A LINEAR PROGRAMMING MODEL OF THE GASEOUS-DIFFUSION ISOTOPE-SEPARATION PROCESS

Donald Fort

P-1231

12 December 1957

The RAND Corporation
1700 MAIN ST. • SANTA MONICA • CALIFORNIA

:25)

A LINEAR PROGRAMMING MODEL OF THE GASEOUS-DIFFUSION ISOTOPE-SEPARATION PROCESS

Donald Fort

Introduction

The purpose of this paper is to show how the gaseous diffusion process for separating uranium isotopes may be treated as a linear programming model. The model is developed by the theoretical or engineering approach, as opposed to empirical methods based on statistical analysis of observations. The model may be useful in the economic analysis of complicated systems involving various feedbacks between gaseous diffusion plant, nuclear reactors, and other facilities involved in the production of nuclear materials and nuclear power.

This paper is in three parts. First is a brief description of the physical separation process. Next we show how the physical relationships may be translated into linear programming terms. Finally we shall indicate briefly how the linear programming model of the gaseous diffusion separation process may enter into larger models of the nuclear materials and power industry. Although the numerical values of the various parameters of the separation process are still highly classified, the relationships described here are generally known and unclassified.

The Gaseous-Diffusion Isotope-Separation Process

Before going into the details of the gaseous diffusion process, let us first take a look at the plant as a whole, viewed as a "black box". In the simplest mode of operation (Fig. 1), natural uranium containing 0.7% U^{235} and 99.3% U^{238} is fed, in the form of gaseous uranium hexafluorides, into the plant. The output of the plant consists of two streams: one, the desired product, is a stream of the gaseous material enriched considerably in the hexafluoride of U^{235}, and the other, the "tails", is the residue, correspondingly depleted in that component. Uranium material is conserved, insofar as possible, within the plant, so that what goes in must come out. The principal input to the plant, other than uranium material, is electric energy, used in large quantities to pump the uranium gas through the plant. The plant itself involves a heavy investment in highly specialized equipment.

A more complicated mode of operation may be called for because of feedbacks between the gaseous diffusion plant and other facilities such as nuclear reactors. An illustrative example of such a case is shown in Fig. 2. Several different compositions of enriched output are required, to meet the needs of fission weapons and various reactors which use enriched uranium as fuel. Moreover, the spent uranium from these reactors, containing a lower proportion of U^{235}, is available to be fed back into the gaseous diffusion plant, after the necessary chemical processing, if such a feedback is economically worth while. Some of the relationships between the gaseous diffusion plant flows and other facilities are illustrated in a later example. Such a system, involving many inputs and outputs and boundary conditions, is particularly appropriate for linear programming treatment.

Figure 1

External Material Flows of Gaseous Diffusion Process:

Simplest Case

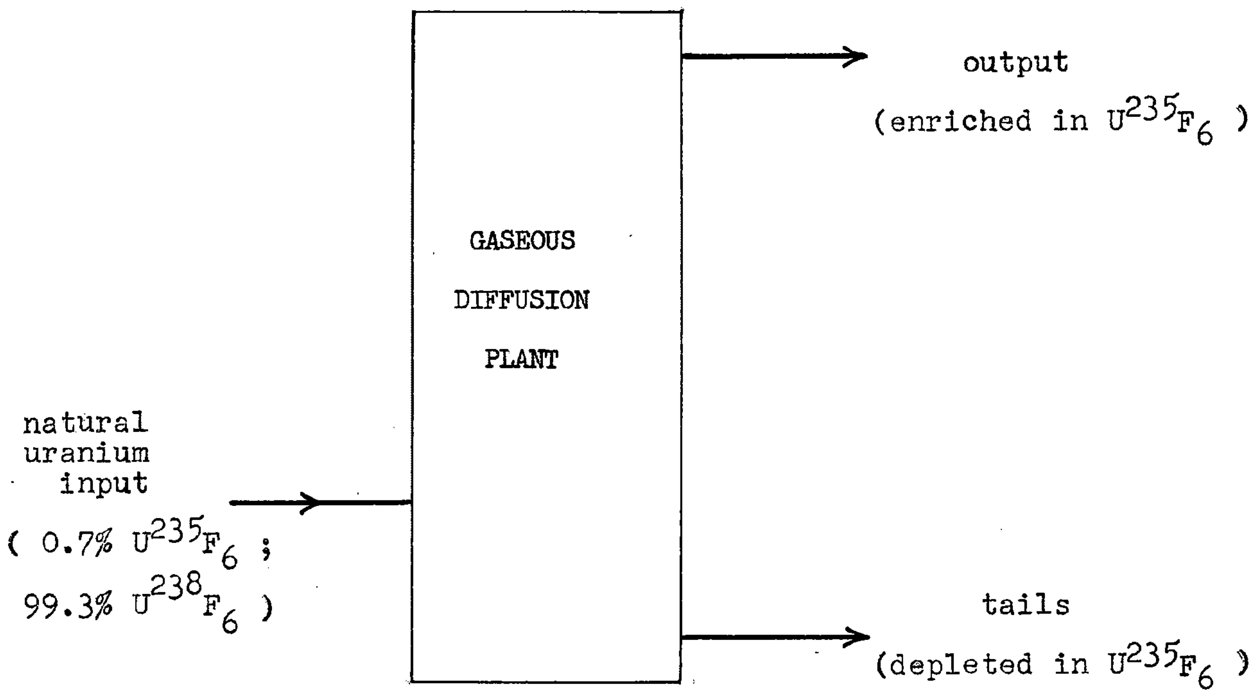
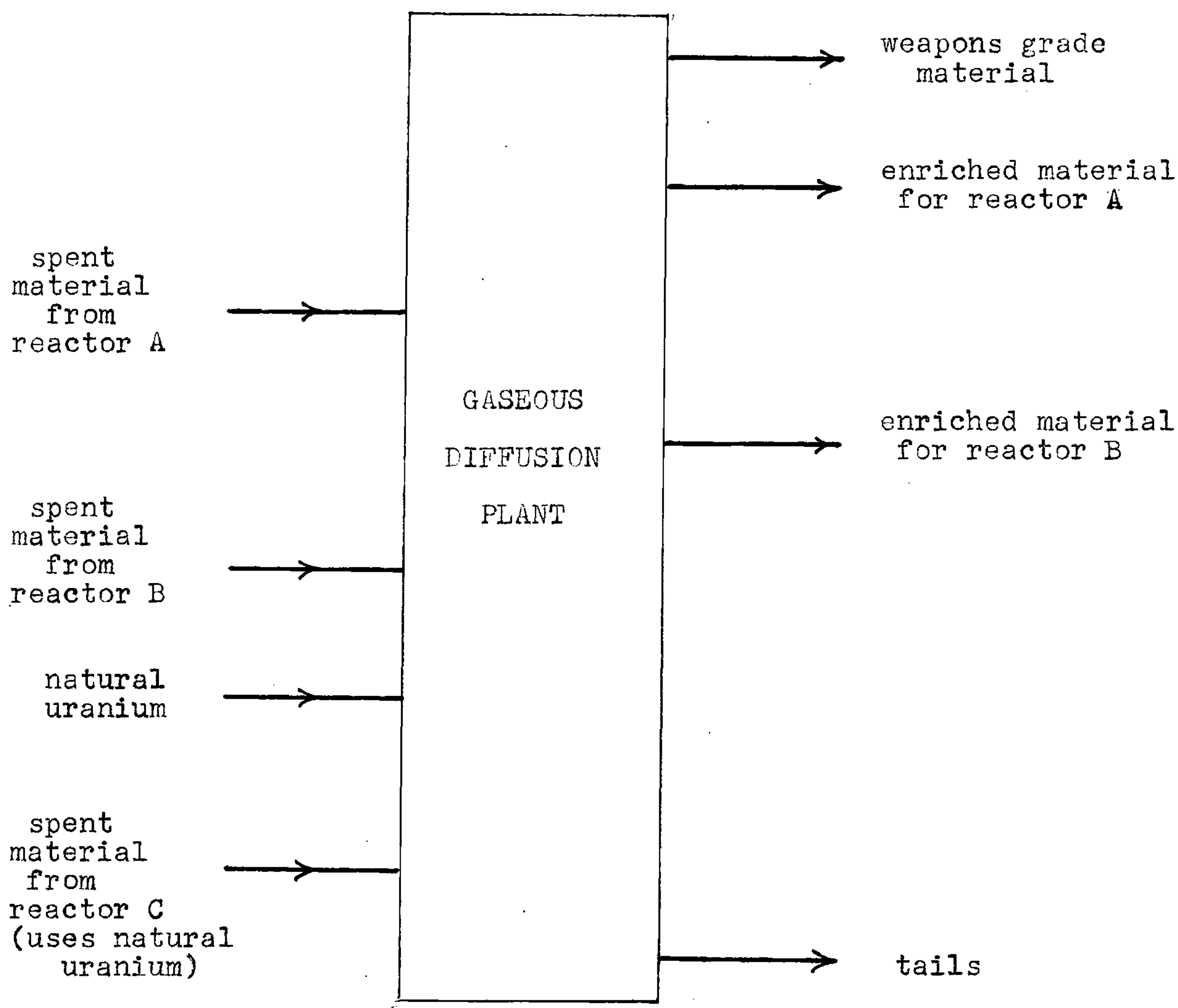


Figure 2

External Material Flows of Gaseous Diffusion Process:

Feedbacks From Nuclear Reactors



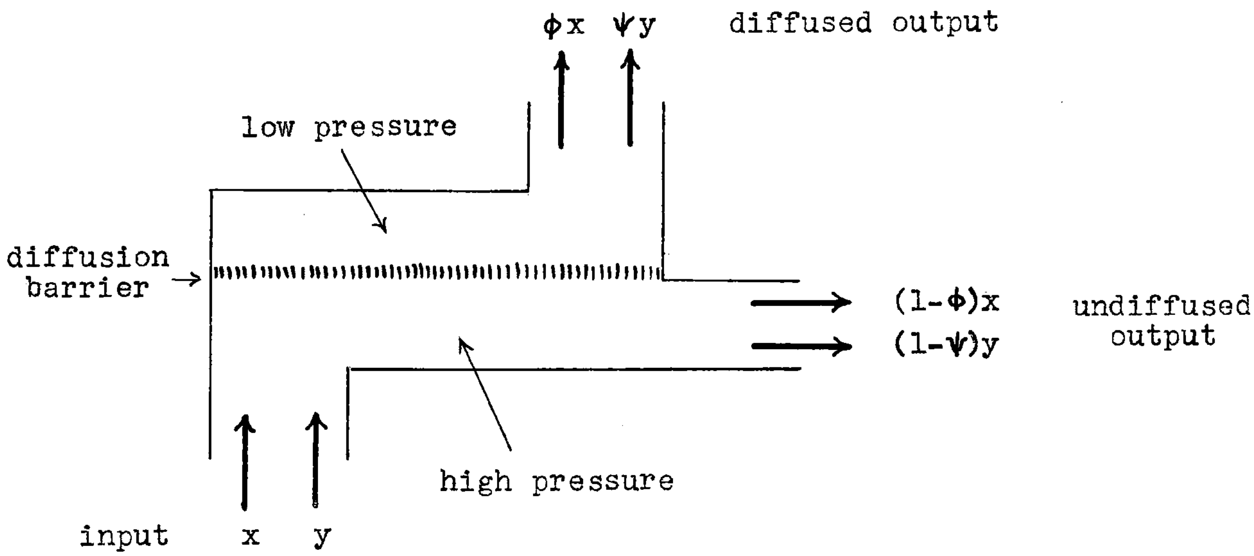
Our object, in constructing a model of the gaseous diffusion plant, is to portray the relationships between the quantities and compositions of the uranium material inputs and outputs, the quantities of the other inputs such as electric energy, and the arrangement and scale of the plant. To do this, we must look inside the plant.

The basic building block of the gaseous diffusion plant is the "separation element", shown schematically in Fig. 3. The gaseous mixture of components x and y (the hexafluorides of the two uranium isotopes) are pumped in under pressure at the left. As they pass along the porous diffusion barrier, some of both components diffuse through the barrier into the low-pressure side (above the barrier in the figure). The lighter molecules, being on the average faster than the heavier molecules, diffuse at a somewhat faster rate. That is, ϕ, the fraction of light component diffused, is slightly greater than ψ, the fraction of heavy component diffused. The undiffused material leaves the separation element at the right.

The "stage separation factor" $\underline{B}$ is defined as the ratio of the fractions of light and heavy components diffused, respectively. The factor $\underline{B}$ depends upon the total fraction θ diffused, according to equation (2) in Fig. 3. Here $\underline{a}$ is a parameter, very slightly greater than unity, whose value depends on the components being separated and the design of the separation element. The total fraction θ diffused can be given any desired value by appropriate adjustments of pumps and valves (not shown). The larger the fraction diffused, the smaller the degree of separation achieved by a given separation element. The optimum value of θ, and hence of B, is determined by considering the gaseous diffusion plant as a whole.

Figure 3

A. Separation Element Schematic



x moles of light component ($U^{235}F_6$) ϕ fraction diffused

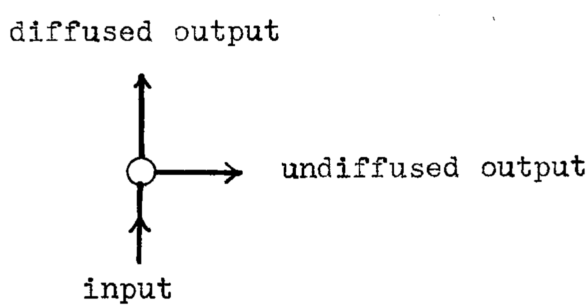
y moles of heavy component ($U^{238}F_6$) ψ fraction diffused

(1) θ total fraction diffused $= (\phi x + \psi y)/(x + y)$

(2) B stage separation factor $\equiv \phi/\psi = 1 + (a-1)\left(\frac{1-\theta}{\theta}\right)\ln\left(\frac{1}{1-\theta}\right)$

a separation element parameter

B. Separation Element or Stage: Simplified Schematic



For simplicity in the remaining diagrams, the separation element will be indicated schematically as shown at the bottom of Fig. 3. This representation will also be used to indicate a "separation stage", consisting of a bank of separation elements operated in parallel, each being fed the same composition of input and delivering the same compositions of diffused and undiffused outputs. For simplicity all separation elements in the plant will be assumed to be identical, although this is not necessarily the case in practice.

Because of the slight differences in the molecular weights of $U^{235}F_6$ and $U^{238}F_6$, the degree of separation achieved in a given separation stage is very slight, the theoretical maximum value of B being 1.0043, with a perfect separation element with an infinitesimal fraction diffused. In practice, the separation element is not perfect, and an appreciable fraction must be diffused, so that B will be more nearly 1.001 or 1.002. Thus, if a substantial degree of separation is desired, it is necessary to operate hundreds or even thousands of separation stages in series in a "separation cascade", in which the diffused output of one stage becomes the input of the next stage. For most efficient operation the undiffused output of a given stage must be fed back (refluxed) as an input to an earlier stage. The optimum refluxing arrangement has been found to be one in which the undiffused output of a given state is fed back as an input to the immediately preceding stage, and with a fraction θ diffused of almost exactly 0.50 in each stage. This arrangement, which avoids mixing materials of unequal compositions, is called the "ideal cascade". For given quantities and compositions of the various desired external input and output streams of uranium material, the ideal cascade arrangement uses fewer separation elements and associated equipment, holds up a smaller stock of uranium within the plant, and consumes less electric power, than any other arrangement. The value of B in the ideal cascade is approximately 1 + 0.69(a-1).

An illustrative example of an ideal cascade with only 12 stages, operated with one external input and two external outputs, is shown in Fig. 4. The composition of enriched output is determined by the number of stages in the "rectifying section" above the input point, and the composition of the tails is determined by the number of stages in the "stripping section" below the point of input. The number of separation elements (not shown) in each stage depends on the required flow rates, and is greatest at the point of input, tapering off from there in both directions.

A somewhat more complicated mode of operating an ideal cascade, with two inputs and three outputs, is illustrated in Fig. 5. Here there are four different "sections" bounded by the various external streams and denoted by the dotted boxes. The compositions of the various streams are determined by (or, conversely, determine) the number of stages in the various sections. More generally, there may be any number of inputs and outputs of different compositions, and in practice each section of the cascade will contain hundreds or thousands of stages. Moreover, each stage typically consists of a bank of many separation elements.

Before proceeding with the general case, one interesting point suggested by Fig. 5 is worth mentioning. In that figure, stream 3, an output, is flanked by input streams 2 and 4. Stream 3 is of composition intermediate between that of the two input streams. Thus the net effect of the overall process is to mix portions of the two input streams within the

Figure 4

Ideal Cascade With One Input and Two Outputs

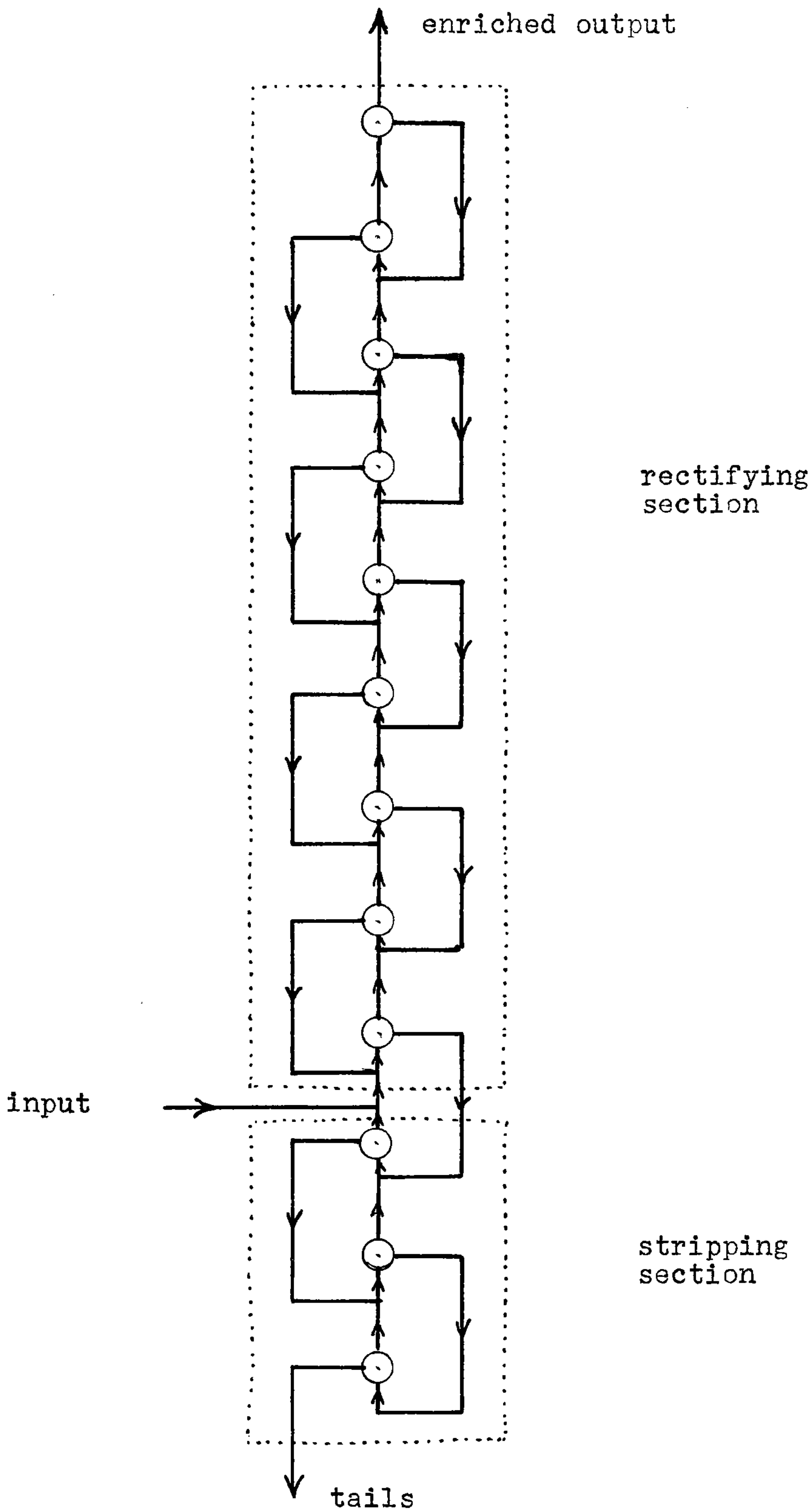
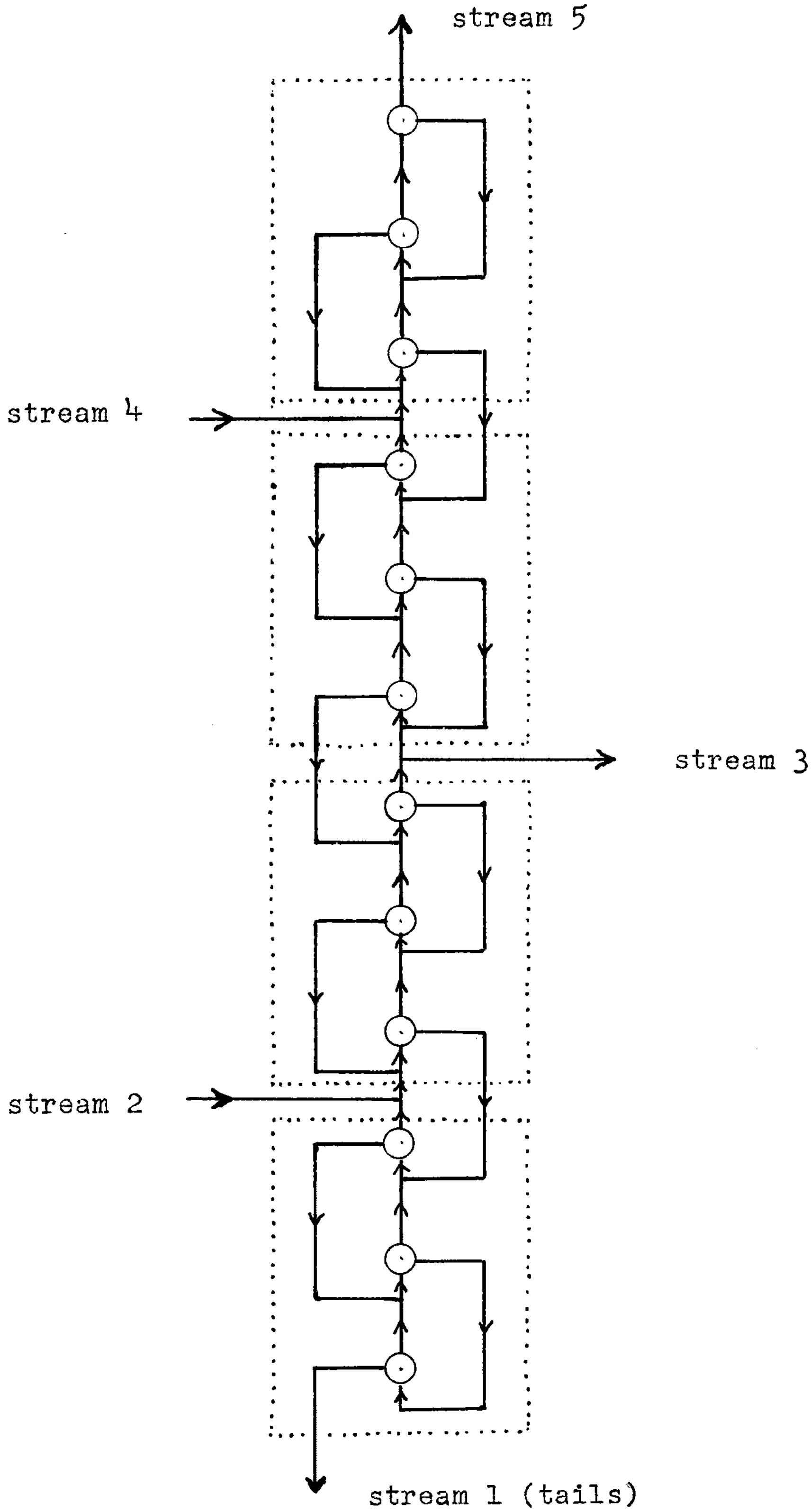


Figure 5

Ideal Cascade With Two Inputs and Three Outputs



cascade to produce the output stream 3 of intermediate composition.[1] At first glance such a process may seem obviously uneconomical, since it is not necessary to use such elaborate facilities simply to mix materials. Output 3 could indeed be obtained by mixing the required portions of streams 2 and 4 outside the cascade, feeding only the remainder of streams 2 and 4 into the cascade to be separated into the streams 1 and 5 of more extreme compositions. However, it can be shown that it costs less in terms of separation plant capacity and electric power consumption to perform the mixing within the cascade rather than outside. An intuitive explanation is that the mixing activity, being the opposite of separation, cancels out some of the separation activity required to produce the streams 1 and 5. Whether or not, all things considered, it will pay to do the mixing within the cascade will depend on how the savings in separation costs compare with the extra costs of chemical processing required for the inputs and outputs of the separation process.

The general case of the ideal cascade is portrayed schematically, again in black-box terms, in Fig. 6. The molar flow rates of the various external streams are denoted by Z_1, $Z_2,\ldots,Z_m$, positive if outputs and negative if inputs. The mole fractions of light component in the various streams are denoted by N_1, $N_2,\ldots,$ N_m, respectively. There are three sets of relations that the cascade must satisfy. The first of these are the requirements for conservation of uranium material within the cascade, under steady state operation. These can be expressed as two material balance

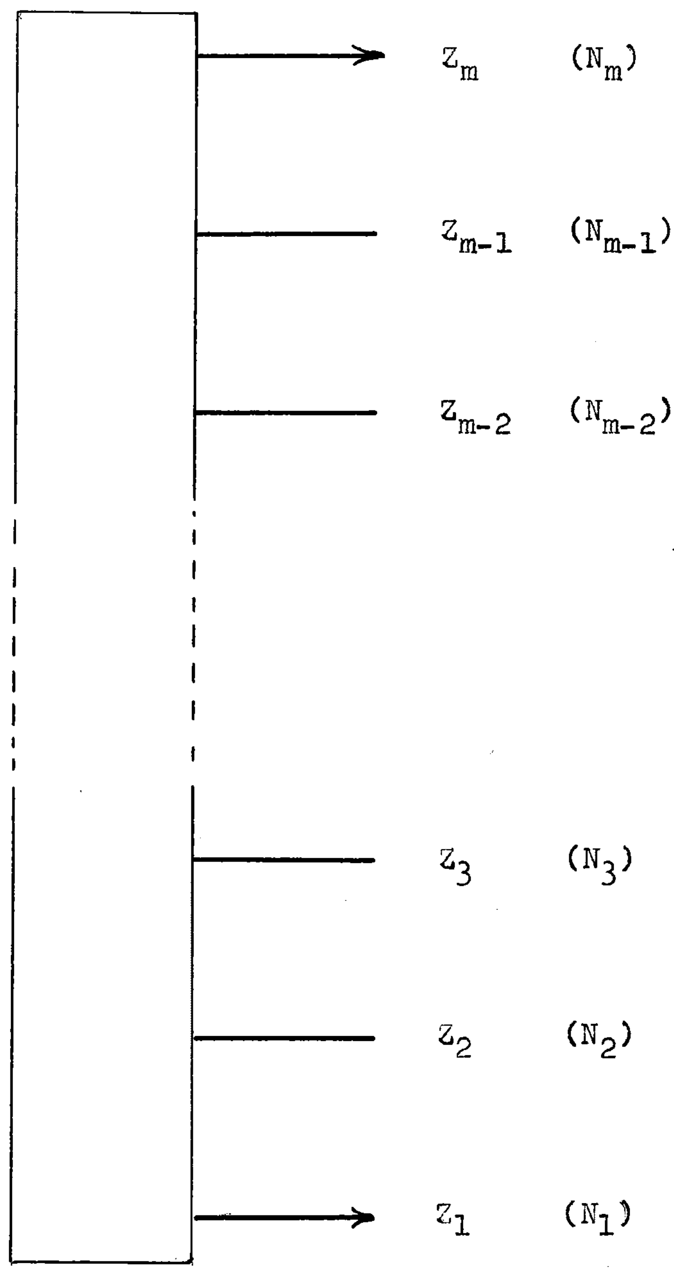
1. Although the overall effect is one of mixing, at no point within the cascade is there any mixing of materials of different compositions.

Figure 6

Ideal Cascade: General Case

Z_i molar flow rate of stream i

N_i mole fraction of light component in stream i



equations:

(3) $$\sum_{i=1}^{m} Z_i = 0$$

(4) $$\sum_{i=1}^{m} Z_i N_i = 0$$

The first equation refers to total-material balance and the second to light-material balance.

A second set of relationships concern the capacity of the cascade. A given set of external streams of given compositions imply given flows of material through the various stages of the ideal cascade. The sum of the molar diffusion flow rates in all stages, denoted by D, is given by the following equation:

(5) $$D = \frac{1}{(B-1)^2} \sum_{i=1}^{m} Z_i (2N_i - 1) \ln\left(\frac{N_i}{1-N_i}\right) .$$

D is a measure of the cascade capacity required to achieve the external flows in question. Each separation element in the cascade is characterized by a given diffusion flow capacity. It is either used at capacity or not at all. Thus D is a measure of the total number of separation elements that must be in operation to achieve the given external flows. The quantities of pumps, valves, plumbing, etc., are very nearly proportional to the number of separation elements, and so is the electric power required. Hence D is a reasonably good measure of the scale of all diffusion plant facilities, as well as power consumption, required to achieve a given set of external flows. If the cascade capacity is given, equation (5) establishes bounds on the magnitudes and compositions of the various streams consistent with that capacity.

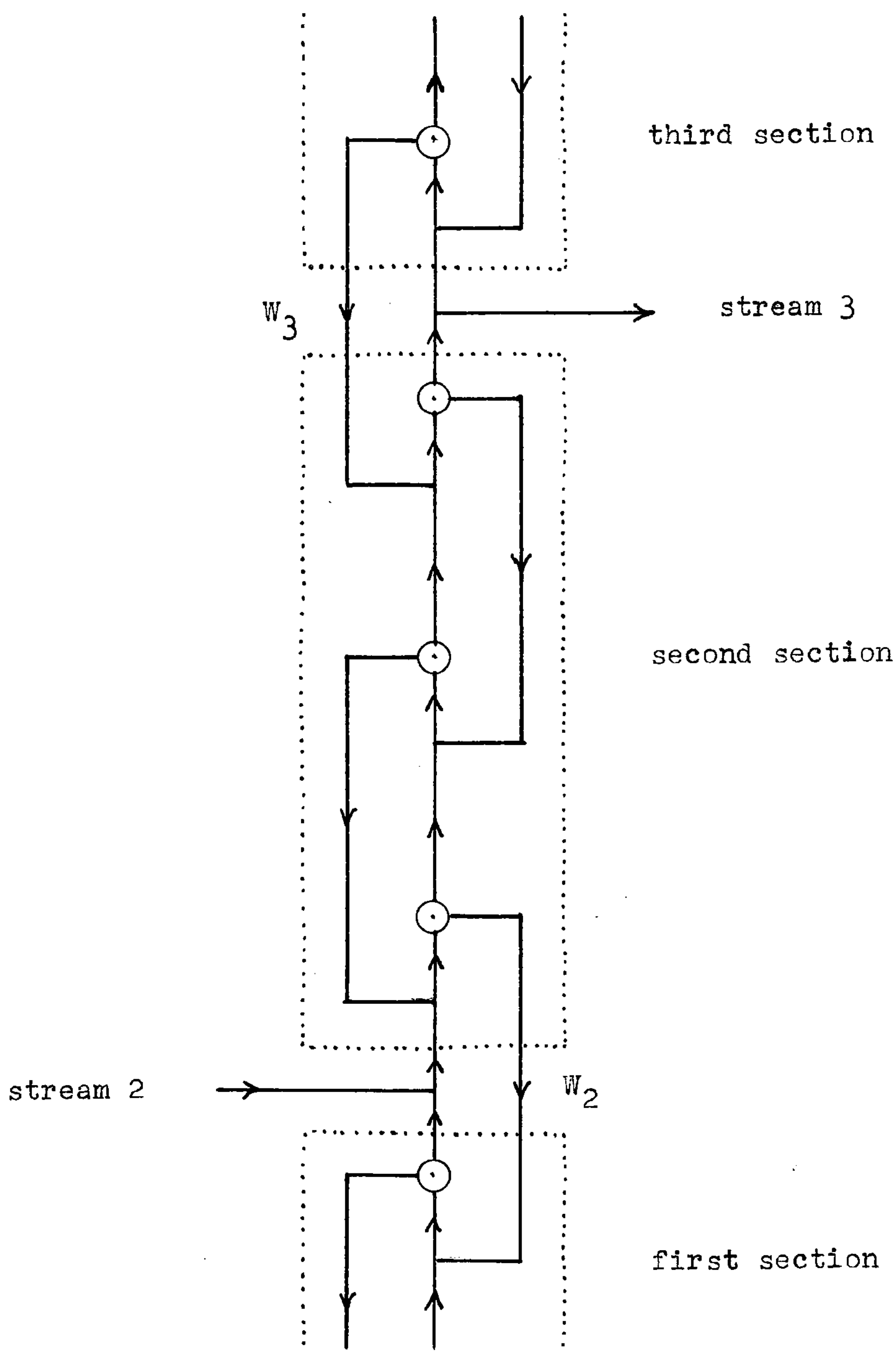
The third set of relationships are concerned with the directions of flows of material <u>within</u> the cascade. Each separation element is a one-way device, and will never be operated in reverse. However there are some combinations of external streams, consistent with equations (3), (4) and (5), and conceivably interesting to the analyst, which imply reverse directions of flow through the separation elements of some stages. Such combinations must be ruled out by appropriate boundary conditions. Thus, in the example of Fig. 5, there is an upper limit to the magnitude of stream 3, relative to the magnitudes of streams 1 and 5, consistent with proper implied directions of flow within the cascade. Within that limit it is permissible to consider producing stream 3 entirely by "mixing" within the cascade, as previously mentioned. However, if this limit were exceeded it would really imply literal mixing within the cascade, in that it would imply some separation stages being operated backward as mixing stages. Thus if one wishes to consider producing material of the composition of stream 3 in excess of the limit in question, the extra material must be produced by mixing materials of the compositions of streams 2 and 4 <u>outside</u> the cascade.

In formulating the boundary conditions in question, the concept of "sections", as previously introduced, is useful. A section is defined as the set of stages bounded by two adjacent external streams. Thus, for example, four sections, outlined by dotted boxes, are shown in Fig. 5. The second section from the bottom in that figure is reproduced on a larger scale in Fig. 7, showing its connections to the bounding external streams and to the adjacent sections.

There are four streams entering or leaving the second section. For present purposes we are concerned with two of these, W_2 and W_3, which are "downflows" of the undiffused outputs of the bottom stages of sections two and three, respectively. It can be shown that a specification of the

Figure 7

Second Section of Ideal Cascade of Figure 5



quantities of the two downflows entering and leaving a given section is sufficient to determine the flows in each stage of that section; if the two downflows are actually flowing downward, proper directions of flow are implied in each stage of the section. This means that individual stage flows need be considered no further, and each section can be treated as a black box, as shown in Fig. 8.

Applying material balance relations to each section in Fig. 8, it can be shown that the specification of the between-section flows W_2, W_3 and W_4 is sufficient to determine the flow rates of the five external input and output streams. Thus, all external streams consistent with proper internal directions of flow in each stage can be generated by considering all possible combinations of non-negative values of W_2, W_3 and W_4. Analogous conclusions apply to the general case.

The general case is shown in Fig. 9, again treating each section as a black box. The following relationships must be satisfied in order that proper directions of internal flows are implied:

(6) $W_i \geqslant 0 \qquad i = 2, 3, \ldots, m-1$

For simplicity it is convenient to define variables U_i as follows, since, in the equations relating the external streams to the downflows, the downflows always enter in expressions of the form of the right hand side of the defining relation:

(7) $U_i \equiv W_i(B-1)N_i(1-N_i) \qquad i = 2, 3, \ldots, m-1$

In terms of the U_i the external stream-downflow relationships are:

$$\left\{\begin{aligned} Z_1 &= \frac{U_2}{N_2-N_1} \\ Z_2 &= \frac{U_3}{N_3-N_2} - \frac{U_2(N_3-N_1)}{(N_2-N_1)\,(N_3-N_2)} \\ Z_3 &= \frac{U_4}{N_4-N_3} - \frac{U_3(N_4-N_2)}{(N_3-N_2)\,(N_4-N_3)} + \frac{U_2}{N_3-N_2} \end{aligned}\right.$$

Figure 8

Sectional "Black Box" Treatment of Ideal Cascade of Figure 5

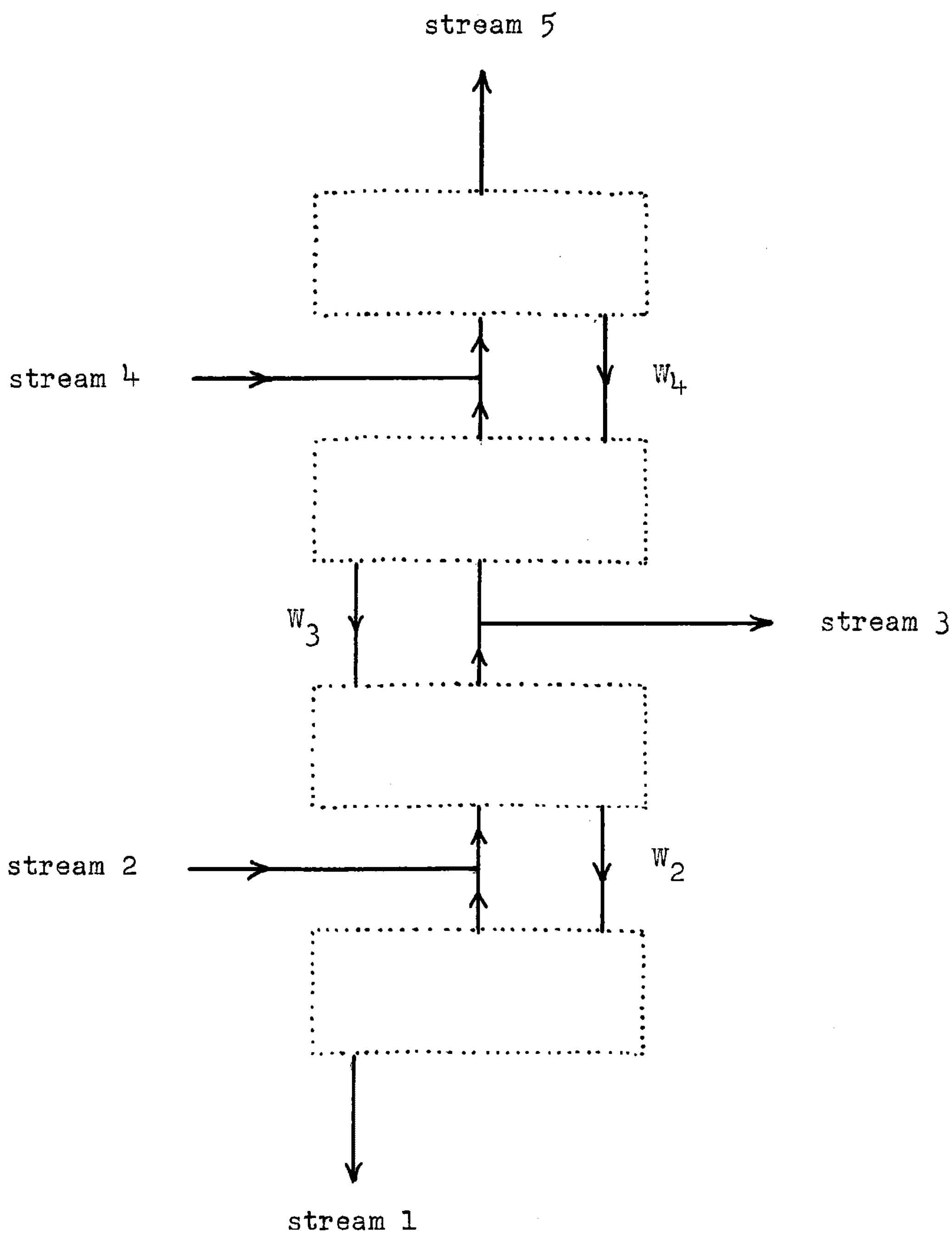
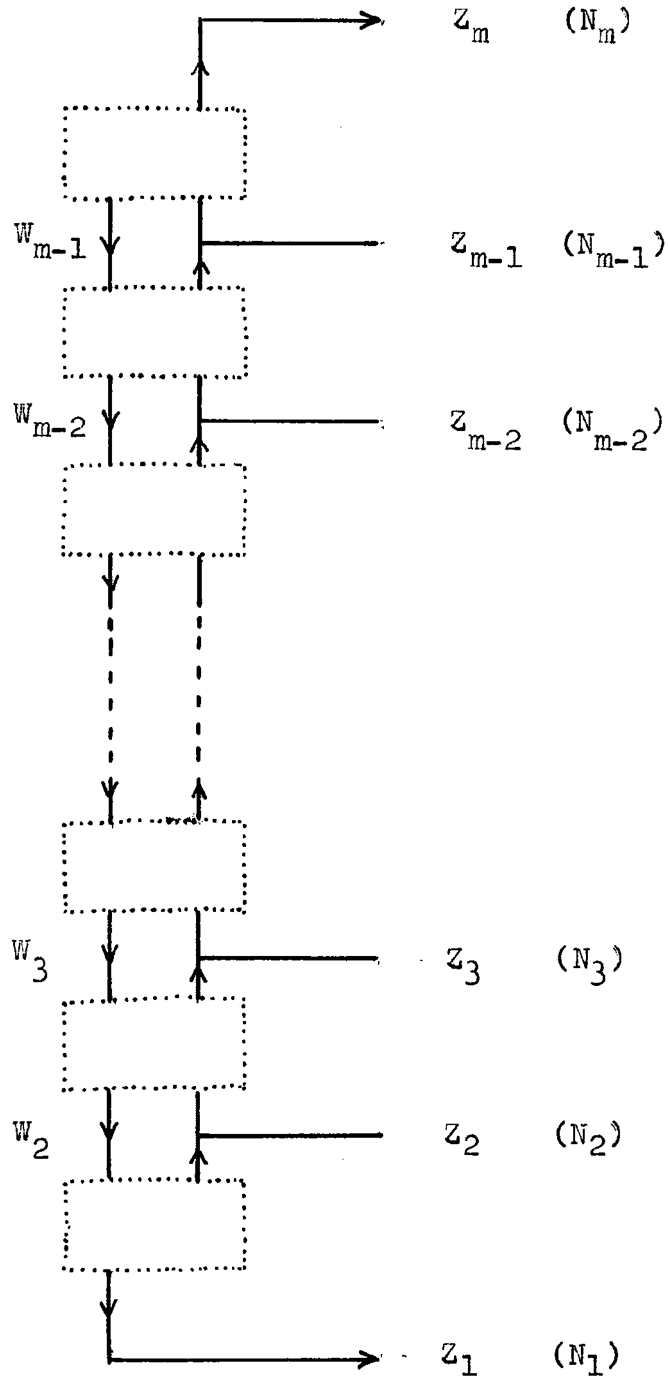


Figure 9

Sectional "Black Box" Treatment of Ideal Cascade: General Case



$$
(8)\quad \left\{
\begin{aligned}
&\vdots\\
Z_{m-2} &= \frac{U_{m-1}}{N_{m-1}-N_{m-2}} - \frac{U_{m-2}(N_{m-1}-N_{m-3})}{(N_{m-2}-N_{m-3})(N_{m-1}-N_{m-2})} + \frac{U_{m-3}}{N_{m-2}-N_{m-3}}\\
Z_{m-1} &= -\frac{U_{m-1}(N_m-N_{m-2})}{(N_{m-1}-N_{m-2})(N_m-N_{m-1})} + \frac{U_{m-2}}{N_{m-1}-N_{m-2}}\\
Z_m &= \frac{U_{m-1}}{N_m-N_{m-1}}
\end{aligned}
\right.
$$

For brevity in the remaining discussion it will be convenient to write set (8) as follows:

$$(8a)\qquad Z_i = \sum_{j=2}^{m-1} F_{ij}U_j \qquad i = 1, 2, \ldots, m.$$

where the F_{ij} are nonlinear functions of $N_1, N_2, \ldots, N_m$, as given by (8). Thus $F_{12} = 1/(N_2-N_1)$, $F_{13} = 0$, etc.

Since all terms in the coefficients of the W_i in (7) are positive, we have,

$$(9)\qquad U_i \geqslant 0 \qquad i = 2, 3, \ldots, m-1.$$

For any given set of external streams $Z_1, Z_2, \ldots, Z_m$ of given compositions $N_1, N_2, \ldots, N_m$, which are consistent with all the relations introduced above, there are no degrees of freedom left within the ideal cascade. That is, the number of stages, the number of separation elements in each state, and the diffusion flow rates in each stage are all completely determined. All degrees of freedom within the cascade have been optimized away by the assumption of the ideal cascade arrangement. The only degrees of freedom we are concerned with have to do with alternative combinations of external streams of various magnitudes and compositions.

The Linear Programming Model

All the equations needed for the linear programming model of the gaseous diffusion process have now been introduced. For convenience they are repeated here:

(3)	$\sum_{i=1}^{m} Z_i = 0$		total-material balance
(4)	$\sum_{i=1}^{m} Z_i N_i = 0$		light-material balance
(5)	$D(B-1)^2 = \sum_{i=1}^{m} Z_i (2N_i - 1) \ln(\frac{N_i}{1-N_i})$		diffusion capacity equation
(8a)	$Z_i = \sum_{j=2}^{m-1} F_{ij} U_j$	$i = 1, 2, \ldots, m.$	external flow-downflow relations
(9)	$U_i \geq 0$	$i = 2, 3, \ldots, m-1.$	boundary conditions for proper directions of internal flows

Since the U_i are nonnegative, and since they enter the various relations linearly, they may be used as activities. Eliminating the Z_i, equations (3) - (5) become:

(10) $$\sum_{i=1}^{m} \sum_{j=2}^{m-1} F_{ij} U_j = 0$$

(11) $$\sum_{i=1}^{m} N_i \sum_{j=2}^{m-1} F_{ij} U_j = 0$$

(12) $$D(B-1)^2 = \sum_{i=1}^{m} (2N_i - 1) \ln(\frac{N_i}{1-N_i}) \sum_{j=2}^{m-1} F_{ij} U_j$$

The F_{ij}, it will be recalled, are nonlinear functions of $N_1, N_2, \ldots, N_m$, of forms given by (8).

Equations (9) - (12) are linear in the U_i and in D. The diffusion capacity D must be nonnegative, and thus can be treated as an activity. Thus, for given external stream compositions $N_1, N_2, \ldots, N_m$, equations (9) - (12) constitute a set of constraints in a linear programming model involving activities $U_1, U_2, \ldots, U_m$, and D. The Z_i are not treated as activities, but in any application of the model they will enter the picture via equation set (8a), since it is largely through these external streams that the gaseous diffusion plant interacts with the rest of the economy.[1]

---

1. Note that (10) and (11) are identities. That is, the forms of the F_{ij}, as shown in set (8), are such that the material balance conditions are satisfied by any set of values of the U_i. Thus, on the basis of relations introduced so far, the U_i are constrained only by the requirements of nonnegativity (9) and consistency with permissible values of D, according to (12), if diffusion capacity is limited.

Applications

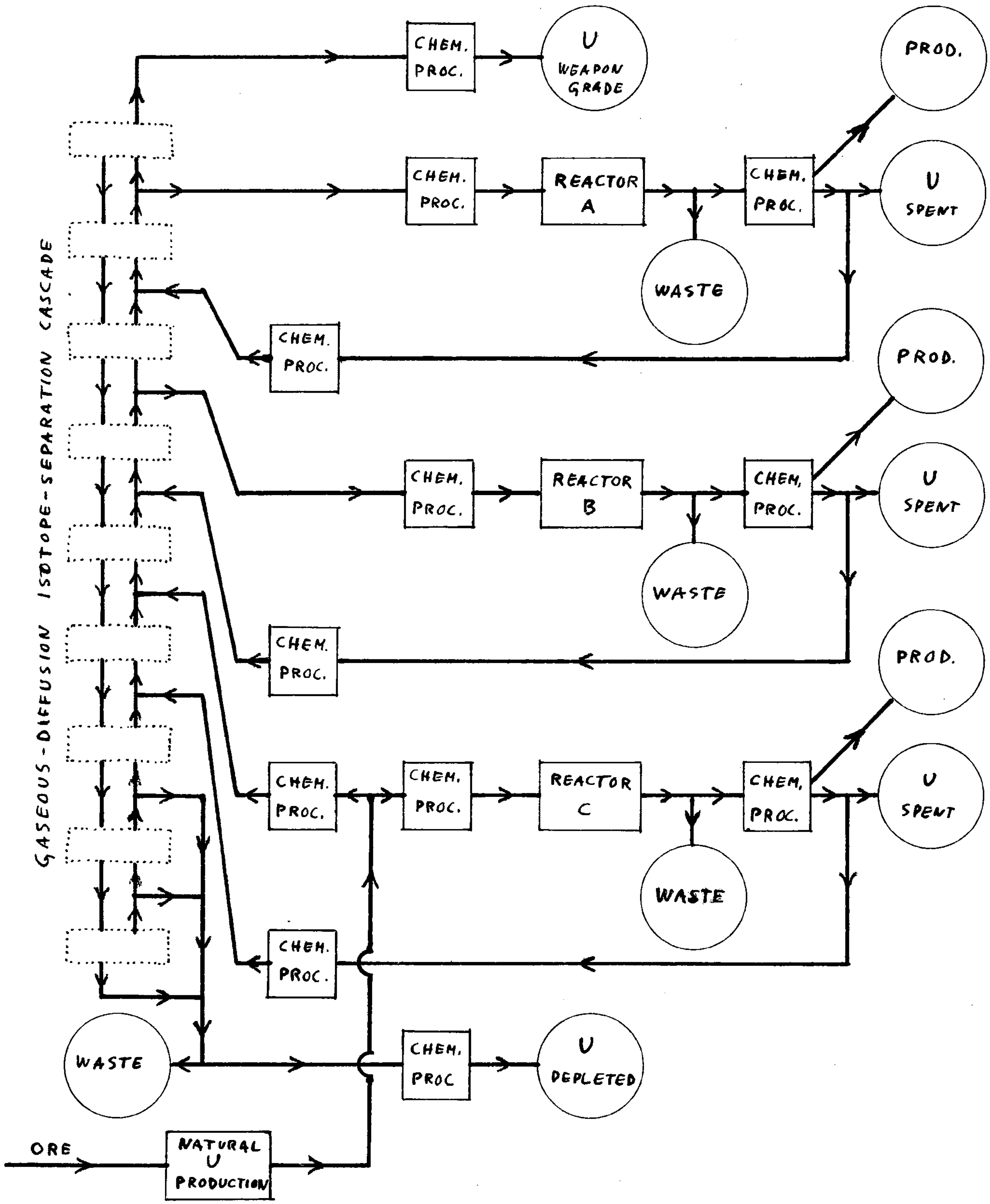
In the application of the model described above to economic problems, the external stream material may be treated as subject to given prices, subject to given bounds on the quantities that may be obtained or disposed of at given prices, or, as indicated later, the streams may be explicitly treated as linked to other facilities such as nuclear reactors. The total diffusion flow D may be taken as a measure of both the rate of consumption of electric energy and other resources (excluding uranium materials) by the plant, and the magnitude of the required plant investment.

The linear programming approach employed here does not permit treating the stream compositions as variables, since the N_i enter the various relations nonlinearly. However, in an important class of problems, the object may be, among other things, to determine the "best" composition for certain streams, such as the "tails". For such purposes the relevant N_i may be varied, in effect, by the following device. A small discrete number of stream compositions are specified, covering what is thought to be the region of interest. The linear programming routine is then employed to determine the "best" of these given compositions. If a more precise answer is desired, the process can be repeated, using a newly specified set of compositions grouped closely around the value obtained on the first approximation; and so on.

A hypothetical example of a larger model incorporating not only the gaseous diffusion plant but also other facilities is illustrated in the flow diagram of Fig. 10. The pattern of flows entering and leaving the gaseous diffusion plant correspond to those previously illustrated in Fig. 2. Three different types of nuclear reactors are shown, requiring different

Figure 10

Hypothetical Model of Nuclear Materials Interactions



compositions of uranium for "fuel", including one reactor using natural uranium. The outputs of material other than uranium from these reactors might include plutonium, useful radioactive isotopes, and various waste products. The spent uranium from the reactors may be either processed and fed back to the gaseous diffusion plant, or disposed of in other ways. Several different tails streams are shown for the gaseous diffusion plant, not because the plant would actually emit tails of more than one composition, but because several streams must be provided for in the approach previously described, if the question of the "best" tails composition is at issue.

Fig. 10 is incomplete as a representation of the activities that would have to be considered in an actual analysis. The inputs and outputs of thermal and electrical energy are not shown. Some of the chemical processing activities, shown separately in the figure, may actually use common facilities . The options of mixing certain streams outside rather than inside the cascade, in order to produce streams of intermediate compositions, are not shown. However, the example should give some idea of the potentialities and limitations of the approach we have described.