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Optimized Q-cascade approach for stable cadmium isotope purification using Newton-Raphson and direct substitution methods

Mohamad Mahdi Shadman[a,*], Fatmeh Mansourzadeh[a], Valiyollah Ghazanfary[b], Younes Amini[a]

[a] *Nuclear Fuel Cycle Research School, Nuclear Science and Technology Research Institute, P.O. Box 11365-8486, Tehran, Iran*
[b] *Reactor and Nuclear Safety Research School, Nuclear Science and Technology Research Institute, Tehran, Iran*

HIGHLIGHTS

- The study applies the Q-cascade model, a novel approach, to optimize parameters for cadmium isotope separation.
- The research determines the optimal value of the M^* parameter to enhance the separation process.
- Researchers developed a combined Newton-Raphson and direct substitution algorithm.
- This is an innovative computational approach to solve the nonlinear equations in the cascade design.

ABSTRACT

In this study, the objective was to separate cadmium isotopes using the Q-cascade approach. The optimal value of the parameter M^* was determined by minimizing the function $\frac{\sum L\varepsilon_0^2}{2P}$ within the Q-cascade. To overcome computational challenges, the Newton-Raphson solver and direct substitution algorithm were employed to solve the system of nonlinear equations. The direct substitution method was used to provide a suitable initial guess for the Newton-Raphson method. Validation of the developed algorithms using stable cadmium isotopes showed that the value of M^* undergoes slight changes with variations in the target isotope concentration in the product and waste streams. For enriching Cd-106 in the product, M^* was approximately 108.5, corresponding to the average molar mass of Cd-106 and Cd-111. As the target isotope concentration in the product increases, the number of enrichment stages increases more steeply compared to the number of stripping stages. Similarly, increasing the Cd-116 enrichment in the waste leads to a larger increase in the number of stripping stages compared to enrichment stages.

KEYWORDS

Isotope separation
Newton-Raphson solver
Q-cascade model
Optimization
Direct Substitution Method

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1 Introduction

Cascade separation of stable isotopes is academically important for extracting and purifying specific isotopes in various scientific disciplines. It enables precise investigation of isotopic properties and behavior, advancing knowledge in fields like chemistry, physics, materials science, and environmental studies. Additionally, it has valuable applications in medical research, providing isotopically enriched compounds for radiopharmaceuticals and diagnostics. Cascade separation plays a crucial role in academic research by controlling isotopic compositions effectively. The study of continuous flow cascade models is useful for the rapid evaluation of cascade separation parameters and their optimization for isotope separation. Cascade separations of multicomponent mixtures, consisting of separation stages with overall enrichment factors ε_{ij} significantly less than one plays a crucial role in the separation of multicomponent systems. The design calculation of such separation cascades for specific concentration values of a target component in the product and waste streams involves determining the total number of separation stages in a cascade N, the number of stages in the enrichment sections S_P and in the stripping sections S_w, the distribution of flow rates in the cascade stages $L(s)$, and the determination of the product or waste stream values (Zeng et al., 2018). However, the set of obtained parameters should be consistent with the optimal values. In the theory of mixture separation, this issue is addressed through the concept of the Model Cascade of Continuous Profile (MCCP). Cascade parameters are obtained analytically using this tool. These calculations provide a basis for further calculations and optimization of square cascades (Borisevich et al., 2011; Song et al., 2010; Zeng et al., 2012). Various

*Corresponding author: mshademan@aeoi.org.ir

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models have been proposed for cascade design in multi-component isotope separation (Zeng et al., 2018, 2012, 2011, 2016; Zhang et al., 2015). The Q-cascade model has been proposed for rapid evaluation of cascade parameters and their optimization in isotope separation. While the Q-cascade model is valid for cascades with low separation factors, it is also applicable to cascades with high separation factors (Zhang et al., 2015; Sulaberidze and Borisevich, 2001). Borisevich et al. investigated the separation of cadmium isotopes Cd-114 and Cd-116 using the Q-cascade and determined the necessary number of stages to minimize interstage flows (Borisevich et al., 2011).

In this study, the equations and solution methods of the MCCP model for stable cadmium isotopes are presented using the Newton-Raphson and direct substitution methods. Stable cadmium isotopes, particularly Cd-111 and Cd-110, have various applications in industries and research. For example, Cd-111 is used as a radioactive source in hydroponic and liquid culture techniques. Additionally, Cd-110 is used as a radioactive source in medical diagnosis (Zhong et al., 2020).

In the second part of this paper, the governing equations for the Q-cascade are examined. Subsequently, the methods for solving these equations are discussed, employing both the Newton-Raphson algorithm and the direct substitution algorithm. Finally, the results for the stable isotopes of cadmium are presented.

2 Modeling

2.1 Governing Equations

The main computational idea in the Model Cascade of Continuous Profile (MCCP) is to replace the component concentration, $C_i(s)$, in the separating mixture with certain characteristic functions $\varphi_i(s)$, which leads to the mass transfer equation in a cascade in the following form:

$$\begin{aligned}\varphi_i(s) + \sum_{j=1}^{m} \frac{2PC_{j,P}}{L(s)} \int_0^{S_P} \varphi_i(t)\exp\big(\varepsilon_{ij}(S-t)\big)\mathrm{d}t \\ = \sum_{j=1}^{m} \frac{C_{i,F}L_F}{L(s)}\exp\big(\varepsilon_{ij}S\big) \qquad i = 1, ..., m\end{aligned} \tag{1}$$

where P is the product stream flow of the cascade, $C_{j,P}$ is the concentration of component j in the product stream, $L(s)$ is the flow rate in stage s, $C_{i,F}$ is the concentration of component i in the feed stream, F is the feed stream flow of the cascade, and L_F is the input feed flow rate to the feed stage (Zeng et al., 2011, 2016). One of the mathematical functions that can be assigned to the characteristic function $\varphi_i(s)$ is given by (Borisevich et al., 2011):

$$\varphi_i(l) = \exp(Q_i l) \tag{2}$$

where, Q_i and Q_j parameters are related as:

$$Q_i - Q_j = \varepsilon_{ij} \tag{3}$$

The general separation parameter ε_{ij} (equivalent to the logarithmic factor α_{ij}, which is the separation factor) is defined as:

$$\varepsilon_{ij} = \varepsilon_0(M_j - M_i) \tag{4}$$

where M_i and M_j are the molar mass of components i and j, respectively. ε_0 is the separation factor of unit mass difference. The parameter Q_i for isotope i can be calculated from (Borisevich et al., 2011):

$$Q_i = \varepsilon_0(M^* - M_i) \tag{5}$$

where M^* is a hypothetical mass number, and separations are measured based on it. Components with molar mass values less than M^* ($M_i < M^*$) are in the light stream, while components with mass numbers greater than M^* ($M_i > M^*$) are in the heavy stream. By establishing the overall mass balance, partial mass balance, and the condition $\sum_{j=1}^{m} C_j = 1$ the distribution of component concentrations in the product and waste streams can be obtained as (Borisevich et al., 2011):

$$C_{i,P} = \frac{\dfrac{1-\exp(Q_i S_w)}{\exp(-Q_j S_P) - \exp(Q_j S_w)} C_{i,F}}{\displaystyle\sum_{j=1}^{N_c} \frac{1-\exp(Q_j S_w)}{\exp(-Q_j S_P) - \exp(Q_j S_w)} C_{j,F}} \tag{6}$$

$$C_{i,w} = \frac{\dfrac{\exp(-Q_i S_P) - 1}{\exp(-Q_j S_P) - \exp(Q_j S_w)} C_{i,F}}{\displaystyle\sum_{j=1}^{N_c} \frac{\exp(-Q_i S_P) - 1}{\exp(-Q_j S_P) - \exp(Q_j S_w)} C_{j,F}} \tag{7}$$

The ratio of product flow to feed flow and the ratio of waste flow to feed flow is obtained as:

$$\frac{P}{F} = \sum_{j=1}^{m} \left(\frac{\exp(Q_j S_w) - 1}{\exp(Q_j S_w) - \exp(-Q_j S_P)} C_{j,F} \right) \tag{8}$$

$$\frac{w}{F} = \sum_{j=1}^{N_c} \left(\frac{1 - \exp(-Q_j S_P)}{\exp(Q_j S_w) - \exp(-Q_j S_P)} C_{j,F} \right) \tag{9}$$

The total interstage flow rate is also determined using:

$$\begin{aligned}\sum L = 2 \sum_{i=1}^{m} \Big\{ & \frac{PC_{i,P}[\exp(-Q_i S_P) - 1]}{Q_i^2} \\ & + \frac{wC_{i,w}[\exp(Q_i S_w) - 1]}{Q_i^2} \\ & + \frac{PC_{i,P}S_P - wC_{i,w}S_w}{Q_i} \Big\}\end{aligned} \tag{10}$$

2.2 Solution Algorithm

In this study, the goal is to design an optimal cascade using the Q model. To find the optimal value of the parameter M^*, the function $\dfrac{\sum L\varepsilon_0^2}{2P}$ is calculated for a specified range of the target isotope in the product and waste streams of the Q-cascade. The term $\sum L$ represents the interstage flows, while the parameter ε_0 is a constant value ranging between zero and one. It is more important, in terms of cascade separation objectives, to have lower interstage flows and a higher production yield. Therefore, the mentioned objective function represents a relative interstage flow that needs to be minimized. The solution algorithm consists of the following steps:

a) The target isotope (n) is selected, and design values are assigned to $C_{n,P}$ and $C_{n,w}$.

b) A value for the unit mass difference separation factor, ε_0, is chosen.

c) If the total number of isotopes is m, the parameter M^* is selected within the bounds of M_1 to M_m.

d) Considering the known values of $C_{n,P}$ and $C_{n,w}$, Eqs. (6) and (7) are formulated for the target isotope n, and the values of S_P and S_w are solved. The nature of Eqs. (6) and (7) is nonlinear, and there is no analytical solution for them. It should also be noted that if M^* is exactly equal to one of the components M_1 to M_m, the solutions of the problem become divergent. To overcome these computational difficulties, two methods are proposed to find the values of S_P and S_w.

2.2.1 First Algorithm

In the first algorithm, the Newton-Raphson method is proposed to solve the system of two nonlinear equations. This method relies on developing an equation based on an approximation of the root of the original equation and converges to the root using successive approximations. The system of two nonlinear equations and successive solution is as follows:

$$F_1(S_P,S_w)=\frac{C_{n,P}-\dfrac{1-\exp(Q_nS_w)}{\exp(-Q_nS_P)-\exp(Q_nS_w)}C_{n,F}}{\displaystyle\sum_{j=1}^{N_c}\frac{1-\exp(Q_jS_w)}{\exp(-Q_jS_P)-\exp(Q_jS_w)}C_{j,F}} \tag{11}$$

$$F_2(S_P,S_w)=\frac{C_{n,w}-\dfrac{\exp(-Q_nS_P)-1}{\exp(-Q_nS_P)-\exp(Q_nS_w)}C_{n,F}}{\displaystyle\sum_{j=1}^{N_c}\frac{\exp(-Q_iS_P)-1}{\exp(-Q_jS_P)-\exp(Q_jS_w)}C_{j,F}} \tag{12}$$

$$\begin{bmatrix}S_P\\S_w\end{bmatrix}^{(k+1)}=\begin{bmatrix}S_P\\S_w\end{bmatrix}^{(k)}-\begin{bmatrix}\dfrac{\partial F_1}{\partial S_P}&\dfrac{\partial F_1}{\partial S_w}\\[2mm]\dfrac{\partial F_2}{\partial S_P}&\dfrac{\partial F_2}{\partial S_w}\end{bmatrix}^{-1}\times\begin{bmatrix}F_1(S_P,S_w)\\F_2(S_P,S_w)\end{bmatrix}^{(k)} \tag{13}$$

where k represents the number of computational iterations. It should be noted that if the solution diverges for a specific value of M^*, the range of M^* needs to be adjusted. Various solvers for Eqs. (11) and (12) were examined, including the Solve function in MATLAB, which resulted in divergent solutions. The Newton-Raphson method is an iterative technique used to find the roots of a function. This method is particularly effective due to its high convergence rate, meaning that it typically requires fewer iterations to converge to the root of the equation compared to other root-finding methods.

The high convergence rate of the Newton-Raphson method is a consequence of the fact that it is based on the Taylor series approximation of the target function. By using the first-order Taylor series approximation, the method assumes that the function behaves like a linear function in the vicinity of the initial guess. This allows the method to quickly converge to the root, as the linear approximation becomes more accurate as the iterative process progresses. The key steps of the Newton-Raphson method are as follows:

1. Start with an initial guess for the root of the equation.
2. Calculate the value of the function and its derivative at the current estimate.
3. Use the function value and its derivative to compute the next estimate of the root using the Newton-Raphson formula.
4. Repeat steps 2 and 3 until the desired level of accuracy is achieved.

The Newton-Raphson method is particularly effective for functions that are well-behaved and have a continuous derivative in the vicinity of the root. This method can converge quadratically, meaning that the number of correct digits in the solution roughly doubles with each iteration. This rapid convergence makes the Newton-Raphson method a popular choice for solving a wide range of root-finding problems in various fields, such as engineering, physics, and mathematics. By applying this method to different isotopes, it can be observed that it has limited dependence on the initial guess for some isotopes (Shadman and Mansourzadeh, 2023). Generally, it is necessary for the user to provide an appropriate initial guess to solve the problem, considering the non-linearity of Eqs. (11) and (12). If the desired method diverges, this issue can be examined from two perspectives. First, the problem may not have a solution for certain design values of $C_{n,P}$, $C_{n,w}$, and M^*. Second, the initial starting point entered as the initial guess for the Newton-Raphson method may have caused the divergence. Therefore, before using the Newton-Raphson method, it would be beneficial to utilize a direct substitution method.

2.2.2 Second Algorithm

In the direct substitution method, a range is initially considered for searching the solution for the values of S_P and S_w. All values within this range are substituted into Eqs. (11) and (12). If the following condition is satisfied:

$$\begin{aligned}|F_1(S_P,S_w)|<\varepsilon\\|F_2(S_P,S_w)|<\varepsilon\end{aligned} \tag{14}$$

where ε is a small tolerance value, the solutions for S_P and S_w are recorded, and the search continues. This process is performed with different values of , such as 0.1, 0.01, and 0.001, covering all the examined regions. If all the examined regions do not yield any solutions, it means that the cascade design problem does not have a solution for the given input values of $C_{n,P}$, $C_{n,w}$, and M^*, and using the Newton-Raphson method and other methods would be pointless.

Table 1: The natural abundance ratio of stable isotopes of cadmium (Borisevich et al., 2011).

Isotopes	Cd-106	Cd-108	Cd-110	Cd-111	Cd-112	Cd-113	Cd-114	Cd-116
Molar Percent	1.25	0.89	12.49	12.80	24.13	12.22	28.73	7.49

Table 2: Optimal design values for the target isotope Cd-116. The columns *Ref.* refer to the work presented by Borisevich et al. (Borisevich et al., 2011).

		$\frac{\sum L\varepsilon_0^2}{2P}$		M^*		S_P			S_w	
$C_{n,w}$ (%)	Ref.	First Algorithm	Ref.	First Algorithm	Ref.	First Algorithm	Validation Error	Ref.	First Algorithm	Validation Error
40	1.20248	1.2024	112.593	112.6001	18.7481	18.7986	0.27	6.26228	6.2395	0.36
60	2.43388	2.43393	113.057	113.0501	25.9765	25.8489	0.49	21.3022	21.4228	0.57
70	4.04923	4.05117	113.320	113.3001	35.2939	34.4929	2.27	37.0640	38.0725	2.72

Table 3: The optimal variable values Cd-106 from the second algorithm for $M^* = 109.2$ and $C_{n,p} = 60\%$.

S_P	34	41	34	39
S_w	2	6	2	5
$C_{n,p}(\%)$	0.60125	0.59776	0.60125	0.59708
$C_{n,w}(\%)$	0.00720	0.00237	0.00720	0.00313
Error based on $C_{n,p}(\%)$	0.20844	0.37338	0.20844	0.48713
Error based on $C_{n,w}(\%)$	44.08226	52.60044	44.08226	37.36365

e) With the values of S_P and S_w determined, the values of $C_{i,P}$ and $C_{i,w}$ are calculated using Eqs. (6) and (7).

f) The value of $\frac{\sum L\varepsilon_0^2}{2P}$ is calculated using Eqs. (8), (9), and (10).

2.3 Model Parameters

Table 1 presents the natural abundance of stable isotopes of cadmium.

3 Results and discussion

3.1 Validation

To validate the developed code, the Ref. (Borisevich et al., 2011) was used. The molar concentration of the target component, Cd-116, in the waste stream was set to 40%, 60%, and 70%, while in the product stream it was set to 5%. The factor ε_0 (unit mass difference separation factor) is considered to be 0.0875. According to Table 2, the predicted values using the Newton-Raphson method have an error of less than 3% compared to the Ref. (Borisevich et al., 2011). However, as the molar percentage of the target isotope in the waste stream increases from 40% to 70%, the error in the trend with respect to S_P and S_w increases from 0.27% to 2.72%.

3.2 Optimization of Cd-106

The results of designing a cascade for the target isotope Cd-106 are presented below. In the current modeling, the concentration of the target component, Cd-106, in the waste stream was set to 0.5%, and the concentration of the target component, Cd-106, in the product stream was set to 20%, 40%, and 60%. The graph of the parameter $\frac{\sum L\varepsilon_0^2}{2P}$ versus the parameter M^* for different values of the concentration of Cd-106 in the product stream is shown in Fig. 1. This figure demonstrates the optimization process using the Newton-Raphson algorithm, as explained above. It can be observed that the Newton-Raphson algorithm has acceptable convergence. Fig. 1-c shows the graph divided into two different regions. the initial estimation for both S_P and S_w in the black region ($M^* < 109.2$) was set to 10. The continuation of solving for M^* greater than 109.2 will not converge with this initial estimation. For the blue region, ($M^* > 109.2$, the direct substitution method was used, and according to Table 3, the initial estimation for S_P and S_w was chosen as 41 and 6, respectively.

In Table 4, the optimal variable values for the target isotope Cd-106 from the first algorithm are demonstrated. It can be seen that M^* depends significantly on the target isotope. The value of M^* changes slightly with variations in the percentage of the target isotope in the product stream and is approximately 108.5, which is the average molar mass of Cd-106 and Cd-111, indicating the condition of no mixing for these two isotopes. As the concentration of the target isotope in the product stream increases, the number of enrichment stages increases compared to the number of stripping stages with a sharper slope.

Table 4: Optimal values for the Cd-106 from the first algorithm.

$C_{n,p}(\%)$	$\frac{\sum L\varepsilon_0^2}{2P}$	M^*	S_P	S_w
20	2.352	108.901	12.541	3.663
40	5.508	108.701	18.3128	3.9255
60	9.003	108.501	23.9007	4.2302

Table 5: The optimal variable values Cd-106 from the second algorithm for $M^* = 106.1$ and $C_{n,p} = 20\%$.

S_P	7	7	8	7
S_w	10	11	10	3
$C_{n,p}(\%)$	0.19907	0.20411	0.19661	0.19907
$C_{n,w}(\%)$	0.00512	0.00481	0.00913	0.00512
Error based on $C_{n,p}(\%)$	0.46413	2.05393	1.69740	0.46413
Error based on $C_{n,w}(\%)$	2.35502	3.71378	82.69433	2.35502

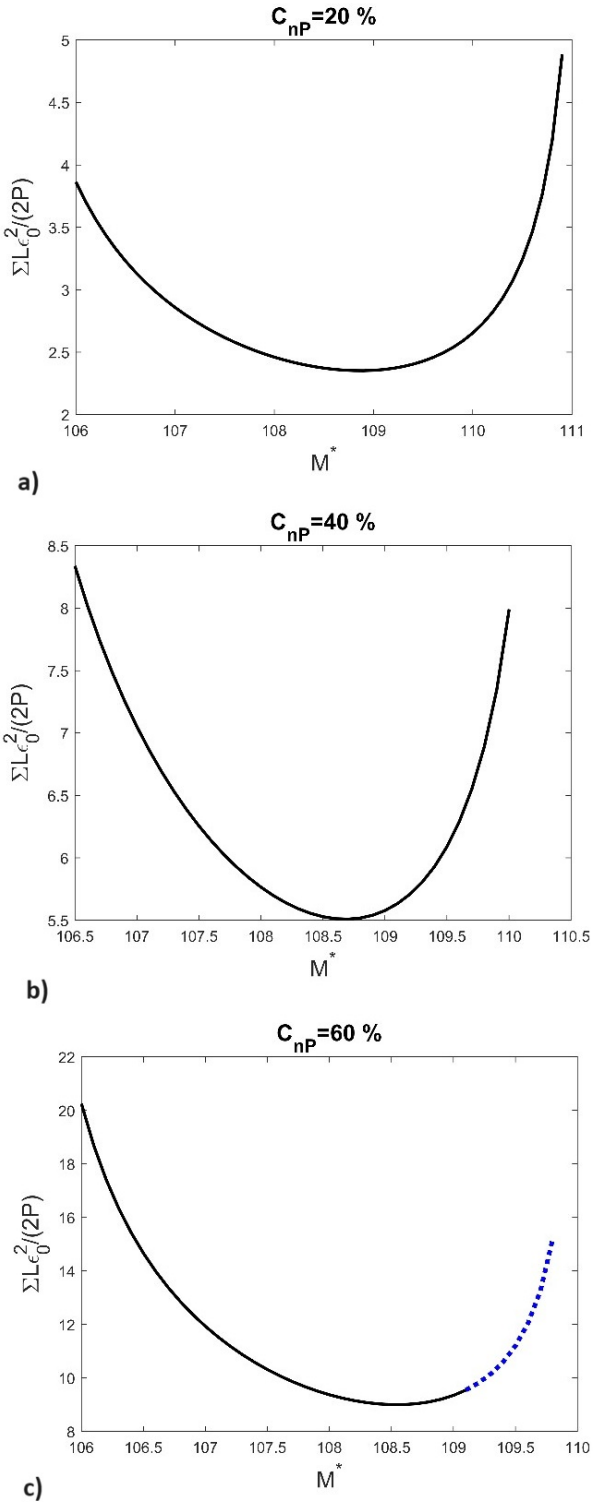

Figure 1: Optimization of M^* for different concentrations of the Cd-106 in the product stream.

Table 5 presents the optimal values of the S_P and S_w from the direct substitution algorithm. If the value of in Eq. (14) is changed, the number of solutions will vary. Using this table, initial estimation values and the possible solution region can be obtained. For example, according to Fig. 1-a and the direct substitution algorithm, designing a cascade for M^* equal to 111 will not be feasible.

3.3 Optimization of Cd-116

The results of modeling for the target isotope Cd-116 in the waste stream are presented below. In the current modeling, the concentration of the target component Cd-116 in the product stream was set to 1%, and the concentration of the target component Cd-116 in the waste stream was set to 20%, 40%, and 60%. The graph of the parameter value $\frac{\sum L\varepsilon_0^2}{2P}$ versus the parameter M^* for different values of the concentration of the Cd-116 target component in the waste stream are shown in Fig. 2.

Table 6 presents the optimal variable values for the target isotope Cd-116 from the first algorithm.

Table 6: Optimal values for the Cd-116 from the first algorithm.

$C_{n,p}(\%)$	$\frac{\sum L\varepsilon_0^2}{2P}$	M^*	S_P	S_w
20	0.559	1114.201	13.6397	8.2848
40	0.817	114.601	16.9676	16.9367
60	0.999	114.701	18.3434	26.3388

4 Conclusions

The Q-cascade model has been proposed for the rapid evaluation of cascade parameters and their optimization in isotope separation. The design calculation of such separation cascades for specific concentration values of a target component in the product and waste streams involves determining the total number of separation stages in a cascade (N), the number of stages in the enrichment sections (S_P), the number of stages in the strip sections (S_w), the flow distribution in cascade stages, as well as determining the product or waste stream values. In the context of isotope separation using the Q-cascade algorithm, the parameter M^* is defined as a hypothetical mass number, and separations are quantified based on it. Isotopes with a molecular weight less than M^* ($M_i < M^*$) are separated in the light stream, while isotopes with a molecular weight greater than M^* ($M_i > M^*$) are separated in the heavy stream of the cascade. In this study the objective is the separation of cadmium isotopes using the Q-cascade.

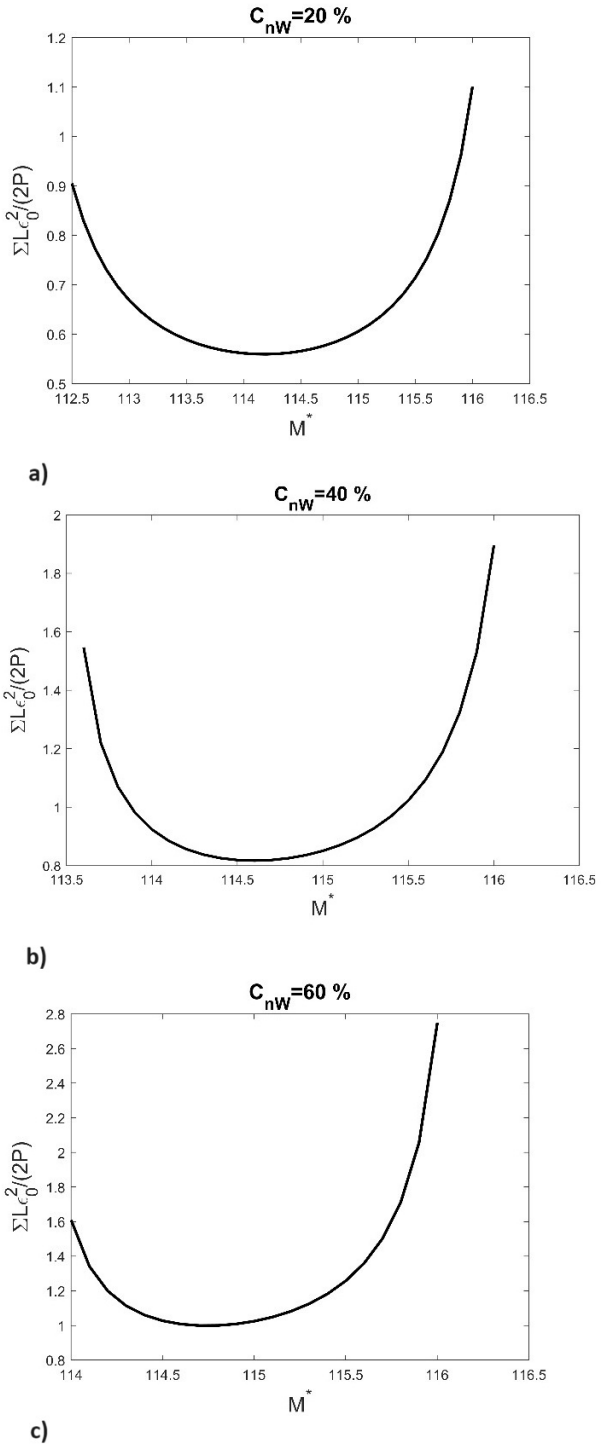

Figure 2: Optimization of M^* for different concentrations of the Cd-116 in the waste stream.

To find the optimal value of the parameter M^*, the function $\frac{\sum L\varepsilon_0^2}{2P}$ is calculated for specific values of a target isotope in the product and waste streams. To overcome computational difficulties, the Newton-Raphson algorithm and the direct substitution algorithm have been proposed for solving the system of two nonlinear equations. Using this approach, different possible solution sets are examined for values of $C_{n,P}$, $C_{n,w}$, and M^*, and a suitable initial guess is obtained for the Newton-Raphson method.

To validate the developed algorithms, stable cadmium isotope modeling is compared with a parameter value of ε_0 equal to 0.0875. The validation results show an error of less than 3%. As the molar percentage of the target isotope in the waste stream increases from 40% to 70%, the validation error increases from 0.27% to 2.72% for S_P and S_w. The results indicate that the value of M^* changes slightly with variations in the percentage of the target isotope in the product and waste streams. For the enrichment of Cd-106 in the product, the value of M^* is approximately 108.5, which is the average molar mass of Cd-106 and Cd-111, indicating the condition of no mixing for these two isotopes. As the target isotope concentration in the product increases, the number of enrichment stages increases with a higher slope compared to the number of stripping stages, with respective increases of 90% and 13%. Furthermore, as the enrichment of Cd-116 isotope in the waste stream increases from 20% to 60%, the number of enrichment stages increases by 34%, and the number of stripping stages increases by 218%.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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