)z_0} + \frac{(1 - 2z_0)(z - z_0)}{z_0(1 - z_0)} \tag{16}$$

The value function is now independent of β and is the same as that for the close separation case. Noting $Z_0 = 1$ for $z_0 = 0.5$, we get

$$v(Z;1) = v(z;0.5) = (2z - 1)\ln \frac{z}{(1 - z)} = \phi(z) \tag{17}$$

where $\phi(z)$ is the elementary value function. Since the value balance around the separator will remain the same whether $v(Z;Z_0)$, $v(Z;1)$, or $v(z;0.5)$ is used, $\phi(z)$ itself can be considered as a value function. In retrieving the expression for the elementary value function for large β, we had only set up the condition that the separative duty of the separator shall remain independent of the concentration. While deriving the expression for the elementary value function $\phi(z)$, we were not constrained by the nature of the process being nonequilibrium or not having low β. Thus, there is no loss of generality in applying it to the LIS process. Davis et al. (*11*), using a thermodynamicist's approach as shown below, concluded that the value function is a fundamental expression. For a binary mixture of species A and B,

$$\phi = x_A \frac{\partial s}{\partial x_A} + x_B \frac{\partial s}{\partial x_B}$$

where x_A and x_B are the concentrations and s, the specific entropy of mixing, is given by

$$s = x_A \ln x_A + x_B \ln x_B$$

It can be shown that the expression for ϕ used by Davis et al. can be put in the form of an elementary value function as in Eq. (17). Moreover, on examining the nature of the value function we find:

(1) The value function is independent of the separator's parameters.
(2) It serves the objective of quantitatively defining the work related to separation. It can be unambiguously combined with the material balance and energy balance around the separator. It provides a method of quantifying the value balance around the separator.
(3) This is a unique function and has a reference point; e.g., $\phi(z) = 0$ at $z = 0.5$ or $v(z_0;z_0) = 0$.
(4) For a given concentration, it is single valued.
(5) Although it may have the same value for two concentrations, because of the choice of the reference point, the values always add/increase when the material flows through the separator.

Since the value function fulfills the above-mentioned conditions, we can treat it like a state function associated with the streams entering or leaving a separator.

Role of Value Function for Optimal Cascade Operation

Let us now consider a separation plant (cascade) as shown in Fig. 1. The net value produced in the separation plant in terms of SWU per kg feed is given by

$$\Delta U/\text{kg } F = \theta v(y;z_0) + (1 - \theta)v(x;z_0) - v(z;z_0) \qquad (18)$$

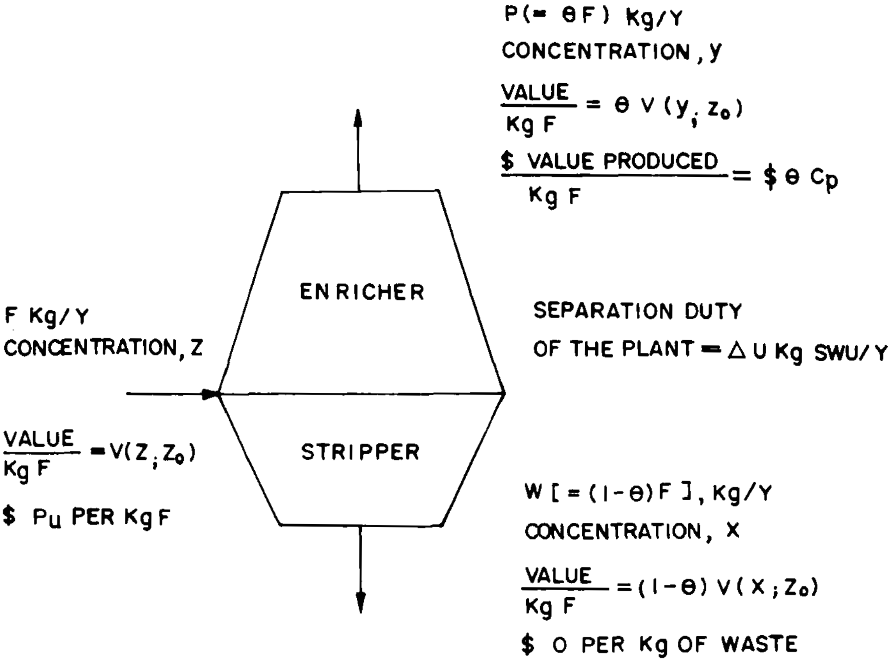

FIG. 1. Separation plant (cascade) of capacity ΔU SWU/Y.

and cost of product produced per kg of F processed in dollars is

$$\theta C_p = p_u + p_e(\Delta U) \tag{19}$$

where C_p = cost of product per kg P at concentration y
p_u = price of feed of concentration z
p_e = cost of unit SWU
$\theta = P/F$

Combining Eqs. (18) and (19), we get

$$C_p = \frac{p_u}{\theta} + \frac{p_e}{\theta}[\theta v(y;z_0) + (1-\theta)v(x;z_0) - v(z;z_0)] \tag{20}$$

because the value balance will not change if we write the values with x as the reference point. Further noting that $v(x;x) = 0$, we get

$$C_p = \frac{p_u}{\theta} + p_e[v(y;x) - v(z;x)/\theta] \tag{21}$$

Applying the criterion for optimal design/operation of the plant (*12*), namely, $\partial C_p/\partial x = 0$, one can show that this condition is equivalent to the criterion that no value should be lost through the stripper, and the cost of retaining the value of the feed should be equal to the cost of the feed (see Appendix A).Thus, the optimum condition for operation of the plant is

$$p_u = p_e v(z;x) \tag{22}$$

and the minimum cost of producing 1 kg of product at concentration y is

$$C_p = p_e v(y;x) \tag{23}$$

Equation (23) can be shown to give the same value of C_p as Eq. (20).

With this criterion we can also determine the limit to the price that can be paid for feedstock of concentrations z' which is different from the natural concentration. If p_e is the current market price of unit SWU and x_w is the lowest concentration of the desired atoms in the waste streams, then

$$p_{u,z'} = p_e v(z';x_w) \tag{24}$$

We suggest that x_w should be taken as the lowest concentration of the desired atoms encountered anywhere in the use of these isotopes. In the case of uranium, the lowest concentration of U^{235} occurs in the used fuel of SEU-fueled PHWR reactors, i.e., 0.09%. If we treat this as x_w, the range of concentrations of depleted uranium (DU) and reprocessed uranium (RU) can be priced as given in Eq. (24). This equation can also be used to set the economic goal of enrichment processes as shown in Table 1.

Table 1 is prepared by using as the base case the processing of a depleted feed of concentration z' to get a product concentration equal to that of natural uranium ($y = 0.0072$) and a tailing concentration of $x_w = 0.0009$. Since the product is equivalent to a natural feedstock, its price can be taken as the market price of natural feedstock. With the value of C_p known, the optimal value of p_e, p_e^*, can be calculated using Eq. (23). This value of

TABLE 1
Pricing of Feedstock of Different Concentrations[a]

Feedstock concentration, z'	$v(z';x_w)$	$p_{u,z'} =$ $p_e^* v(z';x_w)$ in \$	Product concentration, y	$p_e^* v(y;x_w)$ in \$
0.0025	0.76	15.4	0.0072	100
0.003	1.13	23	0.03	588
0.0072	4.94	100	0.03	588
0.009	6.58	133.5	0.03	588

[a] C_p = \$100; $x_w = 0.0009$. Thus p_e^* = \$20.30.

p_e^* is used to calculate the economic prices of varying feedstock concentrations given a tailing concentration x_w.

Thus we have shown that the value function and separative work unit (SWU) are powerful modes of representation of any separation process including the LIS processes, and that they help in quantitatively determining the relevant process and economic parameters.

MODELING THE LIS STAGE

We now discuss how the LIS stage itself can be modeled as a separator where the conventional process engineering concepts can be used.

Laser isotope separators are broadly of two types:

(A) Single-phase and single-component (chemical) reactors like those used in atomic vapor processes.
(B) Two-phase and multicomponent reactors where the reactant species changes phase or undergoes chemical reaction and the product is removed from the reaction site.

While modeling a reactor of Type A, it can be seen that the reactor will act like a flow splitter when the light is off, while in a multicomponent reactor of Type B the second phase/product will not occur when the light is off. Thus, Type B reactors generally cannot be modeled as flow splitters. Here we discuss the modeling of laser reactors taking the Type A reactor as an example. In a Type A reactor with the light off, the flow material balance is given by

$$F = P' + W' \tag{25}$$

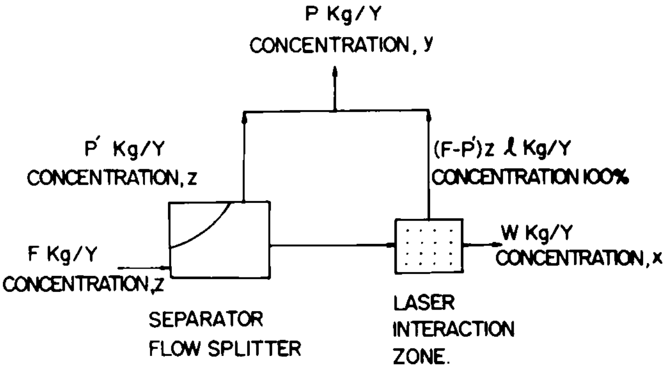

FIG. 2. Flow splitter model of Type A laser photoreactor.

and with the light on the material balance is

$$F = P + W \tag{26}$$

As shown in Fig. 2, the total desired species on which the light shines and interaction takes place is given by $(F - P')z$. The transport of the desired material from the feed stream to the product stream is $P(y - z)$.

Spectral and Process Separation Factors

A process engineer usually looks for achievable maximum separation factors. These are also known as ideal separation factors or separation factors limited by the physical laws of the separation process. In the laser-based processes an ideal separation factor or spectral selectivity is given in terms of the ratio of the laser photon absorption cross-sections of the species. Thus, the greater the isotopic shift, the greater will be the spectral selectivity.

To a process engineer the spectral selectivity, which is usually very high, matters insofar as the photon efficiency and the energy efficiency are concerned. The process separation factors are dictated by the population distribution of the reacting species which can interact with the laser photons. For example, in an atomic vapor the population of the reacting species is distributed over several electronic energy levels 0 (ground), e_1, e_2, e_3, . . . according to the Maxwell–Boltzmann distribution. The laser or lasers that shine on this vapor stream will be directed at exciting one or more levels to the subsequent excited states. If we, for the sake of simplicity, assume that two colors of laser light are used to excite levels 0 and e, then the maximum recovery of the desired species by photo interaction will be

$$\frac{n_0 + n_e}{n_{\text{total}}} = l_{(0+e)} \tag{27}$$

In order to determine the limiting separation factor, we shall assume that the excited species fraction, l, is removed from the reactant with 100% efficiency. Thus the maximum recovery, limited by the population which can react with the photons, is l:

$$l = \frac{Py - P'z}{(F - P')z} \tag{28}$$

Usually the feed composition, z, is the natural abundance of the reacting species, which in most cases is very small compared to 1, such that $P \cong P'$. Equation (28) then reduces to

$$\frac{P(y - z)}{Wz} = \frac{(z - x)(y - z)}{(y - z)z} = l$$

and

$$x = (1 - l)z \tag{29}$$

If the separation factor Z/X is expressed as β_2, then $\beta_{2,\text{ideal}}$ can be derived as follows.

$$\beta_{2,\text{ideal}} = \left(\frac{1}{1 - l} - z\right)\frac{1}{1 - z} \tag{30a}$$

$$= \frac{1}{1 - l} \quad \text{for } z << 1 \tag{30b}$$

If z is not small compared to unity, then P is not equal to P'. If the selectivity is very high (say infinite) and the collection efficiency is 100%, then the photoseparated species will quantitatively be equal to $(Py - P'z)$. The desired and total material balance then can be written as

$$Py = (F - P')zl + P'z \tag{31a}$$

$$P = (F - P')zl + P' \tag{31b}$$

Case I

If sufficient photons are available in the whole range of the feed concentration, then l is a constant in Eqs. (31a) and (31b). Combining the desired material balance with the total material balance equation, we get

$$y = z\frac{P' + (F - P')l}{P' + (F - P')zl} \tag{32a}$$

$$= z\frac{f + (1 - f)l}{f + (1 - f)zl} \tag{32b}$$

where

$$f = P'/F$$

It is to be noted that $y = 0$ for $z = 0$ and $y = 1$ for $z = 1$. The total and desired material balance can also be written in terms of W and x as follows:

$$W = F - P = (F - P') - (F - P')zl \tag{33a}$$

$$Fz - Wx = Py = (F - P')zl + P'z \tag{33b}$$

Now, by eliminating y in the material balance equation, we get

$$x = \frac{z(1 - l)}{(1 - lz)} \tag{34a}$$

$$z = \frac{x}{1 - l + lx} \tag{34b}$$

Here again, $z = 1$ at $x = 1$. Substituting for z in Eq. (32b), we obtain

$$y = x\frac{f + (1 - f)l}{f(1 - l) + lx} \tag{35}$$

Here also, $y = 1$ at $x = 1$. The behavior of the separator can be found from the above-mentioned equations as shown in Table 2. From Table 2 it can be seen that l has a stronger influence on SWU output per kg F than does f. At higher values of z, the SWU per kg F shows a marginal increase caused by increasing values of θ and a smaller difference between $v(y;z_0)$ and $v(z;z_0)$.

TABLE 2
SWU Output per kg F for Unlimited Photon Source

Feed	$f = 0.1$		$f = 0.15$	
z (%)	$l = 0.6$	0.8	0.6	0.8
0.32	0.64	1.24	0.56	1.11
0.72	0.64	1.24	0.56	1.11
1.0	0.64	1.24	0.56	1.11
3.0	0.64	1.24	0.56	1.11
10.0	0.66	1.26	0.57	1.11
50.0	0.75	1.34	0.61	1.13
70.0	0.79	1.38	0.63	1.14
95.0	0.85	1.43	0.66	1.14
Separation factor, α	16	41	11	27.7

Case II

If the availability of laser photons is limited in a system consisting of a separator and a photon source, then there will be a maximum limit to the quantity of photoseparated species. If the photons are constant and the flow through the separator is invariant, then $(F - P')$ will also be constant. Thus, for the regime $z < z_m$: $(F - P')zl < C_m$, and the x–y–z relations of Case I will apply. For the regime $z \geq z_m$:

$$(F - P')zl = C_m \tag{36}$$

Here z_m is the limiting feed concentration where the maximum fraction l of the species is excited and separated. Thus for $z \geq z_m$, we derive the following relations:

$$Py = P'z + C_m \tag{37a}$$

$$P = P' + C_m \tag{37b}$$

$$y = \frac{C_m}{C_m + P'} + \frac{P'}{C_m + P'}z = \frac{r}{r + f} + \frac{f}{r + f}z \tag{38}$$

where we have defined

$$r = \frac{C_m}{F} = \frac{\text{moles of photoseparated species}}{\text{moles of feed}}$$

$$= \frac{\text{moles of photons}}{n\ (\text{moles of feed})} \tag{39}$$

In Eq. (39), n denotes the number of photons absorbed per molecule or atom of the feed species. Rewriting the total and desired material balance with W and x as before, we can get the following relations for $z \geq z_m$:

$$z = \frac{C_m}{(F - P')} + \frac{(F - P') - C_m}{(F - P')}x$$

$$= \frac{r}{(1 - f)} + \frac{(1 - f - r)}{(1 - f)}x \tag{40}$$

Substituting for z in Eq. (38), we can get for $z \geq z_m$

$$y = \frac{r}{(r + f)(1 - f)} + \frac{(1 - f - r)f}{(r + f)(1 - f)}x \tag{41}$$

The behavior of the separator for the Case II can be calculated from the above-mentioned equations as shown in Table 3. Here it is seen that beyond the limit z_m the separation factor as well as the SWU output decreases rapidly compared to the unlimited photon case. For both the limited and unlimited cases, the y vs x behavior calculated from Eqs. (35) and (41) is

TABLE 3

SWU output per kg F with Limited Photon Source for $f = 0.1$, $l = 0.8$

Feed	$z_m = 0.0072$		$z_m = 0.0102$	
z (%)	α	SWU	α	SWU
0.40	41	1.24	41	1.24
0.72	41	1.24	41	1.24
1.02	14	0.57	41	1.24
5.0	2.3	0.039	2.95	0.071
10.0	1.6	0.012	1.89	0.022
50.0	1.12	0.0006	1.16	0.0011
95.0	1.06	0.0002	1.09	0.0003

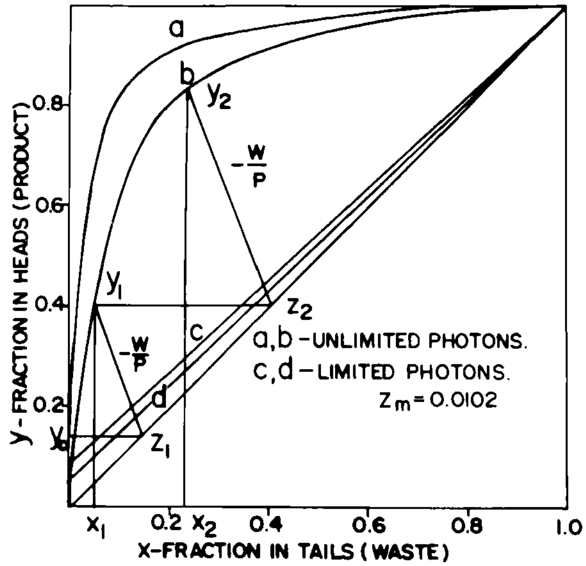

FIG. 3. y vs x behavior of Type A laser photoreactor. $f = 0.1$; for Curves a and c, $l = 0.8$; for Curves b and d, $l = 0.6$.

represented in Fig. 3. The effort per stage or per batch in a laser separator is dependent on the photon cost and also on the cost per kg of input feed through the separator. If the number of stages to attain a given separation objective are known, then the total effort can be easily estimated. The y vs x plot can be used to get the number of stages required for attaining a particular product in the manner described below. We construct this diagram for a case of the LIS separator being operated in a batch campaign mode. In this case the operating line is drawn from $z_1 = y_0$ on the 45° line with a slope $-W/P$, which becomes characteristic of a given separator. The intersection of the operating line with the y vs x curve is y_1, which becomes the feed z_2 for the next stage. The step off begins at $z_2 = y_1$ on the 45° line. This procedure can be continued to get the total number of stages.

It becomes quite obvious from the curve for Case II that the point of optimum operation is around z_m for optimum utilization of the separator effort as well as the laser effort.

CONCLUSIONS

The analysis presented in this paper has shown that the value function and separative work units can be used as valuable tools for analysis of laser separators. With a suitable formulation of value function as $v(z;z_0)$ and $v(z;x)$, a wider range of separator operation can be handled. An example of this is the development of simplified optimal conditions of operation of a cascade or a separation plant. We have also evolved a simple method of determining the maximum feedstock prices for a variety of concentrations above and below the natural concentration, which the separator operator can pay based on the prevailing market prices.

We have shown that the model developed for a Type A laser separator incorporates the limiting features of the photophysical/photochemical reactor. The ideal or maximum separation factor in the separator is governed on the one hand by the fraction of the population of the reactant which can be addressed by the highly selective photon stream and on the other by the flow splitting that takes place even when the photon stream is absent in the reactor (i.e., when the laser light is switched off).

Once the separator is designed and installed, its characteristics are well defined and are no longer flexible. In other words, the y vs x curve (behavior) of the separator is fixed for a given ratio range of photon moles to reactant moles. Apart from the various physical limitations to being able to vary this ratio over a large range, it is only practical to visualize that the separator will be installed with a given laser power pool. Therefore, the process engineer will have to find answers to such questions as "Given a laser isotope separator system, what is the optimum (economically competitive) method of operating the separator for a given separator duty for

Case A: When only the laser isotope separator is available?
Case B: When other separation cascades/plants like the ones based on the diffusive separation processes are also available to the operator?"

The model developed here gives pointers as to how the laser separators should be operated and can help in setting up the decision-making criteria. Our analysis of α and β dependence on z and P'/F and the analysis of y vs x behavior will provide answers to both questions. In Case A, if the entire separation is to be done by the laser separator, then in the regime of operation $0.5 < z < 1$ it would be more economical to excite the minor species of concentration $(1 - z)$ because the moles of photon requirement will be reduced and a higher separation factor will be obtained with the limited photon source. We observe that with knowledge of the behavior of y_A vs x_A (and y_B vs x_B) and the concentrations of Species A and B, an optimization strategy can be built for switching from one mode of excitation (exciting A) to the other (exciting B).

For a limited photon source, the laser separation effort increases for $z > z_m$. For a comparative study of the laser process with the other processes (as required for Case B), the y vs x curve could be used to get the number of theoretical stages. With knowledge of the cost of effort per stage for each process, a basis can be evolved for economic comparison.

For a countercurrent configuration, the usual stepping off method to find the number of stages, with which the process engineer is familiar, can also be utilized effectively here. The basic approach developed in this paper can also be extended to the study of the performance of real systems, where 1) selectivity has a finite value, 2) the parasitic ions are always present, and 3) the product removal efficiency is less than 100%. Thus, this simple and direct model of a separator can easily accommodate many different features of the laser separation process.

APPENDIX A

Equation (21) can be rewritten as

$$C_p = p_u \frac{y - x}{z - x} + p_e\left[v(y;x) - \frac{y - x}{z - x} v(z;x)\right] \tag{A1}$$

The concentration x, at which C_p will be minimum, is obtained by solving for $\partial C_p/\partial x = 0$.

$$\frac{\partial C_p}{\partial x} = 0 = p_u \frac{y - z}{(z - x)^2} + p_e\left[v'(y;x) - \frac{y - z}{(z - x)^2} v(z;x) - \frac{y - x}{z - x} v'(z;x)\right] \tag{A2}$$

Since

$$v(z;x) = (1 - 2z) \ln \frac{(1 - z)x}{z(1 - x)} + \frac{(z - x)(1 - 2x)}{x(1 - x)}$$

$$v'(z;x) = \frac{x - z}{x^2(1 - x)^2}$$

$$v'(y;x) = \frac{x - y}{x^2(1 - x)^2}$$

Equation (A2) can be shown to yield

$$P_u = p_e v(z;x)$$

Acknowledgments

The authors thank Dr. P. R. K. Rao, Head, MDRS, for his keen interest in this work and Dr. B. N. Jagatap for helpful discussions.

REFERENCES

1. J. I. Davis, "Lasers in Materials Processing," *AIChE Symp. Ser. No. 221, 78* (J. R. Merriman and M. Benedict, eds.), 1982.
2. A. E. Cassano et al., "Photochemical Reaction Engineering," *Ind.Eng.Chem.*, *59*(1), 18–38 (1967).
3. V. S. Letokhov, *Nonlinear Laser Chemistry*, Springer Verlag, New York, 1983.
4. G. Sauzay, "Some Characteristics of SILVA and Their Implications for the Nuclear Fuel Cycle," *Nucl. Eur.*, 9, 17 (1986).
5. G. Emanuel, "High-Enrichment Steady-State Cascade Performance," *Nucl. Technol.*, *43*, 314–327 (May 1979).
6. G. Emanuel, "Cascade Performance for Large Separation Factors," *Ibid.*, *51*, 238–243 (December 1980).
7. R. K. Lester, "Laser Isotope Separation for Uranium Enrichment: A Technology Assessment," PhD Thesis, Massachusetts Institute of Technology, 1979.
8. K. P. Cohen, *The Theory of Isotope Separation as Applied to Large-Scale Production of U-235* (National Nuclear Energy Series, Division iii, 13), McGraw-Hill, New York, 1951.
9. B. Brigoli, "Cascade Theory," *Top. Appl. Phys.*, *35* (1979).
10. H. R. C. Pratt, *Countercurrent Separation Processes*, Elsevier, New York, 1967.
11. J. I. Davis et al., *Science, Technology and the Industrialization of Laser-Driven Processes*, UCID-20448, Lawrence Livermore National Laboratories, May 1985.
12. Baumier et al., *Economie de l'energie nucle'aire*, INSTN. CENS, 1986 (in French).

Received by editor April 5, 1991