Two-dimensional multi-isotope separation in a gas centrifuge using finite element analysis

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Abstract: A finite element model of a gas centrifuge is developed to compute the optimal two-dimensional multi-isotope separation. The mass flow field generated using Onsager’s equation without the pancake approximation is used as an input to the diffusion equation for each uranium isotope in the initial form of partial differential equations (PDE). The PDEs are reduced to their weak forms and the resulting integrals evaluated using gauss quadrature. The systems of equations are solved using an optimization routine to satisfy the overall mass and concentration balance inside the machine. The solutions obtained can provide a holistic view of isotopic diffusion inside the centrifuge and the ability to quantify the molecular fraction of various uranium isotopes at a given radial and axial location at any desired initial and operating conditions. While several authors in the past have solved the multi-isotope diffusion problems using 1-D approximations, there are no known 2-D finite element models in literature. The findings of this work, therefore, is not only be significant for the applications of nuclear non-proliferation but also a great analytic tool for nuclear scientific community.

1. Introduction

Gas centrifuges are widely utilized in the nuclear industry for the primary purpose of enriching uranium in the fissionable isotope \(^{235}\text{U}\), which can be used as fuel for nuclear power reactors. The centrifuge is essentially a hollow rotor that is spun rapidly about its axis. This high rotation imparts a strong centrifugal force on the process gas and creates a large pressure gradient between the axis and the wall. During the uranium enrichment process, Uranium Hexafluoride (UF\(_6\)) gas is fed into the centrifuge near the axis through a feed pipe. The radial pressure gradient allows the heavier isotope of uranium, \(^{238}\text{U}\) to be concentrated near the wall and the lighter \(^{235}\text{U}\) molecules to be concentrated near the axis [1]. However, the radial difference in pressure only creates a fraction of overall isotopic separation. To substantially increase the degree of separation, a circulating flow along the length of the centrifuge can be created. If the bottom end of the centrifuge is heated, the warmer gas will rise and flow toward the opposite end along the axis, while the cooler gas will flow in along the wall to replace it. Additionally, a stationary scoop used for removing material can slow the flow of gas, reducing the centrifugal force and allowing it to rise. This then will allow the gas to flow along the centre toward one end and along the wall toward the other, creating an axial separation of lighter and heavier isotopes as shown in Figure 1.
The gas centrifuges can be classified into three different types based on their functionality, including the evaporative, concurrent, and counter-current machines. Because of its ability to significantly improve the separation effect, the counter-current gas centrifuge is the most preferable method for isotope separation and will be the focus of this paper. In the counter-current centrifuge, the gas flow represents a perturbation to the isothermal solid-body rotation which can be driven by a combination of external, mechanical, and thermal drives. The external drive can be established through the introduction of material at the axis and withdrawal at the top and bottom of the centrifuge. The mechanical drive occurs when the bottom scoop exerts drag force on the rotating flow and the thermal drive can be created with a temperature gradient between the top and bottom end caps. These physical mechanisms help maintain a counter current flow pattern inducing a convective mass flux which in turn increases the separation capabilities of the machine [1].

The primary objective of this work is to quantify the separation effect of any arbitrary centrifuge given certain physical and operating parameters of the machine. This process involves mapping the spread of individual isotopes present in the feed gas and determining the concentration of each at every location in the centrifuge rotor. The problem of isotopic diffusion inside a gas centrifuge has been studied previously in literature. However, the solutions are limited to binary mixtures of the $^{235}\text{U}$ and $^{238}\text{U}$ isotopes [3] [4] or 1-D simplifications of the multicomponent mixtures [5] [6] [7] [8]. A full two-dimensional distribution of isotopes in the centrifuge domain is important in minimizing uncertainties associated with determining machine’s separation capability obtained from the simplified models. It is also essential to account for multiple isotopes in the gas mixtures because in its natural state UF$_6$ consists of not only the $^{235}\text{U}$ and $^{238}\text{U}$ isotopes but also the $^{234}\text{U}$ isotope. Furthermore, recycled uranium can also contain traces of the $^{232}\text{U}$ and $^{236}\text{U}$ isotopes that are not considered in the binary mixture separation models. The capability of analyzing multi-component mixtures can provide flexibility in the use of the developed model to study the separation of stable or non-radioactive isotopes [9] [10], further demonstrating the influence of this work. Therefore, a higher fidelity model of the multi-component isotopic diffusion in a gas centrifuge has been developed using Finite Element Analysis (FEA). A brief background on the 2-D multi-component diffusion equation, description of the separation parameters, implementation of the FEA model, and a sample case study are presented in order.

For a mixture of $n$ isotopes, the separation is governed by a set of $n$ diffusion equations with appropriate boundary conditions. These equations are nonlinear two-dimensional partial differential equations (PDEs). The net transport vector $\bar{\phi}_k$ of isotope $k$ in the centrifuge is the balance between three different transport phenomena: pressure diffusion $\bar{\phi}_k^P$, back diffusion $\bar{\phi}_k^B$, and convection $\bar{\phi}_k^C$. Applying the steady-state conservation law for isotope $k$ in the centrifuge, the 2-D diffusion equation for isotope $k$ can be obtained as follows:

$$-D_k \frac{\partial^2 N_k}{\partial z^2} - D_k \frac{\partial^2 N_k}{\partial r^2} \left[ M_k \sum_{j=1}^{n} N_j - \frac{\partial}{\partial r} \left( M_k \frac{\partial N_k}{\partial r} \right) \right] + \rho V_z \frac{\partial N_k}{\partial z} = 0 \quad (1)$$

Equation (1) neglects the radial convection term $\rho V_r \frac{\partial N_k}{\partial r}$ from the continuity equation of fluid dynamics based on the assumption that the radial component $V_r$ of the velocity is predominant over the
axial component $V_z$ only in the very thin Ekman layers near the end caps of the machine. The definitions of the appropriate variables for Equation (1) are as follows:

- $\rho D$ → Constant self-diffusion coefficient for UF6,
- $N_k$ → Mole fraction of isotope $k$,
- $\Omega$ → Angular velocity of the centrifuge,
- $R$ → The specific gas constant,
- $T$ → The average temperature of the gas,
- $M_k$ → The molecular weight of the $k$th component,
- $\dot{m} A_{\text{f}}$ → Axial mass flux obtained from Onsager’s equation without the pancake approximation including the source and sink terms,
- $(r,z)$ → The radial and axial directions of the centrifuge domain respectively.

The boundary conditions associated with Equation (1) are given by the following:

At $r=a$ (radius of the centrifuge):
\[ \frac{\partial N_k}{\partial r} - \frac{\Omega^2 a}{RT} M_k - \sum_{j=1}^{n-1} M_j N_j \ast N_k = 0. \] (2)

At $r=0$ (axis of the centrifuge): \[ \frac{\partial N_k}{\partial r} = 0. \] (3)

At $z=0$ (bottom of the centrifuge):
\[ \int_0^a (-\rho D \frac{\partial N_k}{\partial z} + \rho V_z N_k) 2\pi r dr = -F(1-\theta) N_{w,k} \] (4)

At $z=z_H$ (height of the centrifuge):
\[ \int_0^a (-\rho D \frac{\partial N_k}{\partial z} + \rho V_z N_k) 2\pi r dr = F \theta N_{p,k}. \] (5)

where $F$ is the feed flow rate, and $\theta$ is the “cut”, the ratio of product flow rate over feed flow rate. The above boundary conditions indicate that there is no radial transport at the rotor wall and on the axis while the axial transport over the top and bottom end caps equals the withdrawal flow rates. For a mixture of $n$ isotopes, $n$ equations like Equation (1) together with $n$ sets of boundary conditions expressed above in Equations (2-5) need to be solved. One of the key inputs to the above equation is the mass flux term $\dot{m} A_{\text{f}}$, which comes from the solution of the feed gas flow field inside the centrifuge rotor. The centrifuge fluid dynamics has been studied extensively in literature since the 1960s [11] [12] [13] [14] [15] [16] [17]. The most recently developed finite element solution of the mass flow is utilized to obtain the required axial mass flux term for the diffusion equation above.

Two important parameters used to quantify the separation capability of a gas centrifuge are the overall separation factor, $\gamma$, and the separative work, $\Delta U$.

\[ \gamma = \frac{N_{P}}{N_{P} + \frac{1-N_\text{w}}{N_\text{w}}}, \] (6)

$N_P$ and $N_w$ are the concentrations of the $^{235}$U isotope at the product and the tails, respectively.

\[ \Delta U = F \theta V(N_P) + (1-\theta) V(N_{\text{w}}) - V(N_F), \] (7)

where $F$ is the feed flow rate, $\theta$ is the cut, and $V$ is the value function [1].

2. Methodology

2.1. Finite Element Approximation
The above-described set of 2-D nonlinear partial differential diffusion equations can be solved numerically using FEA. The centrifuge domain is described in Figure 2.
The 2-D axisymmetric centrifuge domain defined in Figure 2 can be discretized into several two-dimensional finite elements. Given the rectangular domain of the centrifuge, four-node rectangular elements are used to simplify the domain. A sample element with its four nodes labelled is presented below in Figure 3. The illustration of a sample mesh with centrifuge domain divided into rectangular elements can be seen in Figure 4. It is imperative to determine the ideal number of elements required for accurate solution derivation. For this purpose, a mesh independence study has been conducted and will be discussed in the next section.

Figure 3. Linear four-node rectangular element

Figure 4. A sample mesh of rectangular elements used to approximate the centrifuge domain. More elements are prescribed near the wall of the machine to capture the gradient effects.

Utilizing the above mesh, the diffusion equation can be simplified to solve for each individual element prior to solving for the entire domain. To ensure that Equation (1) can be solved element wise, it is important to develop its “weak” formulation to reduce the order of differentiation. Let,

\[ H_k = \frac{\nu^2}{RT} (M_k - \sum_{j=1}^{n} M_j N_j) \]  

The weak form of Equation (1) above can be computed by first multiplying the entire equation by an arbitrary weighting function, \( w(r,z) \), and integrating the product over the computational domain of the centrifuge.

\[ \int \int w(r,z) \left( \frac{\partial^2 N_k}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial N_k}{\partial r} \right) - H_k r^2 N_k \right) \rho \frac{\partial z}{\partial r} \left( \frac{\partial N_k}{\partial r} \right) \ d\Omega = 0 \]  

(9)
Taking each term separately and conducting integration by parts in the following expressions:

\[ \iint w \frac{\partial^2 N_k}{\partial z^2} \cdot \partial r dz dr = \int_{r=0}^{a} \int_{z=0}^{z_H} (w \frac{\partial N_k}{\partial z} \cdot \frac{\partial w}{\partial z} - \frac{\partial N_k}{\partial r} \cdot \frac{\partial w}{\partial r}) r dz dr \]  \hspace{1cm} (10)

\[ \iint w \left( \frac{\partial}{\partial r} \left( \frac{\partial N_k}{\partial r} \cdot H_k \frac{\partial z_0}{\partial z} N_k \right) \right) dz dr = \int_{r=0}^{a} \int_{z=0}^{z_H} (w \frac{\partial N_k}{\partial r} \cdot \frac{\partial w}{\partial z} - \frac{\partial N_k}{\partial z} \cdot \frac{\partial w}{\partial r}) r dz dr \]  \hspace{1cm} (11)

\[ \frac{\rho V_z}{\rho D} \iint w \frac{\partial^2 N_k}{\partial z^2} \cdot \partial r dz dr = \frac{\rho V_z}{\rho D} \int_{r=0}^{a} \int_{z=0}^{z_H} (w \frac{\partial N_k}{\partial z} \cdot \frac{\partial w}{\partial z} - \frac{\partial N_k}{\partial r} \cdot \frac{\partial w}{\partial r}) r dz dr \]  \hspace{1cm} (12)

Applying the appropriate boundary conditions and combining the terms, the weak form is obtained as follows:

\[ \iint_{r=0}^{a} \int_{z=0}^{z_H} \left( w \frac{\partial N_k}{\partial z} \cdot \frac{\partial w}{\partial z} - \frac{\partial N_k}{\partial r} \cdot \frac{\partial w}{\partial r} \right) r dz dr = 0 \]  \hspace{1cm} (13)

### 2.2. Interpolation Functions

The FEM approximates \( N_k(r, z) \) over an element \( \Omega^e \) as \( N_k^e(r, z) \). \( N_k^e \) must be differentiable as required in the weak form of the problem, the polynomials used to represent \( N_k^e \) must be complete and all terms in the polynomial should be linearly independent. For an element with four nodes as shown in Figure 3, the polynomial

\[ \sum_{i=1}^{4} c_i \psi_i(r, z) \]  \hspace{1cm} (14)

can be used to interpolate the mole fraction of isotope \( k \) in an element \( e \). In Equation (14), \( c_i \)'s are arbitrary constants to be determined later. The interpolation function should represent the nodal variables at the four nodal points of the element. Therefore, substituting the \( r \) and \( z \) values at each nodal point gives

\[ \begin{bmatrix} N_{k1}^e \\ N_{k2}^e \\ N_{k3}^e \\ N_{k4}^e \end{bmatrix} = \begin{bmatrix} \psi_1(r, z) & \psi_2(r, z) & \psi_3(r, z) & \psi_4(r, z) \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \end{bmatrix} \]  \hspace{1cm} (15)

Here, \( r_1 \) and \( z_1 \) are the coordinate values at the \( i^{th} \) node and \( N_{ki} \) is the nodal value of mole fraction of isotope \( k \). The constants \( c_i \)'s can be obtained by multiplying both sides of Equation (15) by the inverse of the matrix. Substituting the resulting equations for the constants back into Equation (14) results in

\[ N_k^e = \sum_{i=1}^{4} \psi_i(r, z) \cdot N_{ki} \]  \hspace{1cm} (16)

where \( \psi_i^e(r, z) \) are the shape functions for linear rectangular element:

\[ \psi_1^e = \left( \frac{r-a}{b} \right) \left( \frac{z-a}{b} \right), \ \ \psi_2^e = \frac{1}{b} \left( 1 - \frac{r-a}{b} \right), \ \ \psi_3^e = \frac{1}{b} \left( 1 - \frac{z-a}{b} \right), \ \ \psi_4^e = \left( \frac{a}{b} \right) \]  \hspace{1cm} (17)

Here \( a \) and \( b \) are the element length and width respectively as illustrated in Figure 3. Letting the weighting function \( w(r, z) = \sum_{i=1}^{4} \psi_i N_{ki} \) and substituting into Equation (13), the weak form of the diffusion equation can be re-written in terms of the shape functions for each element. The double integral in the weak form of the equation is evaluated using gaussian quadrature to obtain the element stiffness matrix, \([K]^e\). Such process is repeated for every element in the centrifuge mesh and each of the individual element matrices are assembled into the system stiffness matrix, \([K]\). The system stiffness matrix is a diagonally dominant sparse matrix whose size is equal to system degrees of freedom (sdof) \( x \) (sdof). The total system degrees of freedom are evaluated by multiplying the total number of unknowns per node times the total number of nodes in the mesh. Thus, the system equation to be evaluated can be written as follows:

\[ [K]_{sdof} = q^e(N_k)_{sdof} \cdot [H]_{sdof} \]  \hspace{1cm} (18)

The mole fraction of isotope \( k \) is unknown everywhere in the centrifuge domain except for the nodes corresponding to the feed region, where the feed values are known. This ensures that the
right-hand side vector \( f \) in Equation (18) is non-zero. The solution of Equation (18) is obtained by solving for the concentration vector using Matlab’s “mldivide” function.

\[
\{N_i\}_{sdof \times 1} = [K]_{sdof \times sdof} \{f\}_{sdof \times 1}.
\] (19)

The newly developed code named MultiPort builds the coefficient matrix and force vector and solves the matrix in Equation (19) using built-in algorithms. The built-in routine selects the most appropriate method from several different solving techniques, including QR decomposition, a triangular solver, a permuted triangular solver, LDL decomposition, LU decomposition, a Hessenberg or Cholesky solver, a diagonal solver, or a banded solver depending on the characteristic of the coefficient matrix, \( K \). The finite element solver is designed to satisfy several physical properties of the diffusion equation to ensure convergence. The key criteria required to ensure accurate solution are as follows:

1) The sum of the molecular weights of the isotopes multiplied by the concentrations of each at every nodal point equals the molecular weights of the gas. For UF\(_6\) mixture,

\[
\sum_{i=1}^{n} M_i N_i = 352 \text{ kg mol}\n
\text{ where } n = \text{ total number of isotopes present, } M_i = \text{ molecular weight of each isotope } i \text{ and } N_i = \text{ mole fraction of isotope } i.
\]

2) The sum of the concentration of all of the isotopes at every nodal point is 1: \( \sum_{i=1}^{n} N_i = 1 \).

3) The overall balance of isotopic species is established.

\[
N_{F,K} = \theta N_{P,K} + (1-\theta) N_{W,K}
\] (20)

Here, \( N_{F,K}, N_{P,K}, \text{ and } N_{W,K} \) are the concentrations of isotope \( k \) at the feed, product, and tails end respectively. Equation (20) needs to be satisfied for all the isotopes present in the gas mixture for the solution to converge.

3. Results

The output of the new finite element model, MultiPort, has been examined using hypothetical gas centrifuges. The physical as well as the operating parameters of Rome [18], Iguacu [19], and Darmstadt [20] machines found in literature are described in Table 1 below. The diffusion results here are presented for the Rome machine operated at 600 m/s rotor speed.

Table 1. Physical properties of the fictitious gas centrifuges.

| Parameter            | Variable | Unit | Rome  | Iguacu | Darmstadt |
|----------------------|----------|------|-------|--------|-----------|
| Radius               | \( a \)  | \( m \) | 0.25  | 0.06   | 0.25      |
| Length               | \( z_H \) | \( m \) | 5     | 0.48   | 15        |
| Average gas temperature | \( T \) | \( K \) | 320   | 300    | 340       |
| Wall pressure        | \( p \)  | torr | 100   | 60     | 500       |
| Cut                  | \( \theta \) | -    | 0.5   | 0.5    | 0.222     |

The isotopic composition of the UF\(_6\) gas at the feed is given in Table 2. These values for spent reactor uranium fuel are specified in ASTM C787-06 Standard Specification for Uranium Hexafluoride for Enrichment found in Nuclear Material Safeguards for Uranium Enrichment Plants [21].
Table 2. Isotopic composition of the feed gas [molecular %].

| Isotope | %   |
|---------|-----|
| U-232   | 10^-9 |
| U-234   | 0.02 |
| U-235   | 0.9  |
| U-236   | 0.4  |
| U-238   | 98.68 |

Before Equation (1) can be solved, the solution of the mass flow is required. As mentioned previously, the centrifuge fluid dynamics has been studied extensively in literature. For this work, the gas flow field inside the Rome machines was generated using the CurvSOL code developed by Witt [16] and later modified by Thomas [17]. The optimal values of the linear wall temperature, scoop drag force, and the feed rate were combined to simulate the total drive effects in the UF6 gas flow. CurvSOL includes the sources and sinks of mass, momentum, and energy and considers the effect of rotor curvature in the obtained solutions. The mass that is introduced by the feed at the axial midpoint is removed through scoops located at the top and the bottom ends of the machine. The bottom scoop is modelled as a sink of angular momentum while the top scoop is modelled as a boundary since it is shielded from the gas flow region by a baffle. Using the mass flux profile and the centrifuge mesh, MultiPort solves either the binary or the multi-component mixture based on the user input. A mesh independence study was conducted to evaluate the appropriate density of the element mesh required for the centrifuge domain. The results are shown in Table 3.

Table 3. Mesh independence study to determine the optimal number of FEA nodes.

| # of nodes | γ   | % diff | ΔU (SWU yr^-1) | % diff |
|------------|-----|--------|----------------|--------|
| 4750       | 1.678 | 2.483  | 47.401         | 10.915 |
| 9500       | 1.721 | 0.510  | 52.874         | 2.727  |
| 19000      | 1.729 | 0.554  | 54.336         | 2.308  |
| 28500      | 1.739 | 0.596  | 55.605         | 2.118  |
| 38000      | 1.749 | 0.0229 | 56.795         | 0.0772 |
| 47500      | 1.749 | 0.672  | 56.751         | 2.382  |
| 57000      | 1.761 | 0.238  | 58.119         | 0.826  |
| 66500      | 1.765 | 0.672  | 58.601         |        |

The total number of 38000 nodes or the 95x400 grid size, 95 nodes in the radial direction and 400 nodes in the axial direction, is deemed sufficient for the centrifuge in consideration. Any further increase in the number of nodes results in insignificant changes (less than 1%) of the separation parameters but larger than 20% increase in convergence time.

The 3-D surface plots of the individual isotopic diffusion are presented below for the Rome machine distribution. The surface plots in Figure 5 show the concentration of specific isotope at the given radial and axial position inside the rotor volume. The x-axis is the non-dimensional radial location in scale heights from x=0 at the wall and x=15 at the “top of the atmosphere.” The y-axis shows the non-dimensional axial locations with y=0 at the bottom and y=1 at the top. The concentration of the isotope is given in the z-axis in molecular fraction.
The isotopic distribution plots show great agreements with the expected physics that the higher concentration of the lighter isotope is at the top end of the centrifuge rotor while a higher concentration of the heavier isotope is at the bottom end. For the $^{235}$U surface plot in a), the concentration of the isotope increases with increasing axial distance from the feed location at the axial midpoint while decreases with decreasing distance. The radial separation is noticeably minimal compared to the axial, with approximately only 5% increase in concentration between the rotor wall and the top of the atmosphere. The shape of the isotopic distribution plots for the minor isotopes $^{234}$U and $^{236}$U agree with that of $^{235}$U as seen above. For the heavier $^{238}$U isotope, the distribution is reversed with higher concentrations toward the bottom end of the machine and lower toward the top. One of the novel features of the new model is the ability to obtain radial variation of the isotopic concentrations, which was not possible in the older diffusion models. Figure 6 shows the radial distribution of the isotopes at four different axial points in the centrifuge rotor. On the bottom half of the machine, the heavier isotopes are heavily concentrated while at the top half, the lighter isotopes can be found in greater concentrations. The lighter isotopes are enriched at a greater rate towards the axis on the right while the heavier near the wall on the left.

Figure 5. The 3-D surface plots of the individual uranium isotopes a) $^{235}$U b) $^{234}$U c) $^{236}$U and d) $^{238}$U.
The new 2-D model of the diffusion equations has been verified by comparing the results with the previously available simplified code. The comparison was done by radially averaging the solutions of the above output and comparing with the 1-D model [8]. The concentration values reported by the new MultiPort code are within 10% of the radially averaged concentrations obtained from the 1-D model. Since there are no known open-source experimental data available for the separation of uranium isotopes, the code has been validated against the separation of stable isotopic mixtures and shows excellent agreements. The detailed validation against the available experimental data as well as the quantification of uncertainty and sensitivity analysis is a subject of an ongoing study and will be published in the near future.

4. Conclusions
The 2-D diffusion equation was solved using the newly developed finite element code to calculate the multi-isotopic gradients inside a single centrifuge machine. The finite element model utilized linear quadrilateral elements to approximate the mole fractions of arbitrary number of isotopes present in the process gas. This work is beneficial in examining the two-dimensional variation of uranium isotopes that was not available in the past. The new code is designed to solve either the binary mixture or the multi-isotopic mixture according to user’s preference. This feature allows the user to not only limit the calculations to uranium hexafluoride but also to non-radioactive and stable isotopic mixtures. The information obtained here can be used to determine the machine’s separative capability as well as visualize and understand the spread of individual isotopes inside the rotor volume. The new model not only provide the concentrations of uranium isotopes at the outlets of the machines but also information about how the isotopes are distributed in the interior regions. This type of knowledge can be significant in identifying how the machines were utilized and whether any misuse was conducted by the operators and is crucial for implementing nuclear safeguards. Moreover, the separation parameters can be used to conduct additional calculations in the cascade models and has the potential to serve as a beneficial tool for facility level non-proliferation applications. The new software has been verified using available open source 1-D results that shows great agreements with the previously published data. Further refinement of the code in terms of optimizing the mass flow solution and generalizing machine parameters has been completed [22]. As part of the future work, the mass flow and isotopic diffusion codes will be combined into a single stream with improved user interface.

Figure 6. Radial concentration distributions normalized using the feed concentrations of the four minor uranium isotopes at four different axial locations a) $y=0$ b) $y=0.25$ c) $y=0.75$ and d) $y=1$. 

![Radial Concentrations at z/h=0.75](image1)

![Radial Concentrations at z/h=1](image2)
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