Permeation Cascades for the Separation of Krypton and Xenon from Nuclear Reactor Atmospheres

S. A. STERN
and
S.-C. WANG

Dept. of Chem. Eng. and Materials Science
Syracuse University, Syracuse, N.Y. 13210

Multistage permeation cascades have been designed for the removal of radioactive krypton and xenon from nuclear reactor atmospheres. These cascades could serve for the decontamination of the atmosphere within a reactor containment dome following a nuclear accident and for other applications of interest to the nuclear industry. The stages of the cascades are assumed to consist of permeator modules using silicone rubber capillaries as separation membranes. All stages are to be operated in a countercurrent mode with shell-side feed.

It is shown that it is possible to design an ideal cascade for the separation of multicomponent mixtures by matching the concentrations of a suitable key component in interstage streams that are mixed. This procedure minimizes the cascade volume and power requirement. It is also possible to design a cascade with constant *stage cuts* in its enriching and stripping sections that approximates the performance of an ideal cascade. The krypton and xenon content of a feed mixture containing about 1×10^{-3} mol% Kr and 1×10^{-2} mol% Xe in air can be lowered by factors of 10^3 and 10^8 respectively in a 27-stage permeation cascade. Methods of reducing the number of stages and the effects of irradiation on the membrane performance are also discussed.

SCOPE

One of the nuclear safety problems of great current interest is the separation of radioactive ^{85}Kr and $^{133\text{-}135}Xe$ from the atmosphere within a reactor containment dome after an accidental release of fission products. This problem could be a significant factor in building large power reactors in populated areas (Belter 1963; Blumkin et al., 1966), as was strikingly demonstrated by the accident at Three Mile Island near Harrisburg, Pa. The separation of radioactive krypton and xenon is also of interest for two other applications, namely, the decontamination of: 1) the off-gases from plants for reprocessing spent nuclear fuels, and 2) the gas that blankets the liquid-metal-cooled fast breeder reactor (LMFBR).

A promising separation technique which has been studied in connection with the above problems is the selective permeation of gases through nonporous polymeric membranes. This technique has made considerable progress during the last decade (Stern, 1972, 1976; Hwang and Kammermeyer, 1975). The separation of krypton and xenon by permeation from various gas streams has been studied in some detail by Rainey, Carter, and Blumkin (1971), who used silicone rubber membranes in sheet form as separation barriers.

More recently, Stern et al. (1978) investigated theoretically and experimentally the separation of a Kr-Xe-N_2-O_2 mixture by permeation through silicone rubber capillaries. The use of membranes in the form of capillaries, not in sheet form, offers two major advantages: 1) very large permeation areas can be packed per unit volume of permeator module, and 2) capillaries have sufficient strength to support themselves under elevated gas pressures and do not require porous supports as do sheet membranes. The use of capillaries instead of sheet membranes should, thus, substantially decrease the size of permeator equipment. The cost of permeators will then also decrease, provided that the cost of the silicone rubber capillaries is not a limiting factor.

Stern et al. (1978) studied the separation of the Kr-Xe-O_2-N_2 mixture in a single permeation stage, where the separation achieved was relatively small. The objective of the present study was to design multistage permeation cascades that would permit an extensive removal of krypton and xenon from the nuclear reactor atmospheres mentioned above.

Cascade feeds used in this study were chosen with the assumption that the cover gas from an LMFBR was essentially a Kr-Xe-Ar mixture and that the other atmospheres were Kr-Xe-N_2-O_2 mixtures. Argon or nitrogen and oxygen were taken to be the main components of these mixtures, with the latter two gases being in about the same ratio to one another as in air. The concentrations of krypton and xenon in the atmospheres of interest can vary widely because of the variety of conditions in which these gas mixtures can be generated. Therefore, the feed compositions used were selected to illustrate the removal of small amounts of krypton and xenon from mixtures with argon and air, not the processing of any specific atmosphere.

Correspondence should be addressed to S. A. Stern.

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CONCLUSIONS AND SIGNIFICANCE

The separation of radioactive krypton and xenon by selective permeation is a potentially attractive process for the decontamination of nuclear reactor atmospheres and fuel reprocessing off-gas because of its safety and reliability. The two helium-group gases, even when present in very low concentrations, can be removed to the desired extent in multistage permeation cascades using silicone rubber capillaries as separation membranes. The cascades can be designed so as to permit either the recycling of the processed atmospheres or their direct discharge to the surroundings. Permeator modules with silicone rubber capillaries, which form the stages of such cascades, are easy to build, rugged, and very compact. The compactness of permeator equipment may prove advantageous for the construction of mobile plants which could be transported to the scene of a nuclear accident.

It has been found possible to design *ideal* permeation cascades for the separation of multicomponent mixtures similar in concept to the ideal gaseous diffusion cascades devised for the separation of binary isotopic mixtures. Such permeation cascades are obtained by varying the stage cut from stage to stage so as to match the concentration of a suitable key component in interstage streams which are mixed. The *stage cut* is the fraction of feed allowed to permeate in a single stage. Ideal cascades minimize the cascade volume and power requirements. The performance of ideal cascades can be approximated by cascades with constant stage cuts in the enriching and stripping sections. The latter are easier to design and probably simpler to operate. The choice of stage cut must be found by trial-and-error with reference to an ideal cascade. Some of the concepts developed in this study have been explored independently by Pan and Habgood (1978).

The number of stages in a permeation cascade for krypton and xenon separation could be reduced by the use of two different kinds of membranes in each stage and by suitable stage and cascade recycling techniques (Kimura et al., 1978; Ohno et al., 1976-1978). These techniques could significantly lower power and capital investment costs. The permeability and strength of silicone rubber membranes is not appreciably affected by exposure to γ-ray irradiation at doses up to about 10^7 rads.

GENERAL CONSIDERATIONS

The separation obtained in a single permeation stage can be multiplied many times by connecting a number of stages in series to form a countercurrent permeation cascade. Each stage may be composed of a single permeator module or of several modules connected in parallel, depending on the volume of the feed, together with the necessary auxiliary equipment such as compressors and valves. The degree of separation achieved in a stage is determined by the performance of the permeator used.

During World War II, very large cascades were built for the Manhattan District Project to separate the uranium isotopes ^{235}U and ^{238}U by gaseous diffusion through porous barriers. The theory of isotope separation in such cascades is well developed (Cohen, 1951; Pratt, 1967). The design of permeation cascades, in which nonporous polymer membranes are used as separation barriers, is similar in principle to that of gaseous diffusion cascades. The two types of cascades differ, however, in the characterization of their ideal separation factors due to the difference in the mechanisms of gas transport through porous and nonporous barriers.

Additionally, the gaseous diffusion cascades were developed for the separation of binary isotopic mixtures with very small separation factors. As a result, it was possible to design *ideal* cascades, in which mixing interstage streams have the same composition, and the calculations could be simplified by assuming a continuous change in gas composition from stage to stage. An ideal cascade has the advantage of minimizing the total cascade volume as well as the power requirements; it does not necessarily minimize the capital investment costs, because each stage is of different size. Therefore, the ideal cascade must be *squared off* to reduce capital costs (Cohen, 1951; Pratt, 1967).

By contrast, in permeation cascades for the separation of multicomponent mixtures it is not possible to match the concentrations of all components in interstage streams that are mixed. These cascades can be *ideal* only with respect to some key component; that is, only the concentrations of a single component can be matched in mixing interstage streams. Moreover, the separation factors for the multicomponent mixtures of interest in the present study are much larger than those for isotopic mixtures. Consequently, it is not possible to assume a continuous change in composition from one stage to another.

Relatively little information is available on the design of permeation cascades for the separation of gas mixtures with large separation factors. Thus, Hwang and Kammermeyer (1965) have examined different types of cascades for the separation of binary mixtures, and Blumkin (1967, 1968*a*, 1968*b*) has designed permeation cascades for the separation of krypton and xenon from ternary and quaternary gas mixtures. Blumkin assumed that the stages in his cascades consisted of permeator modules using flat silicone rubber membranes and that these permeators were operated in a *cross-flow* mode (Stern 1972, 1976; Hwang and Kammermeyer, 1975).

More detailed analyses of permeation cascades have been reported recently by Ohno et al. (1977, 1978) and by Pan and Habgood (1978). Particularly pertinent to the present work is the theoretical and experimental study by Ohno et al. of a nine-stage recycling cascade for the separation of a Kr-N_2 mixture. This cascade was composed of permeator modules in which both silicone rubber and cellulose acetate capillaries were used as separation barriers (Ohno et al. 1978 *a,b*). Pan and Habgood (1978) performed a parametric study of permeation cascades for the recovery of helium from He-CH_4 and He-CO_2-CH_4 mixtures by means of cellulose acetate capillaries. Ohno et al. assumed *perfect mixing* conditions in the stages of their cascades (Stern, 1972; Hwang and Kammermeyer, 1975), whereas Pan and Habgood selected a *cross-flow* mode of stage operation.

CASCADE DESIGN SPECIFICATIONS

Three types of permeation cascades for removing radioactive ^{85}Kr and ^{133}Xe-^{135}Xe from the following atmospheres were considered in this study: 1) the atmosphere of a reactor containment building after a nuclear accident, 2) the off-gas of a nuclear fuel reprocessing plant, and 3) the cover gas of a liquid-metal-cooled fast breeder reactor (LMFBR).

In view of the fact that the half-lifes of ^{133}Xe and ^{135}Xe are short (5.27 d and 9.2 h, respectively) whereas the half-life of ^{85}Kr is relatively long (10.6 yr), this study was focused on the separation of krypton. Hence, krypton was taken to be the key component in cascade design. Due to the long half-life of ^{85}Kr, it was possible to use the magnitude of krypton concentration, not that

of its radiation, as a criterion of the necessary extent of separation.

The cascades described in the following sections are *ideal* in the sense that the concentrations of a single key component, krypton, are closely matched in interstage streams that are mixed. As mentioned previously, this procedure was adopted because it is not possible to match the concentrations of all components of a multicomponent mixture in such streams. An exact match of concentrations cannot be achieved even for a key component of a multicomponent mixture, as was reported also by other investigators (Thorman, Rhim and Hwang, 1975).

Therefore, in the following sections an ideal cascade is defined as one in which the mole-fractions of krypton in two or more mixing streams do not differ by more than 3%. It is shown that such a cascade, although based on the matching of a single component, is indeed *ideal* because it minimizes the volume and power requirements of the cascade. All cascade calculations presented herein can be scaled up as desired by multiplying the flow rates and capillary length by a suitable factor.

The stages of the cascades have been assumed to consist of permeator modules using silicone rubber capillaries as separation barriers. The construction of such permeators could be similar to that described by Stern et al. (1978). The silicone rubber capillaries were taken to have the nominal dimensions of 635 μm (2.5×10^{-2} in.) O.D. $\times$ 305μm (1.2×10^{-2} in.) I.D. These capillaries would be pressurized externally (shell-side). All stages would operate in the *countercurrent* mode. In other words, the high-pressure (unpermeated) gas streams outside the capillaries and the low-pressure (permeated) gas streams inside the capillaries would flow countercurrently to one another in every stage. No mixing would occur in the axial direction of flow and no concentration gradients would exist in the radial direction. This mode of stage operation yields the largest degree of separation and requires the smallest membrane area, or capillary length. The temperature of operation was selected to be 20°C and the pressure difference across the capillary walls, Δp, was taken to be 2.76×10^5 N/m^2 (40 lb/in.^{2}). Under these conditions, the effective permeability coefficients for krypton, xenon, argon, nitrogen, and oxygen are (Stern, Onorato, and Libove, 1977; Stern et al., 1978):

Gases	Effective Permeability Coefficients at 20°C: $\bar{P}^* \times 10^{15}$ (SI Units)‡	$\bar{P}^* \times 10^8$ [cm^3 (STP) · cm/(s · cm^2 · cm Hg)]
Xenon	73.1	16.6
Krypton	21.0	7.50
Argon	5.36	4.01
Oxygen	4.24	3.96
Nitrogen	1.72	1.84

‡ SI Units: kg$_m$ · m^2/(s · m^2 · Pascal)

Stern et al. (1978) have studied the separation of a Kr-Xe-O_2-N_2 mixture by means of silicone rubber capillaries as a function of pressure, temperature, and *stage cut* (the fraction of feed allowed to permeate in a single stage). They found that the permeability coefficients for pure Kr, Xe, O_2, and N_2, such as those listed above for 20°C., adequately described the permeation behavior of the mixture. Axial pressure drops inside and outside the capillaries were negligible when Δp was 2.76×10^5 N/m^2 (40 lb/in.^{2}), as is assumed in the present analysis.

CASCADE DESIGN PROCEDURES

The design of a permeation cascade requires stage-to-stage calculations, as in that of a fractional distillation column. These calculations are based, also as in distillation, on the use of

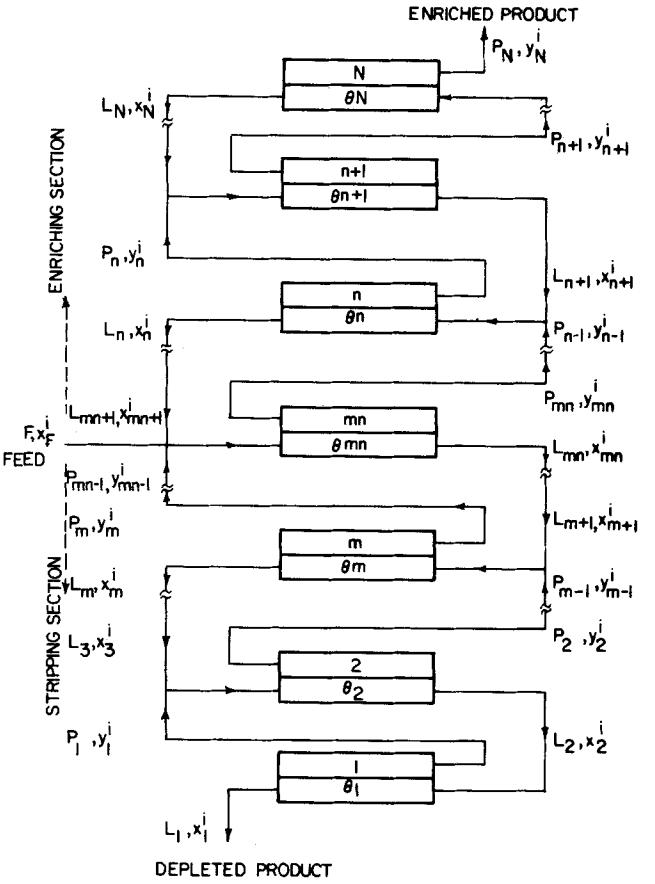

Figure 1. A permeation cascade.

equilibrium line and *operating line* equations. The equilibrium line equation relates the composition of the high- and low-pressure product streams leaving any given permeation stage. This equation is obtained from the rates of transport of the components of a specified mixture across the stage membrane. No phase equilibrium is involved in this case, and the term *equilibrium line* is used only by analogy with distillation.

The operating line equation relates the composition of passing interstage streams and is derived from material balances. If the compositions of the cascade feed and of the product streams from the *top* or *bottom* of the permeation cascade are specified, the compositions of all other streams in the cascade can be calculated stage by stage by alternately using the equilibrium and operating line equations. The total number of stages in the cascade is also obtained in this manner. The derivation of these equations and the general method of cascade design used in this study are outlined below.

Equilibrium Line Equation

An equilibrium line equation for multicomponent feed and countercurrent flow was calculated by the method of Stern et al. (1978), which also yields the total capillary length and permeation area per stage. The permeability coefficients listed above were used in these calculations.

Operating Line Equations

Two operating line equations are required: one for the enriching section of the cascade and another for its stripping section. These equations are derived as follows, with reference to Figure 1:

Enriching section: a total material balance around the top of the cascade and the $(n + 1)$th stage yields:

$$P_n = P_N + L_{n+1} \tag{1}$$

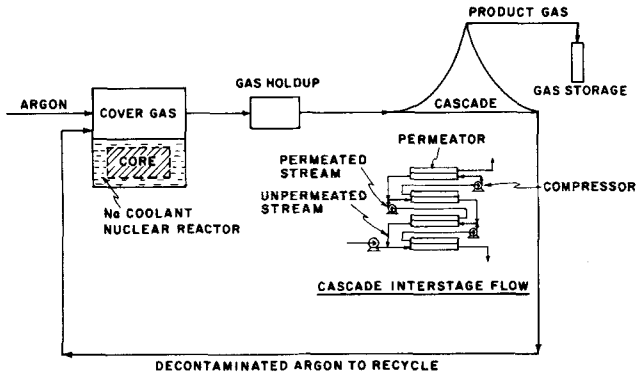

Figure 2. Process for removing Kr and Xe from the cover gas of LMFBR (after Rainey, Carter, and Blumkin, 1971).

A similar component balance for the i-th component yields:

$$y_n^i P_n = y_N^i P_N + x_{n+1}^i L_{n+1}, \tag{2}$$

where P and L are the molar flow rates of permeated (or *upflowing*) and unpermeated (or *downflowing*) streams, respectively, and y^i and x^i are the mol-fractions of the i-th component in these streams, respectively; the subscript designates a particular stage.

The operating line equation for the enriching section is obtained by rearranging Equation 2:

$$y_n^i = (L_{n+1}/P_n)x_{n+1}^i + (P_N/P_n)\, y_N^i \tag{3}$$

Taking the stage cut θ_n of the n-th stage as:

$$\theta_n = P_n/(P_n + L_n) \tag{4}$$

yields

$$L_n = (1 - \theta_n)\, P_n/\theta_n \tag{5}$$

Stripping section: a total material balance around the bottom of the cascade and the m-th stage yields:

$$L_m = L_1 + P_{m-1} \tag{6}$$

A similar component balance yields:

$$x_m^i L_m = x_1^i L_1 + y^i{}_{m-1} P_{m-1} \tag{7}$$

The operating line for the striping section is obtained by rearranging Equation 7:

$$x_m^i = (L_1/L_m)\, x_1^i + (P_{m-1}/L_m)\, y_{m-1}^i \tag{8}$$

The stage cut θ_m of the m-th stage is defined by:

$$\theta_m = P_m/(P_m + L_m) \tag{9}$$

Hence,

$$P_m = \theta_m L_m/(1 - \theta_m) \tag{10}$$

RESULTS OF CASCADE DESIGN CALCULATIONS

Removal of Kr and Xe from a LMFBR Cover Gas

This process is discussed first because it only involves the separation of a ternary mixture of xenon, krypton, and argon (Rainey, Carter, and Blumkin, 1971). The space above the sodium coolant in the reactor vessel of a liquid-metal-cooled fast breader reactor (LMFBR) will be filled with an inert cover gas, namely argon. The reactor will vent fission products into the cover gas during its operation, and these fission products must be removed continuously in order to decrease the hazard to operating personnel.

The separation of krypton and xenon in a cascade of capillary permeators is illustrated in Figure 2. The contaminated cover gas from the reactor is first retained for a short period of time in a hold-up vessel to allow the radiation of short-lived isotopes to decay to less hazardous levels and the temperature of the gas to decrease to the ambient. The cover gas is then fed to the cascade as shown. The radioactive krypton and xenon are concentrated in the enriching section of the cascade, and the product from this section is stored in pressurized cylinders until its radiation decreases to a safe level. The unpermeated product from the feed stage is recycled to the reactor. Since this product is not vented to the atmosphere, it is not necessary to reduce its krypton and xenon content to very low concentrations. Consequently, this cascade does not require a stripping section.

In accordance with a study by Rainey, Carter, and Blumkin (1971) of an 1000 MW (electrical) LMFBR, the mol-fraction concentrations of krypton and xenon in the feed stream to the cascade were assumed to be in the ranges of 10^{-5} and $10^{-5} - 10^{-3}$, respectively, the balance being argon. The feed rate was assumed to be in the range of 10^2 to 10^5 cm^3 (STP)/s. The

TABLE 1. CHARACTERISTICS OF AN IDEAL PERMEATION CASCADE FOR THE SEPARATION OF KR AND XE FROM LMFBR COVER GAS

Stage No.	Length of Capillary, m	Feed Rate, cm^3 (STP)/s	Stage Cut	Mol-Fraction ($\times 10^4$) Permeated Stream Kr	Permeated Stream Xe	Unpermeated Stream Kr	Unpermeated Stream Xe
1 (Feed Stage)	45255	714	0.45	0.47	9.44	0.25	2.04
2	30067	534	0.40	0.64	13.1	0.35	3.23
3	24477	387	0.45	0.87	17.5	0.46	3.77
4	16215	288	0.40	1.19	24.3	0.65	5.99
5	13144	208	0.45	1.62	32.5	0.85	7.03
6	8660	154	0.40	2.21	45.5	1.21	11.2
7	6962	110	0.45	3.01	61.4	1.59	13.3
8	4539	81.1	0.40	4.10	86.9	2.25	21.5
9	3590	57.1	0.45	5.60	119	2.96	26.0
10	2291	41.2	0.40	7.62	172	4.20	42.8
11	1751	28.1	0.45	10.3	242	5.53	53.4
12	1065	19.4	0.40	13.9	357	7.79	90.7
13	751	12.3	0.45	18.4	505	10.1	115
14	493	8.26	0.45	24.3	752	13.6	176
15	284	4.44	0.45	31.8	1194	18.5	292
16 (Top Stage)	119	2.22	0.45	40.0	2000	25.1	535
Total	159662	1529					

This cascade has only an enriching section.
Feed composition (mol-fraction): Kr, 3.50×10^{-5}; Xe, 7.11×10^{-4}; Ar, balance.
Enriched (top) product composition (mol-fraction): Kr, 0.004; Xe, 0.200; Ar, balance.
The slanted lines connect compositions of interstage streams mixing in the cascade.

TABLE 2. CHARACTERISTICS OF PERMEATION CASCADES WITH FIXED STAGE CUTS FOR THE SEPARATION OF KR AND XE FROM LMFBR COVER GAS

Stage Cut	Length of Capillary, m	Number of Stages	Cover Gas (Feed) Mol-Fraction × 10^5		Cascade Feed Rate	Compressor Capacity	Departure from Ideal Cascade,[a] %	
			Kr	Xe	cm^3 (STP)/s		Top Stage	Bottom Stage
0.25	6.24×10^6	11	4.005	2.465	8.86×10^4	1.33×10^5	−23	−40
0.30	8.83×10^5	10	4.146	8.052	8.37×10^3	1.46×10^4	−19	−40
0.35	3.20×10^5	11	3.410	20.85	1.96×10^3	4.24×10^3	−15	−36
0.38	2.02×10^5	12	3.395	37.61	9.18×10^2	2.35×10^3	−12	−27
0.39	2.28×10^5	13	2.747	35.56	9.28×10^2	2.55×10^3	−10.8	−22.9
0.40	1.60×10^5	13	3.605	54.47	5.82×10^2	1.72×10^3	−9.6	−17.1
0.41	1.66×10^5	14	3.344	57.33	5.33×10^2	1.71×10^3	−8.5	−10.5
0.42	1.63×10^5	15	3.381	64.72	4.57×10^2	1.61×10^3	−7.2	−2.7
0.43	1.51×10^5	16	3.714	78.11	3.67×10^2	1.44×10^3	−5.9	+6.0
0.44	1.73×10^5	18	3.450	78.59	3.55×10^2	1.58×10^3	−4.6	+15.9
0.45	1.76×10^5	20	3.717	90.60	3.00×10^2	1.55×10^3	−3.3	+26.6
0.50		∞						
For Ideal Cascade: 0.40~0.45	1.60×10^5	16	3.495	71.11	3.94×10^2	1.53×10^3	±3.0	

[a] The negative sign indicates that the mol-fraction of krypton in the permeated stream is less than that in the unpermeated stream when the two streams are mixed; the positive sign indicates the opposite.

These cascades have only an enriching section.
Enriched (top) product composition (mol-fraction): Kr, 0.004; Xe, 0.200; Ar, balance.
Product flow rate: 1 cm^3 (STP)/s.

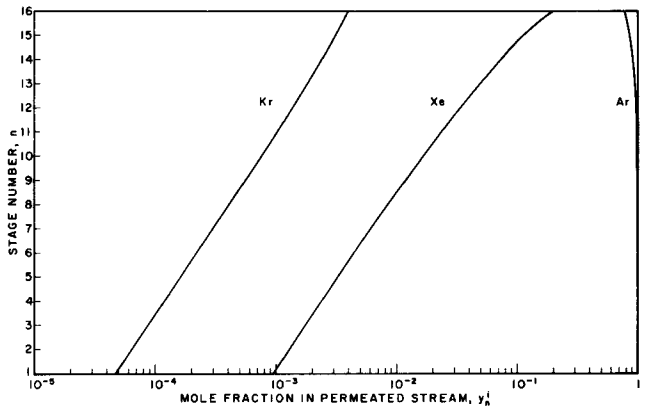

Figure 3. Cascade for removal of Kr and Xe from the cover gas of a LMFBR: composition profile of permeated streams.

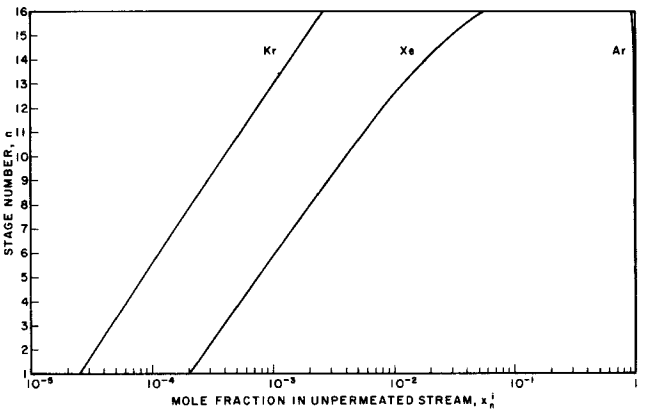

Figure 4. Cascade for removal of Kr and Xe from the cover gas of a LMFBR: composition profile of unpermeated streams.

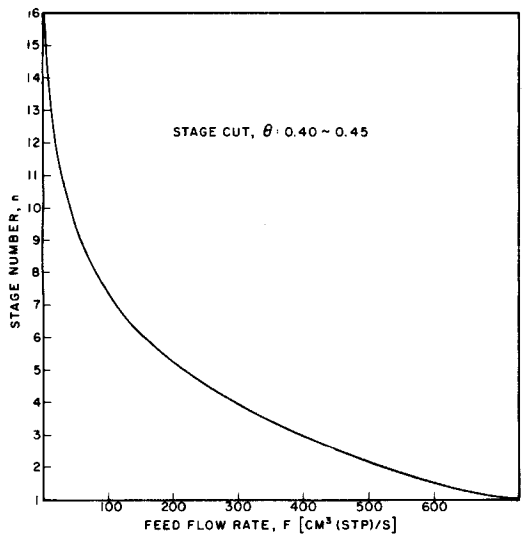

Figure 5. Cascade for removal of Kr and Xe from the cover gas of a LMFBR: stage number vs. feed flow rate.

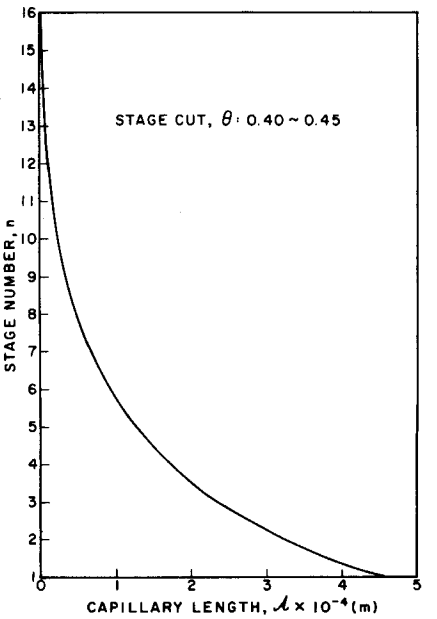

Figure 6. Cascade for removal of Kr and Xe from the cover gas of a LMFBR: stage number vs. capillary length.

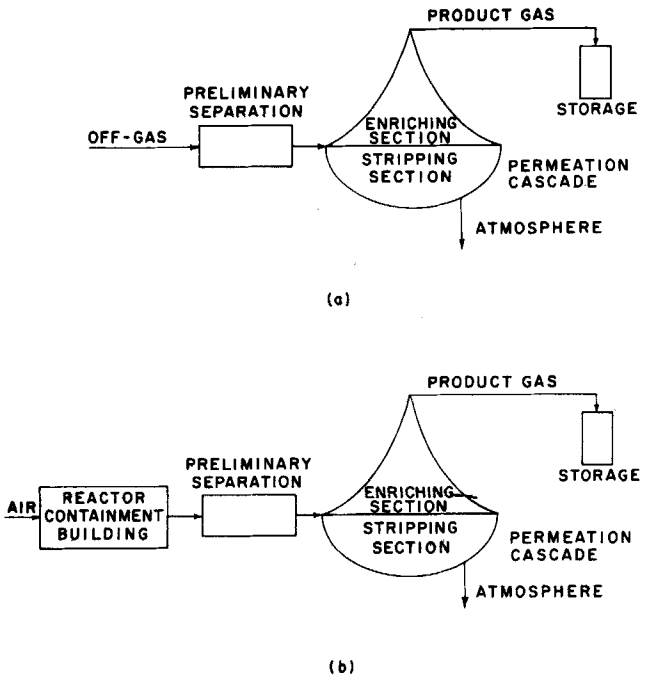

Figure 7. (a) Process for removing Kr and Xe from the off-gas of a nuclear fuel reprocessing plant. (b) Process for removing Kr and Xe from a reactor containment building after a nuclear accident.

enriching section of the cascade was designed to increase the concentration of krypton (the key component) by a factor of 100 above that in the feed stream.

To keep the whole system operating at steady state, it will be necessary to inject argon continuously into the cover gas at the same rate as that at which the concentrated product is removed from the enriching section of the cascade. Additionally, the rate of removal of radioactive krypton and xenon from the cascade must be equal to the rate at which these fission products are vented into the cover gas.

Two different types of cascades were considered in this study. One type was an *ideal* cascade, as defined earlier, in which the stage cut was varied from stage to stage so as to match the krypton concentrations in mixing streams. The other type was a nonideal cascade in which the stage cut was maintained constant. Several nonideal cascades with stage cuts from 0.25 to 0.50 were studied. Table 1 lists the characteristics of the ideal cascade. Table 2 summarizes some characteristics of the nonideal cascades and shows the extent of their departure from *ideality*. The results of the ideal cascade calculations are also shown graphically in Figures 3 to 6.

In Table 2. the first column lists the constant stage cut used in the calculations of the nonideal cascades as the basic design parameter. The second column shows the total capillary length required in each cascade. The third column lists the number of stages needed to obtain the desired krypton and xenon concentration in the product, at the specified stage cut. The fourth and fifth columns show the corresponding concentrations (in mol fractions) of krypton and xenon in the cover gas; an upper permissible value of 4×10^{-5} is specified for krypton.

To compare the performance of cascades with different stage cuts, the flow rate of the product from the enriching section was fixed at 1 cm^3 (STP)/s and the mol-fractions of krypton and xenon were specified to be 0.004 and 0.2, respectively. The sixth column shows the feed rate to the cascades. The seventh column gives the required compressor capacity, which is equivalent to the total flow rate of the low-pressure (permeated) interstage streams in the cascade. These streams must be compressed from 1.01×10^5 N/m^2 (14.7 lb/in.^{2} abs) to 3.77×10^5 N/m^2 (54.7 lb/in.^{2} abs) to maintain a Δp of 2.76×10^5 N/m^2 (40 lb/in.^{2}) in each stage. The last two columns of Table 2 show the extent to which the nonideal cascades depart from the ideal cascade criterion.

It is important to note in Table 2 that the criterion used for the design of the ideal cascade, that is, the matching of krypton concentration in mixing interstage streams (to within ±3%), indeed minimizes the total cascade volume and the power requirement. It should also be noted that, at least for low concentrations of krypton and xenon, the ideal cascade can be approximated by using a suitable constant stage cut, instead of varying the stage cut from stage to stage.

Table 2 shows that a stage cut of 0.43 provides a good approximation to an ideal cascade. This is evident from the data in the last two columns of the table, which show that the nonideal cascade with a constant stage cut of 0.43 deviates least from the ideal cascade. Therefore, a constant stage cut was used in the design of the cascades described in the following sections to simplify calculations. Various stage cuts were tried to minimize cascade volume and power requirements. This simplified procedure is useful particularly for the design of cascades with both enriching and stripping sections, in which the material balance as well as the match of krypton concentrations in mixing streams must be satisfied around the feed stage.

Removal of Kr and Xe from the Off-Gas of a Fuel Reprocessing Plant

The permeation cascade considered in this case has both an enriching and a stripping section, for the following reasons (Rainey, Carter, and Blumkin, 1971). In the aqueous processing of spent reactor fuels, the fuel is usually dissolved in nitric acid. The dissolver off-gas contains water vapor, oxides of nitrogen and other chemically active gases, oxygen, nitrogen, as well as krypton and xenon. The last two gases are mainly fission products and are therefore radioactive. The water vapor, nitrogen oxides, and chemically active gases can be separated from the off-gas by conventional techniques. The krypton and xenon may then be removed in a permeation cascade, Figure 7(a). This treatment will reduce the concentrations of the two helium-group gases in the product from the stripping section of the cascade to the extent where this product can be discharged directly to the atmosphere. The product from the enriching section, concentrated in krypton and xenon, must be collected and stored in gas cylinders under pressure until its radiation decays to a safe level.

The calculations also require the total material balance around the cascade:

$$F = P_N + L_1 \tag{11}$$

where F is the molar flow rate of the feed, and the component balance:

$$x_F^i F = y_N^i P_N + x_1^i L_1 \tag{12}$$

The calculations for an *ideal* cascade then proceed as follows:

(1) Given the feed flow rate, F, the feed composition in terms of the mol fractions x_F^i of all components; and the mol fraction x_1^j of a key component j in the *bottom* product.

(2) Assume the *top* product flow rate P_N and the mol-fractions x_1^i of all other components in the bottom product (i.e., other than x_1^j), then use Equations 11 and 12 to calculate the bottom product flow rate L_1 and the mol-fractions y_N^i of all components of the top product stream.

(3) Assume the stage cut θ_N, calculate L_N from Equation 4, then use the *equilibrium line* equations to calculate the mol-fractions x_N^i in the unpermeated product stream from the top stage (N).

(4) Use Equation 1 to obtain P_{N-1} and operating line Equation 2 for the enriching section to calculate the mol-fractions y_{N-1}^i in the permeated stream from the $(N-1)$th stage.

(5) The general procedure for the n-th stage in the enriching section is then as follows: having obtained P_n and y_n^i for all components as in step (4) for stage $N-1$, assume the stage cut θ_n.

(6) Calculate L_n from Equation 4, then use the equilibrium line equations to determine the mol-fractions x_n^i in the unpermeated stream leaving the n-th stage.

(7) Use Equation 1 to obtain P_{n-1} and operating line Equation 2 to calculate the mol-fractions y^i_{n-1} in the permeated stream from the $(n-1)$th stage.

(8) To design an *ideal* cascade, iterate steps (5) to (7) by assuming new stage cuts θ_n until $x^j_{n+1} \cong y^j_{n-1}$ for the key component. This condition is required to match the concentrations of the key component in the interstage streams which are mixed to form the feed to stage n.

(9) Continue this iterative procedure *down* to the feed stage mn to obtain L_{mn} and x^i_{mn} for all components. Use Equation 1, with P_n replaced by $(P_{mn-1} + F_1)$ to determine P_{mn-1}; also, use Equation 2, with $y^i_n P_n$ replaced by $(y^i_{mn-1} P_{mn-1} + F x^i_F)$, to determine y^i_{mn-1} for all components.

(10) Next, assume the stage cut θ_1 in the bottom stage 1 and, having obtained L_1 in step (2), use Equation 10 to calculate the flow rate P_1 of the permeated stream from stage 1; then use the equilibrium line equations to calculate the mol-fractions y^i_1 of the components in the permeated stream.

(11) Knowing L_1 and P_1, use Equation 6 to obtain L_2; knowing also all y^i_1's, use the operating line Equation 8 for the strippping section to determine the mol-fractions x^i_2 in the downflowing stream from stage 2.

(12) The general procedure at the m-th stage in the stripping section then follows: having obtained L_m and also x^i_m for all components, as in step (11) for stage $m-1$, assume stage cut θ_m.

(13) Calculate P_m from Equation 10; next, use the equilibrium line equations to determine the mol-fractions y^i_m in the upflowing stream from stage m.

(14) Use Equation 6 to obtain L_{m+1} and the operating line Equation 8 to calculate the mol-fractions x^i_{m+1} in the unpermeated stream leaving stage $m+1$.

(15) Iterate steps (12) to (14) by assuming new stage cuts θ_m until the ideal cascade condition $x^j_{m+1} \cong y^j_{m-1}$ is fulfilled for the key component.

(16) Continue the iterative procedure *up* to one stage below the feed stage, where L_{mn}, x^i_{mn}, P_{mn-1}, and y^i_{mn-1} are known.

(17) Adjust the values of P_N; x^i_1 for all components except the key component x^j_1 (which is specified); θ_N; and θ_1; until the values of y^i_{mn-1} and P_{mn-1} obtained both by steps (9) and (16), that is, by iterative calculations in the enriching and strippings sections, respectively, meet the selected convergence criterion. If this requirement is satisfied, then one obtains $x^j_F \cong x^j_{mn-1} \cong y^j_{mn-1}$ for the key component at the feed stage.

It should be noted that the concentrations of the key component in interstage streams which are mixed cannot be matched exactly without forcing other cascade variables, such as stage cuts, to assume impractical values. In this study, these concentrations were matched to ±3%. Nonideal cascades with constant stage cuts in the enriching and stripping sections can be designed by the same procedure but omitting steps (8) and (15). In the special case of a cascade which does not require a stripping section, the design procedure can be simplified by the trial-and-error selection of the top product composition (y^i_N) followed by stage-to-stage calculations until the material balance at the feed stage is satisfied.

TABLE 3. CHARACTERISTICS OF A PERMEATION CASCADE FOR THE SEPARATION OF KR AND XE FROM THE OFF-GAS OF A FUEL REPROCESSING PLANT

Stage No.	Length of Capillary, m	Feed Rate, cm³(STP)/s	Mol-fraction in Permeated Stream			Mol-fraction in Unpermeated Stream		
			Kr	Xe	O_2	Kr	Xe	O_2
1 (Bottom Stage)	15529	161	2.94×10^{-8}	3.01×10^{-11}	0.346	9.35×10^{-9}	4.28×10^{-12}	0.203
2	24024	262	5.11×10^{-8}	9.49×10^{-11}	0.419	1.71×10^{-8}	1.42×10^{-11}	0.258
3	28446	325	8.70×10^{-8}	3.17×10^{-10}	0.486	3.04×10^{-8}	5.00×10^{-11}	0.312
4	30653	364	1.45×10^{-7}	1.09×10^{-9}	0.540	5.27×10^{-8}	1.79×10^{-10}	0.360
5	31700	390	2.37×10^{-7}	3.75×10^{-9}	0.582	8.88×10^{-8}	6.38×10^{-10}	0.400
6	32183	405	3.82×10^{-7}	1.30×10^{-8}	0.612	1.46×10^{-7}	2.26×10^{-9}	0.431
7	32399	414	6.09×10^{-7}	4.47×10^{-8}	0.633	2.37×10^{-7}	7.93×10^{-9}	0.453
8	32493	420	9.62×10^{-7}	1.53×10^{-7}	0.647	3.79×10^{-7}	2.76×10^{-8}	0.468
9	32533	424	1.51×10^{-6}	5.26×10^{-7}	0.657	6.01×10^{-7}	9.52×10^{-8}	0.478
10	32549	426	2.37×10^{-6}	1.80×10^{-6}	0.663	9.46×10^{-7}	3.27×10^{-7}	0.485
11	32555	428	3.71×10^{-6}	6.15×10^{-6}	0.667	1.48×10^{-6}	1.12×10^{-6}	0.490
12	32556	429	5.78×10^{-6}	2.10×10^{-5}	0.669	2.32×10^{-6}	3.84×10^{-6}	0.493
13	32555	429	9.01×10^{-6}	7.16×10^{-5}	0.671	3.62×10^{-6}	1.31×10^{-5}	0.494
14 (Feed Stage)	38212	460	1.47×10^{-5}	2.80×10^{-4}	0.679	5.64×10^{-6}	4.48×10^{-5}	0.497
15	24800	338	2.19×10^{-5}	3.93×10^{-4}	0.817	9.55×10^{-6}	7.26×10^{-5}	0.677
16	16681	248	3.14×10^{-5}	5.46×10^{-4}	0.904	1.49×10^{-5}	1.11×10^{-4}	0.816
17	11599	182	4.39×10^{-5}	7.54×10^{-4}	0.952	2.20×10^{-5}	1.62×10^{-4}	0.904
18	8233	132	6.06×10^{-5}	1.04×10^{-3}	0.976	3.12×10^{-5}	2.30×10^{-4}	0.952
19	5893	96.1	8.30×10^{-5}	1.44×10^{-3}	0.987	4.33×10^{-5}	3.23×10^{-4}	0.976
20	4218	69.3	1.13×10^{-4}	2.00×10^{-3}	0.993	5.96×10^{-5}	4.53×10^{-4}	0.988
21	2998	49.4	1.55×10^{-4}	2.80×10^{-3}	0.995	8.17×10^{-5}	6.37×10^{-4}	0.994
22	2104	34.8	2.11×10^{-4}	3.98×10^{-3}	0.996	1.12×10^{-4}	9.09×10^{-4}	0.996
23	1445	23.9	2.89×10^{-4}	5.79×10^{-3}	0.993	1.53×10^{-4}	1.32×10^{-3}	0.997
24	959	15.9	3.96×10^{-4}	8.66×10^{-3}	0.991	2.10×10^{-4}	1.99×10^{-3}	0.997
25	600	10.0	5.41×10^{-4}	1.35×10^{-2}	0.986	2.89×10^{-4}	3.12×10^{-3}	0.996
26	335	5.63	7.39×10^{-4}	2.25×10^{-2}	0.977	3.97×10^{-4}	5.23×10^{-3}	0.994
27 (Top Stage)	140	2.39	1.00×10^{-3}	4.00×10^{-2}	0.959	5.46×10^{-4}	9.48×10^{-3}	0.990
Total	508392	6544						

This cascade has both an enriching section (14-27) and a stripping section (1-13).
Feed stage: 14. Stage cuts: enriching section = 0.425; stripping section = 0.385.
Feed composition (mol-fraction): Kr, 1.02×10^{-5}; Xe, 4.07×10^{-4}; O_2, 0.210; N_2, balance.
Feed rate: 100 cm³ (STP)/s.
Enriched (top) product composition (mol-fraction): Kr, 1.00×10^{-3}; Xe, 4.00×10^{-2}; O_2 0.959; N_2, balance.
Enrichment rate: 1.016 cm³ (STP)/s.
Depleted (bottom) product composition (mol-fraction): Kr, 9.35×10^{-9}; Xe, 4.28×10^{-12}; O_2, 0.203; N_2, balance.
Depletion rate: 98.984 cm³ (STP)/s
The slanted lines connect compositions of interstage streams mixing in the cascade.

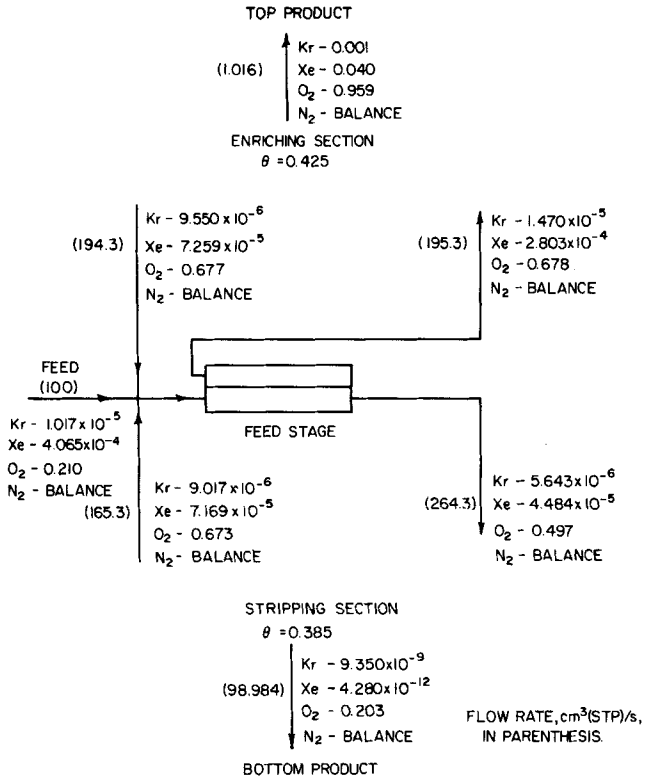

Figure 8. Cascade for removal of Kr and Xe from the off-gas of a nuclear fuel reprocessing plant: diagram of material balances.

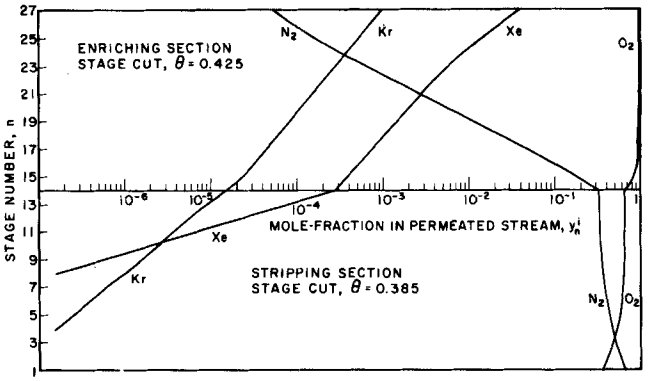

Figure 9. Cascade for removal of Kr and Xe from the off-gas of a nuclear fuel reprocessing plant: composition profile of permeated streams.

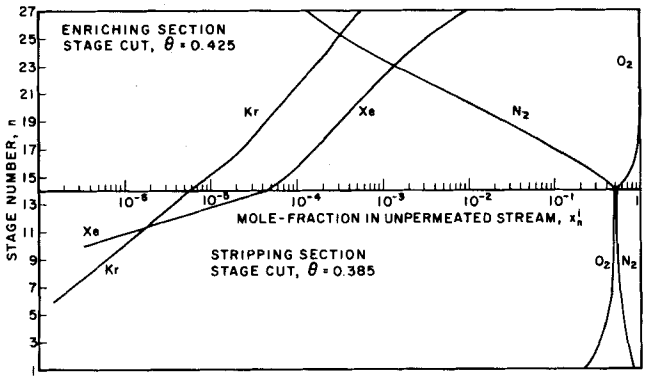

Figure 10. Cascade for removal of Kr and Xe from the off-gas of a nuclear fuel reprocessing plant: concentration profile of unpermeated streams.

In accordance with the simplified design procedure described in the previous section, constant stage cuts were used in both enriching and stripping sections. These stage cuts had to be found by trial-and-error so that the characteristics of the cascade would approximate those of an ideal cascade.

The off-gas to be processed in the permeation cascade was assumed to be a quaternary mixture of krypton, xenon, oxygen, and nitrogen. The concentrations (in mol-fractions) of krypton and xenon were taken to be of the orders of magnitude of 10^{-5} and 10^{-4}, respectively. The concentrations of oxygen and nitrogen were selected to be approximately the same as in air, i.e., 0.21 for oxygen and the balance for nitrogen. The permeation cascade was required to decrease the concentration of the key component, krypton, by a factor of 1000, so that the concentration of this component in the product gas from the stripping section would not exceed about 1×10^{-8} (mol fraction). The concentration of xenon would be reduced at the same time to a large extent because the separation factors $\alpha(\text{Xe-O}_2)$ and $\alpha(\text{Xe-N}_2)$ are substantially larger than $\alpha(\text{Kr-O}_2)$ and $\alpha(\text{Kr-N}_2)$. The separation factors are the ratios of the effective permeability coefficients.

The product from the stripping section could then be vented directly to the atmosphere. Note that the natural abundance of krypton and xenon in air is (in mol-fractions) 1.14×10^{-6} and 8.60×10^{-8}, respectively. Consequently, the concentrations of krypton and xenon in the product from the stripping section are considerably lower than in air. The cascade feed rate was assumed to be 100 cm^3 (STP)/s, while the flow rate of the product stream from the enriching section, concentrated in krypton and xenon, was taken as 1% of the feed.

It was mentioned earlier that, in the calculation of cascades, the concentration of the key component, krypton, is specified in the product from the bottom stage (stripping section), while the concentrations of the other components in that product are adjusted by trial-and-error together with the stage cuts so as to obtain an approximation of an ideal cascade.

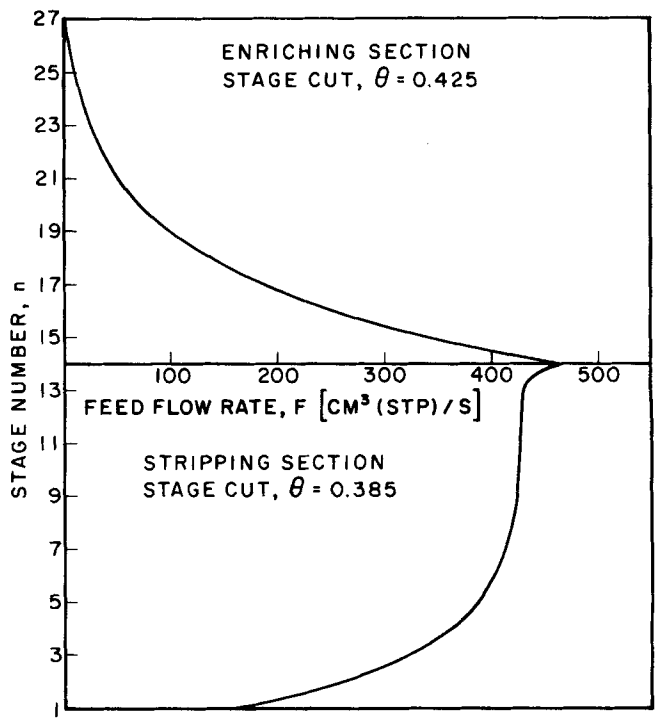

Figure 11. Cascade for removal of Kr and Xe from the off-gas of a nuclear fuel reprocessing plant: stage number vs. feed flow rate.

In the present case, the concentrations of xenon, oxygen, and nitrogen are adjustable. However, the concentration of xenon in the bottom stage is not critical becasue it is so low that this gas has a negligible effect on the design of the cascade. Of the concentrations of the other two components, oxygen and nitrogen, only one needs to be adjusted in the trial-and-error calculations, since the other one will form the balance of the product. In the present study, the concentration of oxygen was taken as the single independent variable.

The results of the calculations are summarized in Table 3 and are shown graphically in Figures 8 to 12. Note that the bottom product has almost the same oxygen and nitrogen concentration as the feed, due to the fact that 99% of the feed is processed in the stripping section. The top product stream, whose flow rate is 1% of that of the feed, is highly concentrated in oxygen in addition to krypton and xenon. The large difference between the oxygen and nitrogen concentration in the feed and that in the permeated and unpermeated streams from the feed stage can be better understood from Figure 7, which shows the flow rates and compositions of the streams around the feed stage. This difference is dictated by the choice of krypton as the key component and the requirement to match krypton concentrations in mixing interstage streams. The oxygen and nitrogen distribution in the cascade would have been very different if the cascade had been designed primarily for the separation of oxygen, not that of krypton. Calculations show that in such a case the concentration of krypton in the top product stream would be much lower than in that of Table 3.

Removal of Kr and Xe from a Reactor Containment Building

The last process investigated was the removal of radioactive krypton and xenon from a reactor containment building after a nuclear accident, such as may be caused by loss of coolant in a water-cooled reactor. The separation process is depicted in Figure 7(*b*). The contaminated air in the reactor containment building is pumped out and gaseous fission products other than krypton and xenon are first removed by conventional techniques. The two helium-group gases are then separated in the same permeation cascade designed for the treatment of the off-gas from a nuclear fuel reprocessing plant (see previous section).

In the present case, the contaminated air being pumped out of the containment building must be replaced continuously by fresh air. Therefore, the concentration of krypton and xenon in the building, and thus also in the cascade feed, will decrease

Table 4. Characteristics of a Permeation Cascade for the Separation of Kr and Xe from Air

Stage No.	Length of Capillary, m	Feed Rate, cm^3(STP)/s	Mol-fraction in Permeated Stream			Mol-fraction in Unpermeated Stream		
			Kr	Xe	O_2	Kr	Xe	O_2
1 (Bottom Stage)	15529	161	3.25×10^{-9}	6.41×10^{-15}	0.346	1.04×10^{-9}	9.11×10^{-16}	0.203
2	24024	262	5.66×10^{-9}	2.02×10^{-14}	0.419	1.89×10^{-9}	3.03×10^{-15}	0.258
3	28446	325	9.63×10^{-9}	6.74×10^{-14}	0.485	3.36×10^{-9}	1.06×10^{-14}	0.312
4	30641	364	1.61×10^{-8}	2.31×10^{-13}	0.540	5.83×10^{-9}	3.80×10^{-14}	0.360
5	31684	390	2.63×10^{-8}	7.98×10^{-13}	0.582	9.83×10^{-9}	1.36×10^{-13}	0.400
6	32164	405	4.23×10^{-8}	2.76×10^{-12}	0.612	1.62×10^{-8}	4.81×10^{-13}	0.431
7	32378	414	6.74×10^{-8}	9.50×10^{-12}	0.633	2.63×10^{-8}	1.69×10^{-12}	0.453
8	32490	420	1.06×10^{-7}	3.26×10^{-11}	0.647	4.20×10^{-8}	5.86×10^{-12}	0.468
9	32531	424	1.67×10^{-7}	1.12×10^{-10}	0.657	6.65×10^{-8}	2.02×10^{-11}	0.479
10	32548	426	2.62×10^{-7}	3.83×10^{-10}	0.663	1.05×10^{-7}	6.96×10^{-11}	0.485
11	32554	428	4.10×10^{-7}	1.31×10^{-9}	0.667	1.64×10^{-7}	2.39×10^{-10}	0.490
12	32556	429	6.40×10^{-7}	4.46×10^{-9}	0.669	2.57×10^{-7}	8.16×10^{-10}	0.493
13	32557	429	9.98×10^{-7}	1.52×10^{-8}	0.671	4.00×10^{-7}	2.79×10^{-9}	0.494
14 (Feed Stage)	38411	460	1.64×10^{-6}	5.99×10^{-8}	0.673	6.25×10^{-7}	9.52×10^{-9}	0.490
15	24905	338	2.44×10^{-6}	8.41×10^{-8}	0.814	1.06×10^{-6}	1.54×10^{-8}	0.671
16	16730	248	3.50×10^{-6}	1.17×10^{-7}	0.902	1.66×10^{-6}	2.37×10^{-8}	0.812
17	11622	182	4.89×10^{-6}	1.61×10^{-7}	0.951	2.44×10^{-6}	3.46×10^{-8}	0.901
18	8245	132	6.73×10^{-6}	2.22×10^{-7}	0.976	3.46×10^{-6}	4.92×10^{-8}	0.951
19	5902	96.1	9.21×10^{-6}	3.07×10^{-7}	0.989	4.80×10^{-6}	6.89×10^{-8}	0.976
20	4225	69.3	1.26×10^{-5}	4.26×10^{-7}	0.994	6.59×10^{-6}	9.65×10^{-8}	0.988
21	3005	49.4	1.71×10^{-5}	5.98×10^{-7}	0.997	9.01×10^{-6}	1.36×10^{-7}	0.994
22	2111	34.8	2.33×10^{-5}	8.50×10^{-7}	0.998	1.23×10^{-5}	1.93×10^{-7}	0.997
23	1452	23.9	3.18×10^{-5}	1.23×10^{-6}	0.999	1.68×10^{-5}	2.80×10^{-7}	0.998
24	965	15.9	4.35×10^{-5}	1.84×10^{-6}	1.000	2.30×10^{-5}	4.19×10^{-7}	0.999
25	606	10.0	5.95×10^{-5}	2.87×10^{-6}	1.000	3.14×10^{-5}	6.54×10^{-7}	1.000
26	341	5.63	8.16×10^{-5}	4.76×10^{-6}	1.000	4.31×10^{-5}	1.08×10^{-6}	1.000
27 (Top Stage)	145	2.39	1.12×10^{-4}	8.56×10^{-6}	1.000	5.91×10^{-5}	1.95×10^{-6}	1.000
Total	508767	6544						

This cascade has both an enriching section (14-27) and a stripping section (1-13).
Feed stage: 14. Stage cuts: enriching section = 0.425; stripping section = 0.385.
Feed composition (mol-fraction): Kr, 1.14×10^{-6}; Xe, 8.60×10^{-8}; O_2, 0.210; N_2, balance.
Feed rate: 100 cm^3 (STP)/s.
Enriched (top) product composition (mol-fraction): Kr, 1.12×10^{-4}; Xe, 8.56×10^{-6}; O_2, 1.000; N_2, 0.000.
Enrichment rate: 1.016 cm^3 (STP)/s.
Depleted (bottom) product composition (mol-fraction): Kr, 1.04×10^{-9}; Xe, 9.11×10^{-16}; O_2, 0.203; N_2, balance.
Depletion rate: 98.984 cm^3 (STP)/s.
The slanted lines connect compositions of interstage streams mixing in the cascade.

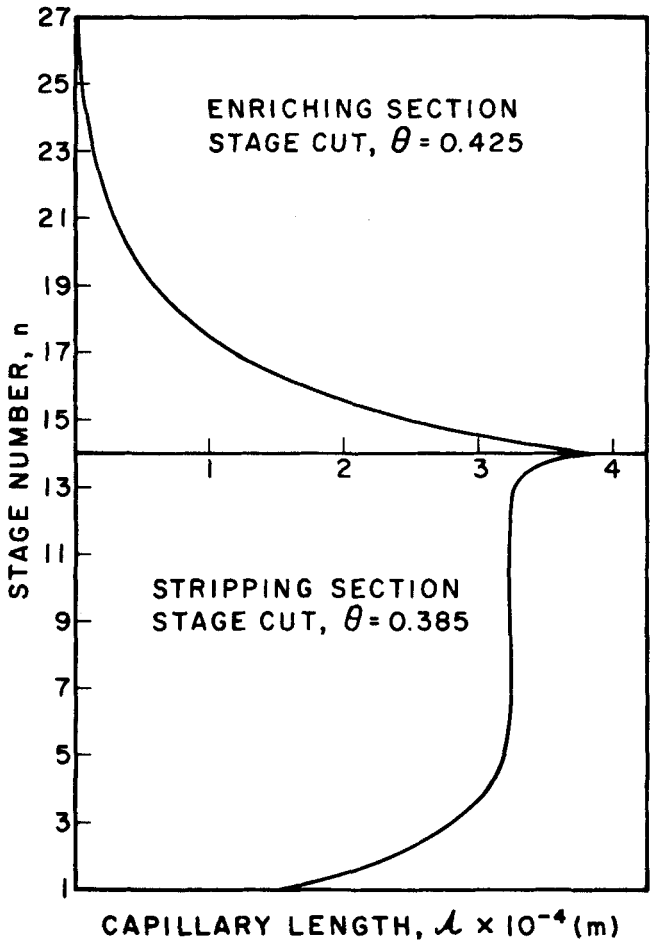

Figure 12. Cascade for removal of Kr and Xe from the off-gas of a nuclear fuel reprocessing plant: stage number vs. capillary length.

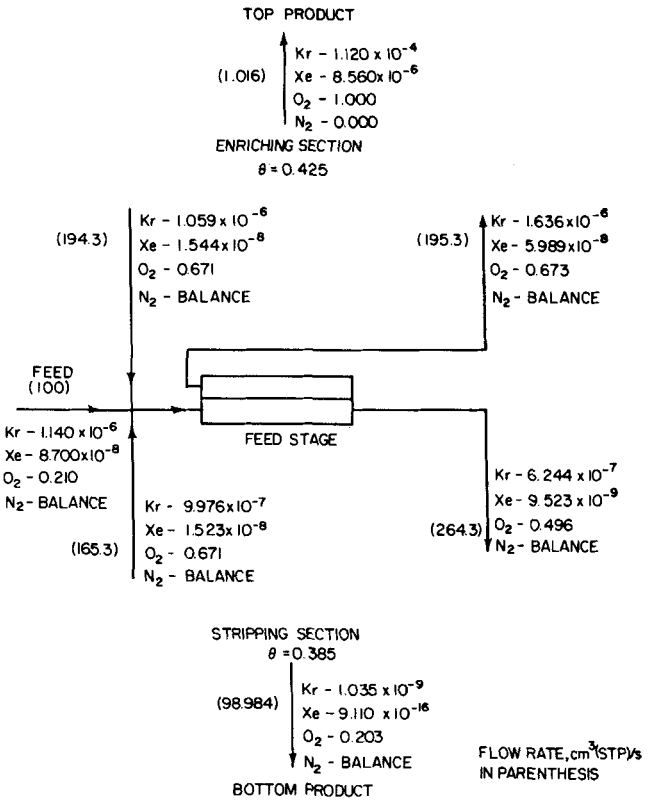

Figure 13. Cascade for removal of Kr and Xe from air: diagram of material balances.

continuously until it becomes equal to the natural abundance of the two gases in air. As a result, the cascade will operate under unsteady-state conditions until the natural abundance of krypton and xenon is attained in the feed. To simplify cascade design under unsteady-state conditions, it was assumed that the capacity of the permeation cascade is small compared to the volume of the reactor containment building. The cascade can then be taken to operate at steady-state over short periods of time.

Calculations were made for two extreme conditions: the initial and final compositions of the feed stream. The initial composition was assumed to be the same as the composition of the feed discussed in the previous section: that is, in mol-fractions, Kr, 10^{-5}; Xe, 10^{-4}; O_2, 0.21; and N_2, balance. The final composition was taken to be that of air: Kr, 1.14×10^{-6}; Xe, 8.60×10^{-8}; O_2, 0.21; and N_2, balance. In the first case the cascade would operate under quasisteady-state conditions, while in the second case it would have reached a true steady state.

Due to the above assumptions, the cascade characteristics for the initial feed composition are the same as those in Table 3 for the separation of krypton and xenon from the off-gas of a fuel reprocessing plant. The cascade characteristics for the final feed composition are summarized in Table 4. For intermediate feed compositions, in-between the initial and final ones, the performance of the cascade can be determined as follows.

An examination of Tables 3 and 4, or of Figures 8 and 13, shows that the mol-fraction concentrations of krypton in the top product, feed, and bottom product streams are in the same ratios of (approximately) $1:1 \times 10^{-2}:1 \times 10^{-5}$ during the initial and final (steady state) stages of the operation. Similarly, the mol fractions of xenon in the three streams are in the same ratios of $1:1 \times 10^{-2}:1 \times 10^{-10}$ during the initial and final stages of cascade operation. If it is assumed that the above ratios remain unchanged during the entire period of cascade operation, the krypton and xenon mol-fractions in the top and bottom product streams can be determined from these ratios for any feed composition during that period. The change in the krypton and xenon concentrations in the feed with time can be calculated assuming *perfect mixing* of the contaminated and fresh air in the reactor containment building, as in a continuous-flow stirred-tank reactor.

EFFECTS OF IRRADIATION

The effects of irradiation on the permeability and physical properties of silicone rubber membranes must be considered in view of the intended applications of the permeation cascades. Robb (1965, 1967) has irradiated samples of silicone rubber containing a few tenths of a percent vinyl groups to doses of 10^7 to 10^8 rads. He found that the permeability of the samples to oxygen, nitrogen, and carbon dioxide decreased by 10 to 20%, and that the separation factors increased by 2 to 20%. The silicone rubber became fairly brittle at 10^8 rads.

Rainey, Carter, and Blumkin (1971) exposed several silicone rubber membranes sealed to a porous backing material in a ^{60}Co source calculated to yield a dose of 8.6×10^6 rads/d at the membrane surface. They found that the membrane failed after irradiation to 1.4×10^8-1.7×10^8 rads due to the formation of small holes, which caused a sudden increase in gas transport through the membranes and a decrease in separation factors. Rainey, Carter, and Blumkin concluded that the physical properties of the membranes should be satisfactory to at least 10^8 rads, provided that the membranes are not subjected to flexing from pressure surges. This exposure limit is probably also acceptable for silicon rubber capillaries.

The effects of irradiation will be most pronounced in the last few stages of the enriching section of a cascade, where the

radioactive krypton and xenon are most concentrated. The size of these stages could be slightly increased to allow for a possible decrease in the permeability of the capillaries due to radiation. Any simultaneous increase in separation factors can have only a beneficial effect on the separation processes. The reactor atmospheres of this study cannot be introduced directly into a permeation cascade, because the silicone rubber capillaries would deteriorate too rapidly. Therefore, these gases must be stored for a period of time to allow a decay in the activity of short-lived isotopes.

Rainey, Carter, and Blumkin (1971) have studied this problem in connection with permeation cascades yielding a *top* product with approximately the same composition as the product of Table 2. They estimated that the life of silicone rubber membranes in the top stage of the cascade would be of the order of several months, if the feed storage time is about 20 days. This life is increased to several years when the storage time is 30 days. The results of Rainey, Carter, and Blumkin were obtained with sheet membranes bonded to a porous backing and supported on screens, and cannot be applied directly to the silicone rubber capillaries considered in the present study. However, their results provide some guidance in this matter.

The effects of γ-ray irradiation on silicone rubber membranes were investigated most recently by Ohno and Morisue et al. (1976) and by Ohno and Kakuta et al. (1977). Their results generally agree with those of Robb (1965, 1967). Silicone rubber membrane were exposed to doses of up to 3.7×10^7 rads and dose rates of up to 2.0×10^4 rad/s. These exposures slightly increased tensile strength and decreased elongation, due to crosslinking, but the gas permeability was not appreciably affected.

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NOTATION

A = capillary length
F = flow rate of cascade feed stream
L = flow rate of unpermeated stream from a stage
P = flow rate of permeated stream from a stage
$\bar{P}^*$ = effective permeability coefficient
x = mol-fraction in unpermeated stream
y = mol-fraction in permeated stream

Greek Letters

θ = stage cut, fraction of feed permeated

Subscripts

m = m-th stage in stripping section of cascade
mn = feed stage of cascade
n = n-th stage in enriching section of cascade
N = top stage of cascade
1 = bottom stage of cascade

Superscripts

i = i-th component in mixture
j = key component in cascade design

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