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Calculation of Multicomponent Flows in the Aerodynamic Separation of Uranium Isotopes

Roger R. Eaton, Ronald L. Fox

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CALCULATION OF MULTICOMPONENT FLOWS IN THE AERODYNAMIC SEPARATION OF URANIUM ISOTOPES

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ABSTRACT

Equations governing gas flow containing uranium hexafluoride and an inert carrier have been developed for mixtures typical of those used in aerodynamic isotope separation. In this report these equations are solved for a simple separation device (two-dimensional channel with external force). The numerical solutions are compared with the results obtained from direct simulation.

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CALCULATION OF MULTICOMPONENT FLOWS IN THE AERODYNAMIC SEPARATION OF URANIUM ISOTOPES

Introduction

Methods for separating the isotopes of uranium U^{235} from U^{238} by processes other than gaseous diffusion are currently receiving increasing attention. The use of fluid dynamic processes for isotope separation has been pursued most actively by research groups outside of the United States. Fluid dynamic separations have been able to achieve enrichment factors 5 to 10 times greater than gaseous diffusion, but have not in their present state of development offered any significant reduction in separative work.

Fluid processes have been examined to determine the methods which offer the greatest promise.[1, 2, 3] This analysis was based on equilibrium between the effective fluid dynamic forces and density gradients. The effective forces which appear in the fluid dynamic relations are due to pressure gradients, external forces, temperature gradients, and viscous effects. The greatest separation was obtained by devices based on pressure gradients and, in order to obtain high separation factors, the uranium-containing gas UF_6 had to be combined with a light carrier, i.e., He or H_2. The method of generating large pressure gradients which is probably most readily available is that using streamline curvature, such as devices using curved nozzles, colliding jets, and vortex flows.

The colliding-jet devices appear to offer advantages over others by reducing viscous wall losses and thus yielding lower separative work. Experimental measurement of colliding jets have shown them to be superior to curved nozzles in certain flow regimes,[4] but the lack of stability of the jet configuration for other regimes can result in poorer ability to separate isotopes. Experiments with incompressible colliding jets has shown the flow to vary from stable to unstable, depending on the separation distance between the interacting nozzles.[5]

Isotope separation devices based on colliding jets meet the criteria for optimum separation determined in the equilibrium analysis and promise less separative work compared to other devices which meet the same criteria. The colliding jets are, therefore, considered one of the prime candidates for further investigation.

The flow field for the simple colliding jet configuration shown in Figure 1 was initially solved for a pure UF_6 flow. The solution was carried out through a modification of an existing Sandia Laboratories finite difference computer code, 2D/LOS.[6, 7, 8] This code was initially developed for calculations of the flow in line-of-sight pipes used in underground nuclear experiments. The code was

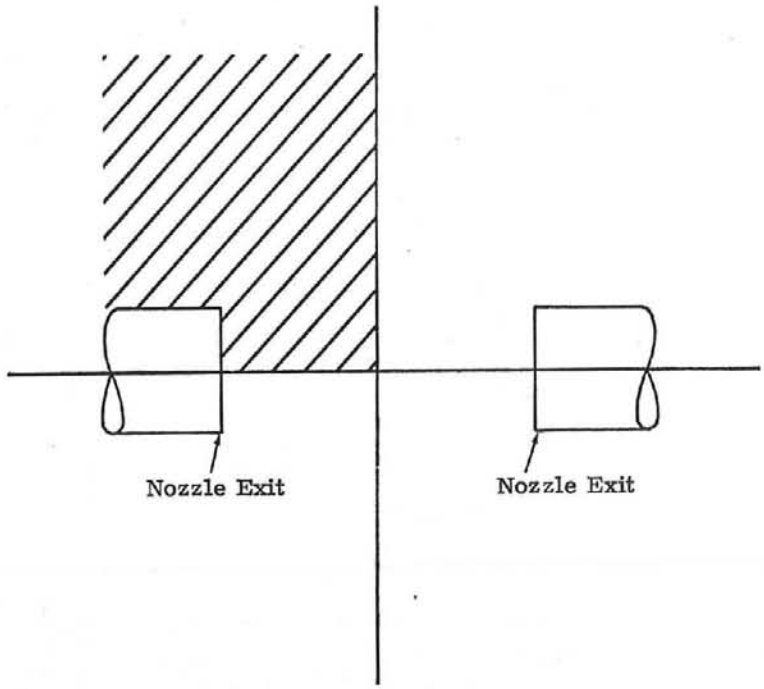

Figure 1. Opposing-Jet Geometry

written for a two-species flow in a cylindrically symmetric geometry. The modifications required for application to isotope separation were a change in geometry and addition of a momentum and energy equation for the second species in the flow.

The finite difference solution was carried out through a modified Lax differencing method. Parametric sets of solutions were carried out for various nozzle designs. The particular parameters investigated include flow speed at nozzle exit, nozzle-separation distance, Knudsen number of the flow, and the ambient- to nozzle-pressure ratio. The finite difference mesh system considered only the quadrant shown crosshatched in Figure 1, the center line of the jets and mid-plane between the jets being considered a line and surface of symmetry. This simplification due to symmetry is applicable to stable flows, but probably would not predict the development of instabilities.

The results of the pure UF_6 analysis for simple colliding jets produced regions of relatively high separation (up to 1.03) but only a low through-put of enriched fluid was obtained at the high separation factors. Flow conditions which produced more acceptable through-put (10 to 20% of the total flow through the nozzle) were found to produce separation factors of 1.01 or less. These results are in general agreement with experimental measurements.[9]

The pure UF_6 system does not meet the equilibrium criteria for optimum fluid dynamic separation; the addition of a light carrier gas is necessary. However, the carrier gas introduces problems in the fluid dynamic analysis due to the ineffectiveness of collisions between two molecules

with disparate mass in driving the specie distribution functions to equilibrium. Thus, the Navier-Stokes equations which assume small deviation from equilibrium may no longer be valid. The development of a set of flow equations which are applicable to the disparate mass flows encountered in aerodynamic separation devices will be presented in the next section.

Fluid Dynamic Analysis

Development of the Flow Equations

The flow equations for a mixture of gas molecules containing species with disparate masses has been the topic of numerous investigations.[10, 11, 12, 13] Typically the flow equations obtained by examining the Boltzmann collision integrals for particular collision models and the analysis is usually applied to a two-component mixture.

For this study the equations governing the flow were obtained from the examination of a test particle moving in a thermal bath of field particles.[14] This test particle analysis is somewhat similar to methods which have been found to be successful in developing equations for ionized gases.[15, 16] The analysis is applied to a ternary gas mixture containing 95% He, 4.96% $U^{238}F_6$, and 0.04% $U^{235}F_6$. This gas mixture was examined to see if flow fields could be obtained that would generate nonequilibrium separation of the gas species.

The individual species in the gas were examined to determine the flow regimes which should be considered in obtaining their governing equations. The ordering of interaction terms between the species was obtained from the test particle analysis. The flow equations could then be stated in terms of collision integrals.

The parameter which is most significant in determining the structure of the analysis appropriate to a particular rarefied flow is the Knudsen number. The Knudsen number is defined as the ratio of the mean free path to the characteristic dimension of the flow. Knudsen numbers of less than 0.01 indicate the flow can be considered continuum. For Knudsen numbers between 0.1 and 0.01 continuum equations with slip are applicable, and for larger Knudsen numbers the flow is either transitional or free molecular. For the purpose of examining a multicomponent mixture, a species Knudsen number for each species was defined as the ratio of the mean free path for collisions between like species to the characteristic dimension of the flow. Species Knudsen numbers less than 0.05 were considered indicative of the applicability of a multifluid analysis. The mean free path for molecular collisions of cross section σ is then

$$\lambda = 1/n\sigma \quad ,$$

where n is the number density of the molecules. For a 95/5 mixture of the He and UF_6, the relative species densities are

$$He \quad - 0.95\ N_0 \ ,$$

$$U^{238}F_6 - 0.0496\ N_0 \ , \text{ and}$$

$$U^{235}F_6 - 0.0004\ N_0 \ ,$$

where N_0 is the total number of density of the fluid.

The species mean free paths based on cross sections calculated in Reference 4 are

$$He - He \quad = 0.071 \times 10^{-16}/N_0 \ ,$$

$$U^{238}F_6 - U^{238}F_6 = 0.124 \times 10^{-16}/N_0 \ ,$$

$$U^{235}F_6 - U^{235}F_6 = 15.4 \times 10^{-16}/N_0 \ .$$

The species Knudsen number of the species with the smallest mean free path was taken as 0.01. It follows that the relative species Knudsen numbers are

$$He - He \quad = 0.01 \ ,$$

$$U^{238}F_6 - U^{238}F_6 = 0.0175 \ , \text{ and}$$

$$U^{235}F_6 - U^{235}F_6 = 2.17 \ .$$

Thus, the He and $U^{238}F_6$ species can be considered as fluids in a multifluid calculation, but the $U^{235}F_6$ must be considered separately.

The interactions between species which must be considered in a computational analysis of the flow can be determined by the number of collisions and the effectiveness of a collision in changing the species distribution function. The number of collisions per unit time per unit volume can be calculated from the cross section

$$\nu_{\alpha\beta} = n_\alpha n_\beta \overline{V}_{\alpha\beta} \sigma_{\alpha\beta} \ ,$$

where $\overline{V}_{\alpha\beta}$ is the average relative velocity between the molecules of type α and type β, and $\sigma_{\alpha\beta}$ is the cross section for collision between α and β.

The effectiveness of a collision in changing the species distribution function depends on the relative masses and velocities of the colliding partners. The relative effectiveness of collisions between the molecules in the UF_6 - He gas is calculated using a solution of the conservation equations based on point interactions.

The comparative effectiveness of collisions between the various species is evaluated by determining the number of collisions of a particular type required to effect an e-folding change in the speed of a test module entering a thermal bath. The collisional effectiveness parameter (E) is defined as the reciprocal of the number of collisions to produce an e-folding. The test molecules and the bath molecules are changed to determine the effectiveness of encounters between each possible species in the He - UF_6 gas in modifying velocity distribution functions. Relaxation times calculated using the test particle approach are in reasonable agreement with those obtained from the kinetic equation.[17] The calculated collisional effectiveness parameters of encounters for equal and unequal masses are 0.2 and 0.007, respectively.[14] The relative ordering of the interaction terms in the transport equation can then be obtained from the product of ν and the collisional effectiveness parameter. Table I presents the relative ordering coefficients obtained from this analysis.

TABLE I

Ordering Coefficients for Each Specie in the Gas

Interaction	ν (arbitrary units)	η (ordering coefficients, $\nu \times E$)
He - He	13.5	2.7
He - $U^{238}F_6$	1.16	8×10^{-3}
He - $U^{235}F_6$	9.3×10^{-3}	$6.5 \times 10^{-5*}$
$U^{238}F_6$ - He	1.16	8×10^{-3}
$U^{238}F_6$ - $U^{238}F_6$	0.0324	6.5×10^{-3}
$U^{238}F_6$ - $U^{235}F_6$	0.00026	$5 \times 10^{-5*}$
$U^{235}F_6$ - He	9.3×10^{-3}	6.5×10^{-5}
$U^{235}F_6$ - $U^{238}F_6$	2.6×10^{-4}	5.2×10^{-5}
$U^{235}F_6$ - U^{235}	2.1×10^{-6}	$4.2 \times 10^{-7*}$

*Designated interactions which are negligible.

The interaction between $U^{238}F_6$ and He is almost negligible for the He gas; however, it is of greater significance than the other neglected interaction terms, and was therefore included in the governing equations.

Examination of the $U^{238}F_6$ partial fluid shows that the auxiliary partial fluid has an influence comparable to self interactions. This could result in deviations from acceptable partial fluid behavior for $U^{238}F_6$ if there are significant differences in the thermodynamic parameters of the species. However, in the devices being considered, the flow areas in which this nonequilibrium condition could exist correspond to regions of low concentration of the heavy isotope.

The kinetic equations for each species

$$\frac{Df_i}{Dt} = -\vec{F}_i \cdot \frac{\partial f_i}{\partial \vec{v}} + \sum_{j=1}^{3} \left(\frac{\delta f}{\delta t}\right)_{ij} \quad , \quad 1 \le i \le 3 \quad ,$$

can be integrated to produce the set of conservation equations

$$\frac{D\rho_i}{Dt} = 0 \quad ,$$

$$\frac{D\rho_i \vec{u}_i}{Dt} = -\vec{F}_i + \sum_{j=1}^{3} \eta_{ij} \int \overline{\left(\frac{\delta f}{\delta t}\right)_{ij}} v_i d^3 v_i \quad ,$$

$$\frac{D\rho_i u_i^2}{Dt} = -\vec{F}_i \cdot \vec{u}_i + \sum_{j=1}^{3} \Omega_{ij} \int \overline{\left(\frac{\delta f}{\delta t}\right)_{ij}} v_i^2 d^3 v_i \quad ,$$

where $\eta(\Omega)$ is the ordering parameter for momentum (energy) transfer. Assuming that the η's and Ω's have the same ordering relationships, the data in Table I can be used to reduce the conservation equation to the form

$$U^{238}F_6 \text{ and He:} \quad \frac{D\langle\ \rangle_i}{Dt} + [\text{pure gas transport}] + \frac{\vec{F}_i}{M_i}\beta(\vec{u})$$

$$= [\text{interaction with the other specie}] \quad ,$$

$$U^{235}F_6: \quad \frac{D\langle\ \rangle}{Dt} + \frac{\vec{F}_i}{M_i}\beta(\vec{u}) = [\text{interaction with } U^{238}F_6] + [\text{interaction with He}] \quad .$$

This statement of the conservation equations makes it possible to use a two fluid solution for the heavy isotope and the light carrier gas. The light isotope flow can then be obtained from the completed solution for the U^{238} and He.

Solution of the Flow Equations for a Simple Separation Device

The requirement for the simultaneous analysis of only two of the flow components makes it possible to apply modifications to the code used for the examination of pure UF_6 to the three-component flow. The code need only be changed from $U^{238}F_6$ and $U^{235}F_6$ to $U^{238}F_6$ and He. The solution for the $U^{235}F_6$ flow can then be calculated from the steady-state solution for the outer species.

The calculation was initially carried out for a simple separation device, namely the flow in a two-dimensional channel which is subjected to a body force at right angles to the flow direction (see Figure 2). The transient equations for the heavy isotope and carrier gases were allowed to reach a steady state. The light isotope distribution was then calculated, assuming the pressure gradient produced by the major flow components to be balanced by the light isotope density gradient[18]

$$\nabla\left(\frac{n_L}{n}\right) = -\left(\frac{n_L}{n} - \frac{\rho_L}{\rho}\right)\nabla \ln P \quad ,$$

where L refers to the light isotope, the unsubscripted variables refer to the major flow components, n is the molecular density, ρ the mass density, and P the pressure.

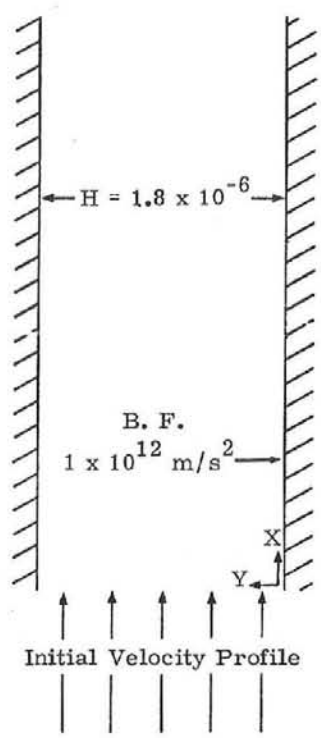

Figure 2. Two-Dimensional Flow Channel Geometry

The mesh system used was 30 x 40. However, the concentration of UF_6 along the wall required a mesh with finer detail near the wall. A step-type coordinate stretch was applied near the wall in order to obtain the desired definition in the UF_6 density; the stretch consisted of two reductions to a final mesh 25 times smaller near the wall than in the main stream (see Figure 3). The density profiles obtained from the finite difference solution for He and $U^{238}F_6$ are shown in Figures 4 and 5.

The density distribution for $U^{235}F_6$ obtained from the finite difference code using the simple diffusion approximation is shown in Figure 6. The results appear reasonable. However, quantitative data for comparison would be desirable before applying the code to more difficult flow geometries. The lack of experimental data for comparison instigated the development of Monte Carlo computer simulation methods. The availability of a Monte Carlo code, in addition to providing a test of the fluid-dynamic code for the simple two-dimensional channel, would supplement the separation program by providing solutions for flow regimes for which the fluid equations are no longer valid.

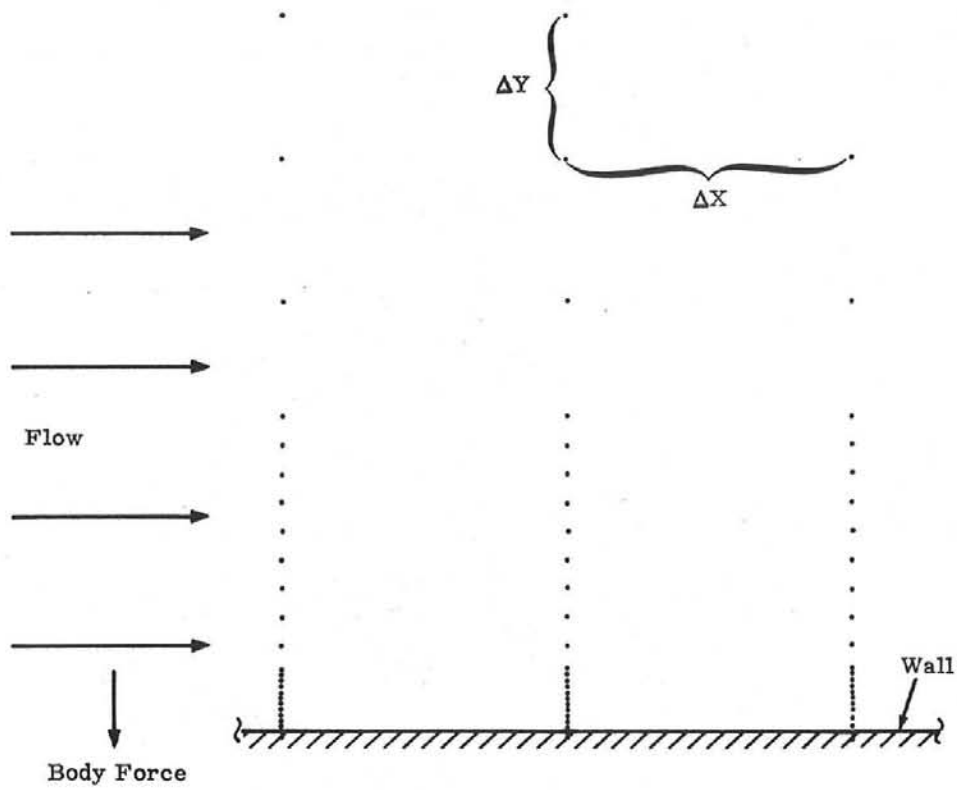

Figure 3. Finite Difference Variable Mesh System

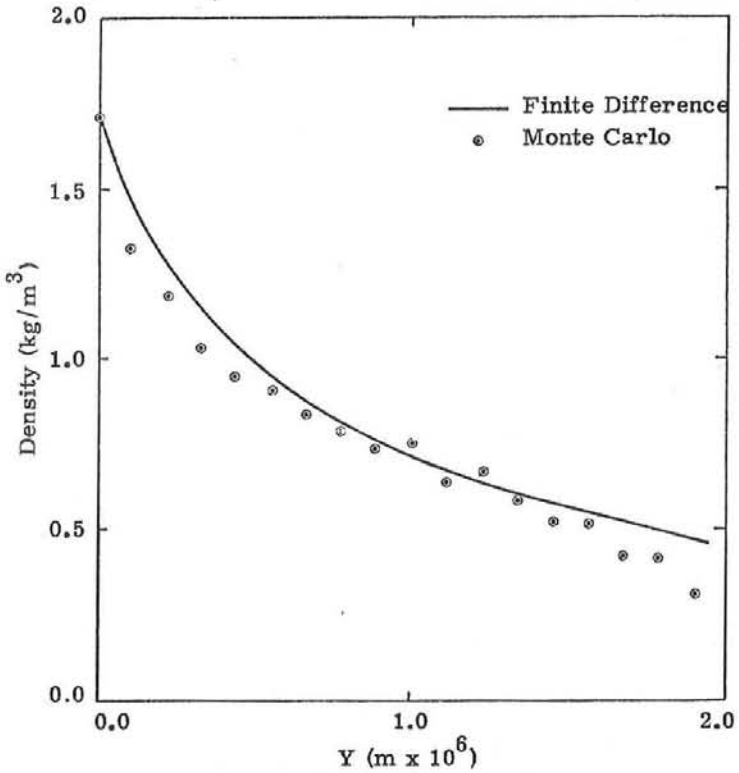

Figure 4. Helium Density Profiles Calculated by Finite Difference and Monte Carlo Methods at X/H = 1.0

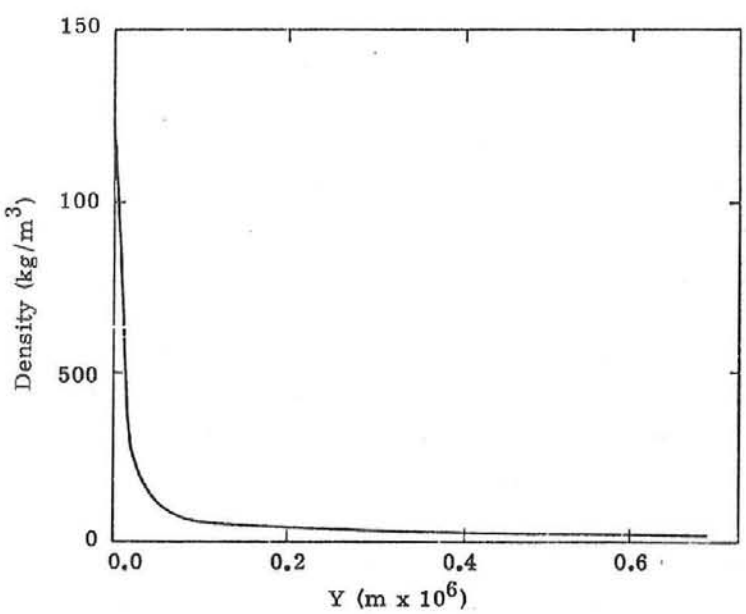

Figure 5. $U^{238}F_6$ Density Profiles Calculated by Finite Difference Method at X/H = 1.0

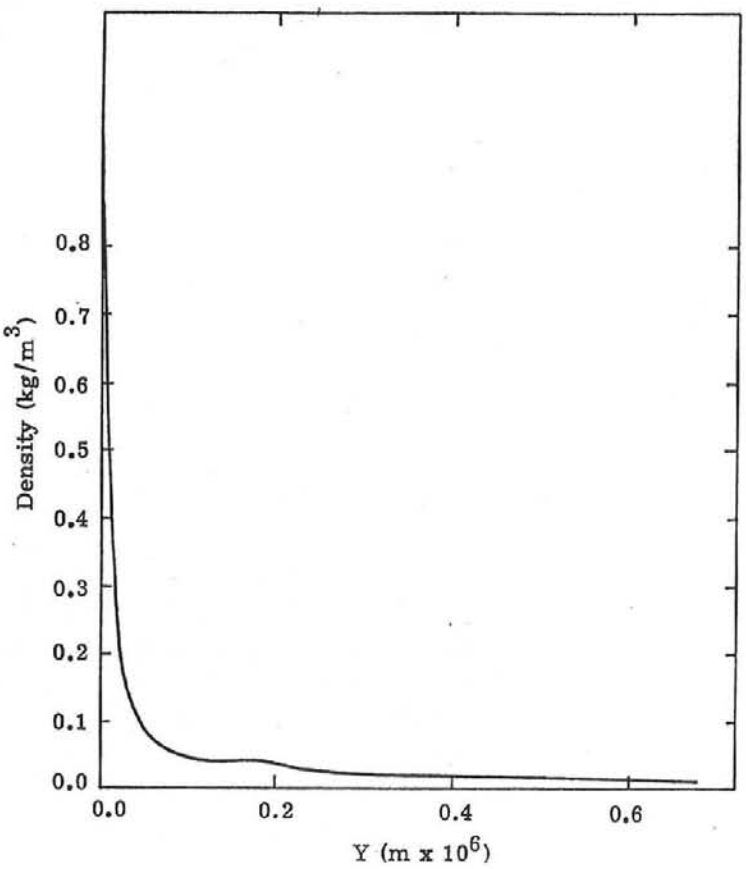

Figure 6. $U^{235}F_6$ Density Profile Calculated by Combined Finite Difference Method and Simple Diffusion Approximation at X/H = 1.0

Development of Direct Simulation Method

A Monte Carlo direct simulation technique has been developed in order to verify the finite difference equations discussed above. The Monte Carlo direct simulation method is in principle applicable to all flow regimes; however, in practice the method is useful for large Knudsen numbers only because of limited computer capacity.

The direct simulation method was developed to describe the flow of a ternary gas in a two-dimensional channel subject to a strong body force perpendicular to the flow (Figure 2 again). The magnitude of the body force is equivalent to the acceleration resulting from flow around a bend whose radius of curvature is equal to 5 μm.

The trajectories of the particles are followed using a combination of an Eulerian-Lagrangean coordinate system. This corresponds to an effective increase in the number of particles by an order of magnitude. The resulting program is applicable to lower Knudsen numbers than would be possible

with a stationary reference frame. The CDC 6600 computer is used for the calculations. Its memory size is adequate to follow 10,000 particles at any given time. With the Lagrangean system this is equivalent to 100,000 particles in a region H by H (Figure 2).

The calculations are started by placing 10,000 randomly distributed particles in the region where $0 < Y < H$ and $0 < X < \frac{H}{10}$. These particles are randomly assigned velocities which satisfy Maxwellian velocity distribution. This subregion is then observed as it moves downstream with the transport velocity. Particles are subjected to the body force, F. The particle trajectories, as described by Newton's equations, are altered by interparticle collisions and diffuse wall collisions. It has been found that the machine time required to directly simulate the interparticle collision is excessive to the point of being impractical. Consequently calculations to date have been made as follows. The region of interest is subdivided into 20 subregions. At each time step the collision frequency for each subregion is calculated based on local particle density. Interparticle collisions are then calculated by randomly choosing the proper number of pairs of particles from this subregion so the calculated collision frequency is satisfied. A typical run on the CDC 6600 takes about 500 seconds. A CDC 7600 machine is expected to be available at Sandia within 6 months. This will increase the storage capacity by a factor of two and decrease the run time required by a factor of four.

Comparison of Monte Carlo and Fluid-Dynamic Methods

The direct simulation method described above and a finite difference solution of the equations derived earlier were applied to a gas at atmospheric pressure containing 95% He and 5% UF_6 flowing between the walls shown in Figure 2. These percentages are characteristic of those encountered in current work in Germany. The results of the two methods are compared in Figures 4, 7, and 8, at a downstream distance of $X/H = 1.0$. This downstream distance is approaching the limit of the finite difference solution in its present state because of the large density gradients which are beginning to form at the lower wall due to the strong body force. A refined numerical grid would improve this situation, but only at great expense in computer time.

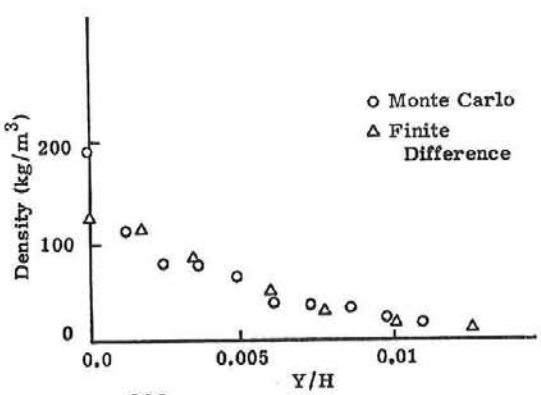

Figure 7. $U^{238}F_6$ Density Profiles Near the Wall, Calculated by Finite Difference and Monte Carlo Methods at $X/H = 1.0$

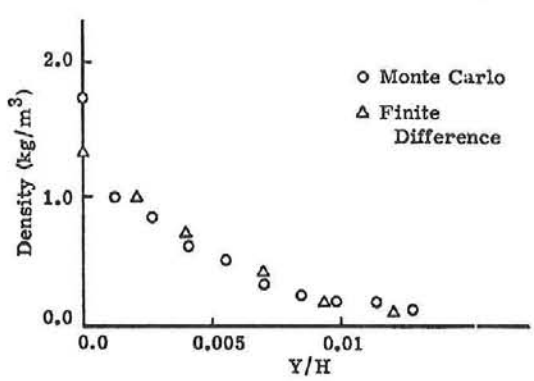

Figure 8. $U^{235}F_6$ Density Profiles Near the Wall, Calculated by Finite Difference/Diffusion and Monte Carlo Methods at $X/H = 1.0$

Figures 9 and 10 show the density profiles near the bottom wall (Y/H = 0.05) for several downstream stations. The solid lines are curves which have been fitted through the Monte Carlo calculated points. It can be seen that the Monte Carlo calculations are approaching the analytical (equilibrium) solution for large X/H distances. Separation factors* can be calculated using the fitted curves on Figures 9 and 10. The results are given in Figure 11. Even though it is realized that there would certainly be problems in actually building and experimentally analyzing a device of this size, the calculations point out the importance of analyzing the nonequilibrium aspects of these types of flow. The nonequilibrium flow both at X/H = 1.5 and X/H = 2.0 results in 50% of the flow with separation factors of 10% or more, while for equilibrium flow less than 0.1% is enriched above 5%.

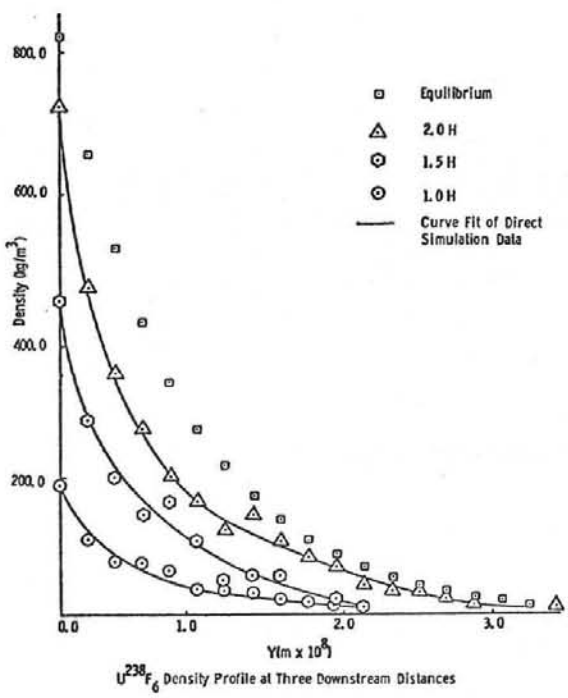

Figure 9. $U^{238}F_6$ Density Profiles Calculated Using Monte Carlo Technique at X/H = 1.0, 1.5, and 2.0

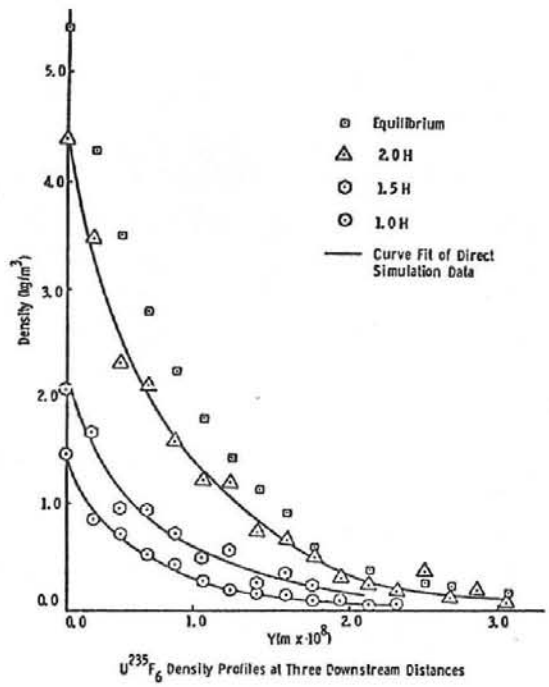

Figure 10. $U^{235}F_6$ Density Profiles Calculated Using Monte Carlo Technique at X/H = 1.0, 1.5, and 2.0

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* $\alpha = \left(\frac{a}{1-a}\right) \Big/ \left(\frac{b}{1-b}\right)$ where $a = \left.\frac{U^{235}}{U^{238}}\right|_X$ and $b = \left.\frac{U^{235}}{U^{238}}\right|_{initial}$.

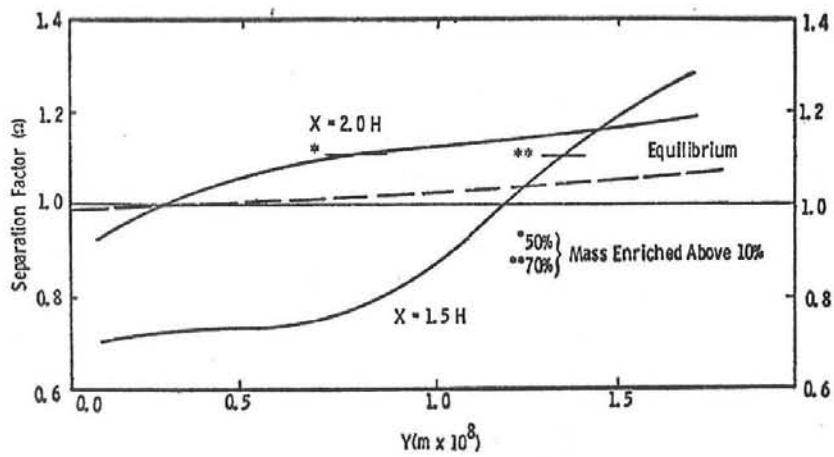

Figure 11. Isotope Enrichment Calculated Using Monte Carlo Profiles

Conclusions

A system of equations for the flow components typical of many aerodynamic isotope separators have been derived. The system of equations consists of a set of coupled flow equations for the heavy isotope of UF_6 and the inert carrier gas (He), with a set of dynamic equations for the light UF_6 isotope which can be solved after solutions to the complex equation have been obtained. The coupled equations for UF_6 heavy isotope and He were solved for a simple separator (2-μm high, 2-dimensional channel with external body force) using a modification of a finite difference code previously used for pure UF_6 flows. The UF_6 light distribution was then calculated using a simple diffusion approximation.

A Monte Carlo direct simulation calculation was developed to test the validity of the finite difference solution and to provide a calculation technique which could be used in certain flow regimes in which the finite difference code is not valid, i.e., high Knudsen number flows. The two-dimensional channel was solved using the Monte Carlo code. Comparison of the Monte Carlo and finite difference solutions are shown in Figures 4, 7, and 8.

It is apparent from these results that there are flow regimes in which both the finite difference solution and the direct simulation solution can be applied to isotope separation devices. It would be desirable to increase the physical size of the device being considered. However, with the current CDC 6600 computer limitations this doesn't appear practical except for devices which display certain properties of symmetry. Increasing the physical size of the device for the numerical calculations is restricted only by the flow detail required of the numerical grid used. This limitation is not as restrictive as is the particle limit for the Monte Carlo method for many problems. It appears from the results of the sample calculation that the equation-ordering discussed under Fluid Dynamics Analysis is applicable to the flow regimes encountered in many aerodynamic separation devices.

Future Work

The finite difference code will be used to obtain solutions for colliding jets with a ternary mixture. The same single-quadrant calculation used in the pure UF_6 jets will be used initially to examine the degree of improvement when an inert carrier is incorporated into the flow. Examination of flow stability of the jet configuration using a finite difference method is difficult. One has to be able to determine whether instabilities are numerical or physical in origin. The direct simulation method will therefore be used initially to investigate the stability of small-scale jets, and the methods of scaling stability criteria to jets of practical dimensions will be examined.

Our recent attempts to solve numerically the flow in curved nozzles have been unsuccessful because of problems associated with subsonic boundary conditions. Solutions for curved nozzles of practical dimensions and flow conditions are being developed based on a Monte Carlo method. It is expected that this approach will eliminate the subsonic boundary problem inherent in a finite difference calculation. In addition, methods for overcoming problems associated with subsonic flows in finite difference solutions of internal flows is under investigation by F. Blottner of Sandia Laboratories.

Improved techniques for calculating the UF_6 light isotope distribution are being investigated. The solution of the dynamic equations for the UF_6 light isotope parameters using a finite difference scheme and Monte Carlo solution of the collision integrals is being investigated under Sandia contract by S. M. Yen at the University of Illinois, Urbana-Champaign. Dr. Yen is also contracted to examine the effect of differences in gas-surface interaction between UF_6 and He on direct simulation. If this effect is found to be significant, the Monte Carlo codes will be modified to include greater details of the molecular behavior at the channel surface.

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