Chapter 2

Separative Performance in a Vacuum Arc Centrifuge

In this chapter, the variation of isotope concentration along the rotation region of a vacuum arc centrifuge is investigated for a two species plasma. Using the fluid equations of motion for both isotope species, together with continuity, the spatial dependence of the isotope concentration and separation factor is numerically solved. As well as comparison with experimental data [13], which details the spatial evolution of the separation profile for a Cu-Ni plasma, the important physical processes occurring in the plasma column as separation takes place are identified.

2.1 Introduction

The general theory of isotope separation in centrifuges operating with neutral gas was expounded by Cohen [43] in the early 1950’s. In 1966, Bonnevieu analysed diffusion due to ion-ion collisions in a multi-component fully ionized plasma [7], and later, in 1971 [8], demonstrated experimentally that a rotating plasma could be used to separate isotopes of different mass. The radial variation of the isotope concentration in a radially configured plasma centrifuge was analysed by Simpson in 1975 [44]. Later, in 1981 [45], Simpson investigated the separative power of radial geometry plasma centrifuges, and found that a single component plasma centrifuge of that type could not match the separative power of a gaseous centrifuge for uranium enrichment. The theoretical development here treats a fully ionized plasma and owes
more to Bonnevier's ground-breaking 1966 paper [7] than the more recent work in partially ionized plasmas [44, 45].

In 1984, Geva et al. [13] presented experimental data on the characteristics of multi-species isotope separation in a CO$_2$ laser triggered vacuum arc centrifuge (see Fig. 2-1). In particular, they measured the enrichment profiles of a Cu-Ni plasma, at various axial positions along the plasma column. From these profiles, they provided an empirical estimate of the average number of orbits each ion undergoes until separation approaches an asymptotic value. To date, no theoretical analysis of the axial variation of isotope separation in an axial plasma centrifuge has been undertaken. The object of the research described in this chapter is to find the spatial evolution of isotope concentration and separation, and compare the findings with measurements made by Geva et al. of the axial evolution of separation profiles in a Cu-Ni plasma [13].

![Diagram of the Yale University Vacuum Arc Centrifuge](image)

Figure 2-1: The Yale University Vacuum Arc Centrifuge.

The chapter is organized as follows: Sec. 2.2 introduces the plasma fluid model of the system, lists any assumptions made, and reduces the problem to a second order partial differential describing centrifugal separation. Section 2.3 describes a numerical solution for separation, whilst Sec. 2.4 sets up the simulation parameters for a numerical solution, using the plasma properties described in the work by Geva [13]. Section 2.5 presents a comparison of the theo-
retical separation profile against the experimental data collected by Geva et al. [13], and plots the isotope concentrations of each ion species. Finally, Sec. 2.6 contains concluding remarks.

2.2 Plasma Model

The plasma in a VAC is fully ionized, and consists of electrons and the material to be separated. The aim of this work is to investigate isotope separation in the rotation region following the anode grid. In this region, the plasma is assumed to consist of ions labelled $i_1$ and $i_2$, which have average charge $Z$, masses $m_1 = m$ and $m_2 = m + \Delta m$, and ion densities $n_{i1}$ and $n_{i2}$ respectively. Separation effects associated with differing ionic charge states [13] are not investigated here. The absence of neutrals reduces the analysis to a 3-fluid steady state model. The geometry of the model is shown in Fig. 2-1, with the axial position $z = 0$ m corresponding to the anode mesh.

Following the established physical picture of VACs, which has arisen from experimental data [11–13, 28, 35, 39] and theoretical analyses [29, 32, 36, 39], the plasma column is assumed to exhibit rigid body rotation at frequency $\omega_0$, and an axial transport velocity $v_{e0}$ which is independent of radius. Like similar treatments [29, 32, 34–36], viscous effects are ignored and the plasma assumed to be isothermal with temperature $T$. Measurements [39] made with a Fabry-Perot interferometer show that the ion temperature does not vary over the duration of the discharge, and is observed to be uniform with radius. A comparison of experimental data from VAC’s indicates that the ion and electron temperatures are similar within the plasma column [35, 39]. An isothermal plasma is thus a reasonable first approximation.

Terms of order $m_e/m$ are neglected, where $m_e$ is the electron mass. The radial profile of the mass density in the plasma column is close to Gaussian [11–13, 27]. Here a more generalized functional form is taken

$$\rho(r) = \rho(0)f\left(\frac{r^2}{R^2}\right)$$

(2.1)

where $R$ is some characteristic radius, $f\left(\frac{r^2}{R^2}\right)$ is a generalized function with $f(0) = 1$, and $\rho(0)$ is the on-axis mass density.

The ion-ion collision interaction is described by the Coloumb cross-section [46]. Applying the Kihara correction factor, which treats the Coulomb shielding process with greater precision,
the on-axis average ion-ion momentum transfer collision frequency \( \nu_{ii}(0) \) can then shown to be

\[
\nu_{ii}(0) = n_i(0) \frac{4\sqrt{2\pi}}{3} \left( \frac{\mu}{k_B T} \right)^{3/2} \left( \frac{Z^2 e^2}{4\pi \varepsilon_0 \mu} \right)^2 (\ln \Lambda - 1.37)
\]

(2.2)

where \( n_i(0) \) is the total ion on-axis density, \( \mu \) is the reduced ion mass and \( \ln \Lambda \) is the Coulomb logarithm. Neglecting the slight density dependence of the Coulomb logarithm, \( \nu_{ii} \) is proportional to the total ion density, and can be written using Eq. (2.1) as

\[
\nu_{ii} = \nu_{ii}(0) f \left( \frac{r^2}{R^2} \right)
\]

(2.3)

For a typical VAC [13, 25], the on-axis ion-ion collision frequency is of the order \( \nu_{ii}(0) \approx 10^6 - 10^7 \text{ s}^{-1} \), the axial length of the centrifuge \( z_f \), is of the order of one metre, and the ion axial velocity is of the order \( v_{z0} \approx 10 \text{ km s}^{-1} \). Using this data it is found that the mean time between on axis ion-ion collisions evaluates to \( 1/\nu_{ii}(0) \approx 0.1 - 1 \mu s \), which is significantly less than the axial transit time, given by \( z_f/v_{z0} \approx 100 \mu s \). This implies that many collisions occur between ions along the length of the centrifuge, i.e.

\[
\nu_{ii} \gg \frac{v_{z0}}{z_f}
\]

(2.4)

This is a key approximation in the following treatment. The assumption is somewhat weaker than the standard diffusion [47] assumption, which is that the velocities of diffusion of the component species are small compared with the bulk plasma velocity \( \mathbf{v} \) of the fluid.

The concentration of the lighter isotope species \( C_i \) is defined as the ratio of the lighter isotope ion density \( n_{i1} \) to the total heavy particle number density \( n_i = n_{i1} + n_{i2} \), that is, \( C = n_{i1}/n_{i2} \). The average mass \( \bar{m} \) is equal to the weighted sum of the two isotope masses

\[
\bar{m} = mC + (m + \Delta m)(1 - C) = m + \Delta m(1 - C)
\]

For each isotope species, the isotope velocities are \( \mathbf{w}_1 \) and \( \mathbf{w}_2 \). In terms of the diffusion velocities for each isotope, \( \mathbf{v}_1 \) and \( \mathbf{v}_2 \), \( \mathbf{w}_1 = \mathbf{v}_1 + \mathbf{v} \), and \( \mathbf{w}_2 = \mathbf{v}_2 + \mathbf{v} \) where \( \mathbf{v} \) is the mass
average or bulk plasma velocity. Neglecting electrons, the definition of \( \mathbf{v} \) implies

\[
\mathbf{v} \equiv \frac{1}{m} [mC\mathbf{w}_1 + (m + \Delta m)(1 - C)\mathbf{w}_2]
\]

which upon simplifying yields

\[
mc\mathbf{v}_1 + (m + \Delta m)(1 - C)v_2 = 0 \quad (2.5)
\]

With the assumptions made at the start of this section, the bulk plasma velocity is given by \( \mathbf{v} = (0, \omega_0 r, v_z) \). The steady state fluid equations of motion for each isotope species can be written as

\[
m_1 n_i (\mathbf{w}_1 \cdot \nabla) \mathbf{w}_1 = Z E n_i (\mathbf{w}_1 + \mathbf{v}_1 - \mathbf{B}) - \nabla p_1 + \frac{n_{i1} n_{i2}}{n_i} \mu \nu_{ii}(\mathbf{w}_2 - \mathbf{w}_1) \quad (2.6)
\]

\[
m_2 n_i (\mathbf{w}_2 \cdot \nabla) \mathbf{w}_2 = Z E n_i (\mathbf{w}_2 + \mathbf{v}_2 - \mathbf{B}) - \nabla p_2 + \frac{n_{i2} n_{i1}}{n_i} \mu \nu_{ii}(\mathbf{w}_1 - \mathbf{w}_2) \quad (2.7)
\]

where \( E \) is the electric field, \( B \) is the magnetic field, the ion species partial pressure \([46]\) are \( p_1 = n_{i1} k_B T \) and \( p_2 = n_{i2} k_B T \), and the last terms on the right hand sides of Eqs. (2.6) and (2.7) represent momentum exchange between the ion species. For typical conditions of a VAC \([28]\), it is reasonable to neglect momentum transfer in electron-ion collisions in Eqs. (2.6) and (2.7).

Dividing Eqs. (2.6) and (2.7) by \( n_{i1} \) and \( n_{i2} \) respectively and taking the difference

\[
m_2 (\mathbf{w}_2 \cdot \nabla) \mathbf{w}_2 - m_1 (\mathbf{w}_1 \cdot \nabla) \mathbf{w}_1 = Z v (\delta \times \mathbf{B}) - \mu \nu_{ii} \delta - k_B T [\nabla (\ln n_{i2}) - \nabla (\ln n_{i1})] \quad (2.8)
\]

where \( \delta = (\delta_r, \omega_1 r, \delta_z) = \mathbf{v}_2 - \mathbf{v}_1 \) is the difference in diffusion velocities. Using Eq. (2.5), one finds

\[
\mathbf{v}_2 = \delta \frac{mc}{m}, \quad \mathbf{v}_1 = \frac{m + \Delta m}{m} (1 - C)
\]

The three components of Eq. (2.8), employing the above substitutions for the diffusion velocities and the expression for the mass average velocity \( \mathbf{v} \), become:

\[
\frac{\Delta m}{m} \omega_0^2 r + 2 \frac{m + \Delta m}{m} \omega_1 \omega_0 r = - \frac{Z e B_0}{m} \omega_1 r + \frac{\mu \nu_{ii} \delta_r}{m} - k_B T \frac{\partial C}{\partial r} \frac{1}{C(1 - C)} \quad (2.9)
\]

\[
-2 \frac{m + \Delta m}{m} \omega_0 \delta_r = \frac{Z e B_0}{m} \omega_1 r + \frac{\mu \nu_{ii} \omega_1 r}{m} \quad (2.10)
\]
\[
\frac{\mu}{m} \nu_{ii} \delta_z = \frac{kT}{m} \frac{\partial C}{\partial z} \frac{1}{C(1-C)}
\] (2.11)

where use has been made of the approximation (2.4). The continuity equation in the steady state for each ion species \( k \) is \( \nabla \cdot (\rho_k \mathbf{w}_k) = 0 \). Subtracting the continuity equations for the two species gives

\[
\nabla \cdot \frac{\rho(r)}{m} [(m + \Delta m)(1 - C)(\mathbf{v}_2 + \mathbf{v}) - mC(\mathbf{v}_1 + \mathbf{v})] = 0
\] (2.12)

Substituting for the diffusion velocities \( \mathbf{v}_1 \) and \( \mathbf{v}_2 \), and the bulk velocity \( \mathbf{v} \), Eq. (2.12) becomes

\[
C(1-C) \frac{\partial \delta_r}{\partial r} + \left( 1 - 2 \frac{m}{m} C \right) \delta_r \frac{\partial C}{\partial r} + \\
C(1-C) \delta_r \left( 1 + 2 \frac{1}{R^2} \frac{j(r^2/R^2)}{f(r^2/R^2)} \right) + \\
C(1-C) \frac{\partial \delta_z}{\partial z} + \left( 1 - 2 \frac{m}{m} C \right) \delta_z \frac{\partial C}{\partial z} = v_{z0} \frac{\partial C}{\partial z} \delta_x
\] (2.13)

For all isotopes separable in VAC’s \( \Delta m/m \sim 0.1 \) at most, and changes in the concentration from the natural abundance ion concentration \( C_0 \) are also typically less than 10%. Thus, it is reasonable to neglect the product. That is, the assumption

\[
\left| \frac{\Delta m}{m} (C - C_0) \right| \ll 1
\] (2.14)

is made, and terms with coefficients of this relative order neglected in the ensuing analysis. Making this assumption means the average mass \( m \) is a constant, \( M \). Using assumption (2.14), and comparing the second term on the LHS of Eq. (2.13) with the first term, the second term can be approximated as

\[
\left( 1 - 2 \frac{m}{M} C \right) \delta_r \frac{\partial C}{\partial r} \approx (1 - 2C) \delta_r \frac{\partial C}{\partial r}
\]

Using Eq. (2.11) gives an expression for \( \delta_z \), appearing in the last two terms on the LHS of Eq. (2.13). For typical plasma parameters (see Table 2.1) the plasma thermal velocity \( v_{\text{th}} = \sqrt{3k_B T/M} \) is at most comparable to the bulk transport velocity \( v_{z0} \). Using the approximation (2.4), the last two terms on the LHS of Eq. (2.13) can then be neglected when compared
to the RHS. With these simplifications, Eq. (2.13) becomes
\[
C(1 - C) \frac{\partial \delta_r}{\partial r} r + (1 - 2C) \delta_r \frac{\partial C}{\partial r} r + C(1 - C) \delta_r \left( 1 + 2 \frac{\gamma^2 f}{r^2} \frac{\gamma^2 f}{r^2} \right) = v_{z0} \frac{\partial C}{\partial z} r
\]  
(2.15)

which can also be written as
\[
\frac{\partial}{\partial r} \left[ C(1 - C) \delta_r r f \left( \frac{r^2}{R^2} \right) \right] = v_{z0} \frac{\partial C}{\partial z} r f \left( \frac{r^2}{R^2} \right)
\]  
(2.16)

The radial component of the difference in diffusion velocities can be eliminated from Eq. (2.16) by solving Eqs. (2.9) and (2.10) for \( \delta_r \), and then substituting back into Eq. (2.16). Equation (2.16) becomes
\[
\frac{\partial}{\partial r} \left( \frac{C(1 - C) \Delta m \omega_i^2 \gamma^2 + k_B T \frac{\partial C}{\partial r} r}{\mu v_i(0) \left( 1 + \frac{\gamma^2}{\nu_i^2} \right)} \right) = v_{z0} \frac{\partial C}{\partial z} r f \left( \frac{r^2}{R^2} \right)
\]  
(2.17)

where
\[
\Omega_i^* = \frac{M}{\mu} \left( \frac{2m}{M} \times \frac{m + \Delta m}{M} \omega_0 + \omega_i \right)
\]
the term \( \omega_i = ZeB_z/M \) is the ion cyclotron frequency, and \( B_z \) the axial magnetic field strength. The term \( \Omega_i^*/\nu_i^2 \) is a generalized Hall term, and can be replaced by a generalized Hall parameter \( \beta_i^* = \beta_i^*(0) f \left( \frac{r^2}{R^2} \right)^{-2} \), where \( \beta_i^*(0) = \Omega_i^*/\nu_i^2(0) \). For \( r \gg R \), the parameter \( \beta_i^* \gg 1 \), and the LHS of Eq. (2.17) becomes small. In this limit, the plasma is said to be “frozen”, where the axial dependence of \( C \) becomes small, and the plasma approaches a collisionless state. The onset of plasma “freezing” occurs when \( \beta_i^* \) becomes comparable to unity.

With the change of variable \( y = \frac{r^2}{R^2} \), and the introduction of a normalized axial position variable \( z' = \frac{z}{z_0} \), Eq. (2.17) reduces to
\[
\frac{\partial}{\partial y} \left[ y \left( \frac{AC(1 - C) + \frac{\partial C}{\partial y}}{1 + \beta_i^2 f(y)^{-2}} \right) \right] = f(y) \frac{\partial C}{\partial z'}
\]  
(2.18)

where
\[
A = \frac{\Delta m \omega_0^2 R_0^2}{2k_B T}, \quad z_0 = \frac{\mu v_{z0} R_0^2}{4k_B T}
\]

In Eq. (2.18), the constant \( z_0 \) scales the axial position, and the constant \( A \) determines the
steady state separation factor $\alpha$ [11–13, 27] for a two-species plasma. The constant $R_0$ in the definitions for $A$ and $z_0$ scales the characteristic radius, $R$. In a VAC, the separation factor $\alpha$ is defined to be the ratio of matter flux of the heavy to light isotope at a given radius $r$, divided by the on-axis ratio of matter flux. That is,

$$\alpha = \frac{\left[ \frac{n_{12} w_{22}}{n_{11} w_{11}} \right]_r}{\left[ \frac{n_{12} w_{22}}{n_{11} w_{11}} \right]_{r=0}} \quad (2.19)$$

It can shown that, for the VAC conditions considered in this work, to a good approximation the species velocities cancel in Eq. (2.19), and $\alpha$ can be written as

$$\alpha = \left[ \frac{1 - C}{C} \right]_r / \left[ \frac{1 - C}{C} \right]_{r=0} \quad (2.20)$$

where both species have average charge state $Z$. In the steady state, Eq. (2.20) reduces to

$$\alpha = \exp \left( A_0 \times \frac{r^2}{R^2} \right) \quad (2.21)$$

Equation (2.18) is functionally equivalent to the diffusion equation derived by Bonnevier [7]. The difference between treatments arises from Bonnevier’s neglect of terms of order $\omega_0/\omega_i$, which are retained here since in some experiments rotation velocities in VAC’s have been reported which approach the ion cyclotron frequency [40]. Applying the condition $\omega_0/\omega_i \ll 1$ to this treatment, Eq. (2.18) reduces to the diffusion equation derived by Bonnevier [7].

### 2.3 Numerical Analysis

A straightforward explicit marching scheme in $z'$ was used to solve Eq. (2.18). A step size $\Delta z = 4.0 \times 10^{-8}$ in $z'$, with $C$ evaluated at 1500 radial points between $y = 0$ and $y = 30$ (giving a step size in $y$ of $\Delta y = 0.02$), was sufficient for numerical convergence and stability. Numerical convergence and stability were checked by doubling $\Delta z$ and $\Delta y$, the effect of which was found to be negligible.
The boundary condition applied to the simulation was that at the outer radial boundary, \( \frac{\partial C}{\partial y} \) was zero. To verify that this zero gradient boundary condition had no effect on predictions, the boundary location was moved inward to \( y = 15 \); this change had negligible effect. Hence, the situation modelled represents a free plasma column with no influence from any vessel walls.

The initial conditions of the simulation specify the concentration \( C \) at \( z = 0 \) is radially uniform, and equal to \( C_0 \), the natural abundance of the lighter isotope. To verify the accuracy of the simulation, transport continuity of mass was checked along the plasma length. This was preserved to within 0.1\% for the results presented below.

2.4 Plasma Model Parameters

In 1984, Geva et al. [13] presented a set of experimental flux and separation profiles, detailing the axial evolution of the flux density and the enrichment profile in a Cu-Ni plasma. The Cu-Ni alloy plasma was generated from laser blow-off of an “Advance” alloy cathode target, which was discharged with a drive current of 3 kA and \( B_z = 0.13 \) T. The “Advance” alloy target consisted of 45\% elemental Ni, and 55\% elemental Cu [48]. The collecting surface was placed successively at five axial locations, at distance of 0.16, 0.35, 0.53, 0.72 and 0.95 m downstream of the cathode target (see Fig. 2-1). With the anode mesh 0.16 m downstream from the cathode target, the five axial locations correspond to \( z = 0.19, 0.37, 0.56 \) and \( 0.79 \) m downstream from the anode mesh \( (z = 0 \text{ m}) \). At each location, a thin film was deposited by the plasma and analyzed by Electron Spectroscopy for Chemical Analysis (ESCA), from which both flux and separative profiles were produced. Comparison of the measured axial evolution of the separative profile of the plasma against our predictions is made in the following section.

Figures 2-2(a), 2-2(b) and 2-3 show the measured axial development of the measured flux profile. As can be seen from these profiles, the observed flux profile is not Gaussian. The flux profile shows a marked dip on axis, which becomes less pronounced with distance downstream of the plasma. As Geva et al. [13] describes, this dip is caused by preferential emission from the outer edges of the cylindrical cathode. Although the observed flux profile also exhibits some radial spreading as a function of axial position, this property is not included in this treatment.
Figure 2-2: Flux profile development of a Cu-Ni plasma, at axial positions $z = 0.19$ m and $z = 0.37$ m respectively from anode mesh. Extracted from “Element and isotope separation in a vacuum arc centrifuge”, by M. Geva, M. Krishnan and J. L. Hirshfield, [13]; $t$ represents thickness of deposited material per shot (nm). Overlayed on (b) is a numerically fitted flux density profile.
Figure 2-3: Plot of the measured separation (×, left axis) and flux (●, right axis) profiles of a Cu-Ni plasma at \( z = 0.79 \, \text{m} \) from the anode mesh. The solid line is a fit of the steady state separation profile against the experimental data (×). Measured data extracted from "Element and isotope separation in a vacuum arc centrifuge", by M. Geva, M. Krishnan and J. L. Hirshfield [13].

In this treatment an intermediate flux profile at \( z = 0.37 \, \text{m} \) is chosen to generate the generalized function \( f(y) \) [see Eq. (2.1)], which describes the radial flux distribution. The solid line fitted in Fig. 2-2(b) shows a function which matches the observed flux profile closely, given by,

\[
f(y) = \frac{1}{1 + \epsilon} \times \left[ \exp\left(-y^2\right) + \epsilon \exp\left(-\kappa y\right) \right]
\]

with parameters \( R = 0.026 \, \text{m} \), \( \epsilon = 0.001 \), and \( \kappa = 0.25 \). The \( y^2 = (r/R)^4 \) dependence in the exponent of the first term on the RHS of Eq. (2.22) gives a profile which falls off with radius more steeply than a Gaussian profile, in agreement with the experimental data. The second term on the RHS of Eq. (2.22) is a small additional Gaussian component which represents background gas outside the plasma column. Since Geva et al. [13] have measured elemental separation in this region, some material must be present. The addition of a background component also
prevents the generalized Hall parameter $\beta_1^*$ from growing too quickly with increasing $y$, thus assisting numerical stability. Providing that $f(y)$ decays to $O(10^{-4})$ at maximum radius, the values of the parameters $\epsilon$ and $\kappa$ do not have a significant impact on model predictions. Finally, the slight radial offset that appears in the flux density profile in Fig. 2-2(b) corresponds to a dislocation in the plasma column, which is not considered in subsequent analysis.

From the work by Geva et al. [13] the on-axis plasma ion density at $z = 0.37$ m can be estimated to be in the range $n_i(0) = 1.5 \times 10^{19}$ m$^{-3}$ to $n_i(0) = 2.5 \times 10^{19}$ m$^{-3}$. A mid-range value of $n_i(0) = 2.0 \times 10^{19}$ m$^{-3}$ is used in the following analysis. In this same work [13], the plasma temperature $T$ is inferred by fitting a predicted separation profile $\alpha(r)$ to the measured separation at the collector plate. Recently, Langmuir probe measurements in the VAC at the Brazilian National Space Research Institute [25] have shown that the ion temperature for a Cu plasma and Ni plasma, under similar device operating conditions, is 1.4 and 2 eV respectively. An ion temperature of $T = 1.5$ eV for a Cu-Ni plasma is used in this treatment.

Figure 2-3 plots the measured separation of Cu to Ni at $z = 0.79$ m, together with the predicted steady state separation profile for $T = 1.5$ eV and the measured flux density profile. Both the predicted and experimental separation are normalized to the separation on axis, as described in relation to Eq. (2.19). In the case of the experimental data, the average of data points within 10 mm of the axis is used for normalisation. Figure 2-3 shows that reasonable agreement is found between the measured data and the steady-state profile. Thus, if it is assumed that the plasma is close to steady-state at $z = 0.79$ m, then the temperature estimate of $T = 1.5$ eV is reasonable.

The on-axis average charge state of both the Cu and Ni ions was measured to be $Z = 3$ [13], using a magnetic sector mass spectrometer [49]. Geva et al.’s [13] steady state predictions show a dependence on the mass to charge ratio, in plasmas where species have differing charges. They also found some experimental evidence for preferential centrifuging of more highly charged ions in Al/Ti plasmas [13]. In the Cu-Ni plasmas under consideration, singly, doubly and triply charged ions are present, and preferential centrifuging may occur; leading to an average charge $Z$ which increases with radius. Although the model developed here is aimed at predicting isotope separation and thus does not take species with different charges into account, predictions have been made for two average charge values: $Z = 2$ and $Z = 3$. 

22
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Table 2.1: Parameters for Cu/Ni plasma.

Finally, based on Geva et al.’s experimental data [13]; the rotation frequency is $\omega_0 = 1.0 \times 10^5$ rad s$^{-1}$ and the bulk axial velocity is $v_{z0} = 6 \times 10^8$ m s$^{-1}$. These parameters now fully describe the plasma model, and allow calculation of the constants that scale Eq. (2.18). The separation figure of merit, $A$ is independent of $Z$, and evaluates to $A = 0.11$. For $Z = 2$, the on-axis generalized Hall parameter evaluates to $\beta^*_i(0) = 0.083$ and the axial scale length evaluates to $z_0 = 3.2$ m; and for $Z = 3$, $\beta^*_i(0) = 0.024$ and $z_0 = 14.8$ m. A summary of the plasma parameters used for the simulation is presented in Table 2.1.

2.5 Results and Analysis

A 3D surface depicting the spatial evolution of the separation profile, for the Cu-Ni plasma with $Z = 2$ described in Sec 2.4 is presented in Fig. 2-4. In conjunction with the surface plot, a contour plot of the separation profile is projected to the top plane. The profile depicts an initial rapid increase in separation around the characteristic radius, as centrifuging takes place. As $z$ increases, centrifuged material begins to accumulate near the edge of the plasma column. At large $z$, the peak in the separation profile moves radially outwards and increases in magnitude.
Figure 2-4: A 3-D surface of the calculated evolution of separation for the Cu-Ni plasma with \( Z = 2. \)

Figure 2-5 plots various calculated plasma parameters as a function of radius, at \( z = 0.37 \text{ m}. \) Figure 2-5(a) plots the mass density \( \rho(r) \) normalized to the on-axis value \( \rho(0) \), together with \( \beta_t^* \), the generalized Hall parameter [see Eq. (2.17)]. Figure 2-5(b) plots \( (1 - C)/(1 - C_0) \), the concentration of the heavier isotope normalized to its natural abundance; together with \( \delta_r \), the radial component of the difference in diffusion velocities, defined following Eq. (2.8). The expression for \( \delta_r \) comes from solving Eqs. (2.9) and (2.10)

\[
\delta_r = \frac{kT}{\mu v_i(0)} \times \frac{1}{f(r^2/R^2)(1 + \beta_t^2)} \times \left( \frac{2A r}{R^2} + \frac{1}{C(1 - C)} \frac{\partial C}{\partial r} \right) \tag{2.23}
\]

From Fig. 2-5 it is seen that near the axis, the radial gradient of the concentration \( C \) is low, and \( \beta_t^* \) is small. From Eq. (2.23), this implies that \( \delta_r \) is driven by centrifuging only [first term in brackets on the RHS of Eq. (2.23)], and increases linearly with radius. Towards the edge of the mass density profile, the concentration of the heavier ion increases and the resultant concentration gradient leads to the decrease in \( \delta_r \) Eq. (2.23) at \( r \approx 20 \text{ mm}. \) The increase in
$\delta_r$ at $r \approx 33$ mm occurs because the effects of centrifuging increase with radius, whereas the concentration gradient is roughly constant over the interval $27 < r < 37$ mm [see Eq. 2.23]. Beyond a radius of about $40$ mm, the concentration of the heavier isotope starts to decrease, leading to the larger than linear increase in $\delta_r$ with $r$ about $r = 40$ mm. Finally, the sharp decrease in $\delta_r$ at about $42$ mm corresponds to the onset of plasma freezing, where the density and the ion-ion collision rate fall rapidly, resulting in a rise in the generalized Hall parameter $\beta_i^*$. Where the plasma is frozen in Fig. 2-5, beyond a radius of about $50$ mm, $\delta_r$, the difference in the ion species diffusion velocities is small, and there is little change in the species concentrations from natural abundances. In this region, the low but non-zero values of $\delta_r$ result from the addition of a small amount of background plasma outside the main column, as described in Section 2.4.

The decrease in separation at large radii associated with plasma freezing is also visible in Fig. 2-5, with a sharp decrease in the separation beyond a radius of about $50$ mm. At smaller radii the separation tends towards the profile expected in the steady state, which is close to parabolic, however the ion-ion collision frequency is so low at outer radii that extremely large axial lengths would be required for the separation to approach steady state.

Comparison of the numerical and measured axial evolution of the separation profile is presented in Figs. 2-6(a) to 2-6(d), for the plasma conditions outlined in Sec. 2.4. Each figure plots the measured separation data points, and the calculated separation profile for charge states of $Z = 2$ and $Z = 3$. Figure 2-6(b) also shows the density profile used for calculations and Fig. 2-6(d) replots the steady state profile, given by Eq. (2.21).

The separation profiles for $Z = 2$ and $Z = 3$ differ due to the strong dependence of $\beta_i^*$ and $z_0$ on the ionisation state, $Z$ [refer to Eqs. (2.17) and (2.18)]. As the ionisation state is increased from $Z = 2$ to $Z = 3$, the Generalized Hall frequency decays from $\beta_i^*(0) = 0.083$ to 0.024, and the axial scaling factor increases from $z_0 = 3.2$ to 14.8 m. The reduction in $\beta_i^*(0)$ moves the freezing region to larger radii, resulting in a separation maximum at a larger radius. The increase in $z_0$ slows the axial evolution of the separation process.

Comparing the density profile in Fig. 2-6(b) with the separation profile, most separation is occurring near the tails of the flux profile, where the ion-ion collision frequency begins to
Figure 2-5: Physical parameters plotted across the plasma at an axial cut of $z = 0.37$ m. Figure (a) plots the flux profile $\rho(r)$ (left axis) together with generalised ion Hall parameter $\beta_i^*(r)$ (right axis). Figure (b) plots the heavy species concentration ratio $(1 - C)/(1 - C_0)$ (left axis), together with the radial component of the relative diffusion velocity $\delta_r$ (right axis).
Figure 2.6: Measured and predicted axial evolution of the separation profile for a Cu-Ni plasma. The four figures [(a) through (d)] correspond to distances downstream of the anode mesh of \( z = 0.19, 0.37, 0.56, 0.79 \) m respectively. In (b), the normalised flux profile used in the calculations is shown, and in (d) the steady state separation (\( \alpha \)) profile is presented. Experimental data is taken from “Element and isotope separation in a vacuum arc centrifuge”, by M. Geva, M. Krishnan and J. L. Hirshfield, [13].
rapidly decay and the plasma freezes. Comparing the predictions with the experimental data in Fig. 2-6, it can be seen that the predictions are close to the experimental data only for radii less than the predicted peak in the separation profile. In this region, comparing Figs. 2-6(a) and 2-6(b), the axial development is approximately correct, and there is at least some experimental evidence for a region of low separation near the axis [Figs. 2-6(a) to 2-6(c)]. However, there is a serious discrepancy between the theory and experiment at radii greater than the predicted separation maximum: the experimental data does not show any evidence of the expected reduction in separation due to plasma freezing.

At present, the reason for the discrepancy is not understood, however one possibility is that the extremely low density plasma outside the main plasma column may have properties which differ significantly from the bulk plasma. In this case, the present treatment would not be valid in that region. It seems possible that plasma contamination may occur in the outer region; hence the plasma temperature in the outer region may be lower than the uniform value of \( T = 1.5 \text{ eV} \) used throughout the analysis, and the bulk transport velocity of the plasma in the outer region may be different from the bulk transport velocity of the column.

A further complication relates to the possibility of a varying charge distribution with radius. The present analysis does not allow for separation of different charge states. As discussed in Sec. 2.4, charge state separation would tend to decrease the effective charge at outer radii [13].

Finally, it should be mentioned that in the case of separation of Cu and Zn alloys in VAC's, at least one measurement [50] suggests that the radial separation decreases with radius at the edge of the plasma column. Evans et al. [50] showed that the radial separation as measured by Proton-Induced X-ray Emission (PIXE) decreases at large radius, whereas the radial separation as measured by Energy Dispersive X-ray spectroscopy (EDX) technique continues to increase. They concluded that the EDX technique becomes unreliable when the deposit thickness falls below some minimum value.
2.6 Conclusions

A numerically determined solution to the spatial evolution of the separation, concentration and radial diffusion velocity has been presented. A key result of the model treatment is that the plasma should freeze at outer radii, where the ion-ion collision rate is dramatically reduced, and there should be little development of separation beyond the plasma column. For the available set of experimental data with which results were compared, no such effect was observed, and the model did not correctly predict the separation outside the plasma column. However, it should be noted that, for any future commercial application, it is the separation factor where a reasonable quantity of material is present which is of importance. The model does provide reasonable estimates of the axial development of separation within the plasma column, and thus provides a useful tool for evaluation of VAC designs.

In addition, the discrepancy between the physical phenomenon of plasma freezing and the experimental data pointed out in this work needs to be resolved. The resolution of this discrepancy may provide further insights into the functioning of vacuum arc centrifuges.

In the next chapter, the effects of electron-ion collisions in the rotation region of a VAC are described by an analytical steady-state calculation. This work builds upon an earlier numerical treatment by Yue and Simpson [36], in which the effects of electron-ion collisions were described for a particular VAC plasma. The advantage of the analytic approach is that the expressions found provide a more efficient method for investigating the effects of electron-ion collisions. Using the analytical solutions the change in steady-state separative performance due electron-ion collisions is discussed, trends with varying temperature, ion rotation frequency and ion axial streaming velocity identified, and conditions under which separative power is maximized suggested. Also, the non-uniformity in the axial magnetic field of a VAC caused by the azimuthal current is investigated.