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Comparative Assessment of Lithium Isotope Separation Processes for Tritium Breeding Blanket Systems in Fusion Reactors

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Abstract

The deuterium–tritium (D–T) fusion reaction is considered a promising approach for future commercial fusion power generation. However, the long-term operation of D–T fusion reactors depends on sustainable tritium production in tritium breeding blanket, where lithium-6 (Li-6) serves as the primary tritium breeding material. Since naturally occurring lithium contains only approximately 7.5% lithium-6, efficient ^{6}Li isotope enrichment is essential to improve tritium breeding performance. This study presents a comparative assessment of four major lithium isotope separation processes, namely Chemical Exchange (COLEX), Electrochemical process, Distillation process, and Ion-Exchange process. However, a systematic comparison of these processes is essential to identify their relative practical process feasibility and separation performance for fusion applications. The processes are evaluated based on isotope separation mechanism, scalability, energy requirement, engineering feasibility, economic feasibility, environmental impacts, separation performance and minimum required theoretical stages. For each process, a representative separation factor (α) was selected considering reported separation factors. Next, a computational simulation was performed using Fenske equation based on the representative separation factor (α), natural composition (7.5% ^{6}Li) and target enrichment (60% ^{6}Li) for the four processes. The minimum theoretical stage requirements were determined as 51, 55, 75, and 99 for distillation, COLEX, electrochemical and ion-exchange processes, respectively. The evaluated characteristics and minimum stage numbers are presented in a comparison table. The study provides a systematic assessment of lithium isotope separation methods that can contribute to the development of sustainable fusion fuel-cycle infrastructure.

Keywords: Lithium isotope separation; Lithium-6 enrichment; Tritium breeding blanket.

1. Introduction

For future fusion energy, deuterium-tritium (D-T) fusion is considered a leading approach due to its relatively favorable fusion reaction characteristics. Since there is limited availability of tritium, it requires the tritium breeding in fusion reactor blanket through neutron-induced reactions with lithium-6 enabling a sustainable D-T fuel cycle[1]. The tritium production takes place in the fusion breeding blanket according to the reaction:

$$^{6}_{3}Li + {}^{1}_{0}n \rightarrow {}^{4}_{2}He + {}^{3}_{1}H + 4.8MeV \text{ [1]}$$

In this reaction, ^{6}Li isotope accept one neutron and breeds tritium producing helium. For this tritium breeding ^{6}Li isotope is obtained through ^{6}Li isotope separation from its natural resource. The natural composition of ^{6}Li is about 7.5% [2]. In some fusion tritium breeding, 60% enriched ^{6}Li is used [3] and so in our computational simulation, minimum theoretical stage requirement is calculated to enrich ^{6}Li from 7.5% to 60%.

Although various lithium isotope separation processes have been investigated individually, a comparative evaluation of their separation characteristics, practical feasibility, and minimum theoretical stage requirements under a common enrichment target remains limited. In the comparative study, four processes such as distillation process, COLEX process, electrochemical process and ion-exchange process have been presented systematically.

For the process comparison, several process characteristics: isotope separation mechanism, scalability, energy requirement, engineering feasibility, economic feasibility, environmental impacts, separation factors are discussed with appropriate references, and minimum required theoretical stages are determined with python simulation and included in process comparison. The comparative characteristics and minimum required stages are presented in a comparison table and discussed.

This comparative study presents COLEX as a established process in lithium isotope separation, however it has health and environmental issues as it uses toxic mercury in lithium-amalgam phase. Ion-exchange process has moderate environmental impact and its separation performance is relatively low. Both electrochemical and distillation processes have low environmental impacts and have engineering feasibility, while the representative separation factor (α) of distillation process is greatest among the four, and the distillation process can be a promising and balanced process for lithium isotope separation.

2. Methodology

For this comparative study, the seven characteristics and minimum theoretical stage requirement are considered:

1. Separation mechanism
2. Scalability
3. Energy requirement

4. Engineering feasibility
5. Economic feasibility
6. Environmental Impacts
7. Representative separation factor
8. Minimum theoretical stage requirement

Based on the reported separation factors of each process, a representative separation factor (α) is selected to represent the separation performance (α) of each process. In minimum theoretical stage determination, all separation processes are considered under ideal stage conditions, except for the separation factor, which is selected as a representative value based on reported literature data for each process.

The seven characteristics are discussed in detail with appropriate references for each process, and the minimum theoretical stage requirements are determined with computational simulation with python coding. Next, the comparison is presented in a table.

The minimum stage requirements are estimated according to the minimum stage requirement equation derived by M. R. Fenske, the minimum number of theoretical stages in respect of product isotope,

$$N_P = \frac{ln\{\frac{x_P(1-x_F)}{x_F(1-x_P)}\}}{ln(\alpha)} \quad \text{.....(1) [4].}$$

For the convenience in computational coding, we transformed Equation (1) to small letter subscripted version,

$$N = \frac{ln\{\frac{x_p(1-x_f)}{x_f(1-x_p)}\}}{ln(\alpha)} \quad \text{.....(2)}$$

Where, N = minimum number of theoretical stages

x_p = ^{6}Li fraction in product

x_f = ^{6}Li fraction in feed

α = Separation factor.

A representative target enrichment of 60% ^{6}Li was selected for the comparative stage calculation, as ^{6}Li enrichment in the range of 30-60% has been considered for several solid breeder blanket concepts [5]. For each process to enrich ^{6}Li from its natural composition (approximately 7.5%) to 60%, the minimum stage requirements are calculated with computational simulation in Google Colab. The computational Colab notebook is provided in **Supplementary Code LHR2** in Section 10.

3. Distillation Process

In general concept, molecular distillation is a physical isotope separation process that exploits the difference in vaporization behavior between ^{6}Li and ^{7}Li isotopes under high-vacuum conditions. The separation process enables preferential enrichment of the lighter ^{6}Li isotope in the vapor phase, of the heavier ^{7}Li tends to remain relatively enriched in the liquid phase [6].

Mechanism: (Vapor-liquid equilibrium)

Molecular distillation generally is used to separate lithium isotopes based on the isotopic mass difference in the vaporization behavior of ^{6}Li and ^{7}Li in vapor-liquid equilibrium. In this separation process, metallic lithium is heated by a heat source under high-vacuum conditions in a molecular still, where the short mean free path of the vapor allows lithium atoms to travel from the evaporating surface toward a nearby condenser [6]. Since ^{6}Li is lighter than ^{7}Li, ^{6}Li enrichment occurs in vapor phase, while remaining ^{7}Li enrichment occurs in liquid phase. The vapor condensed and enriched ^{6}Li can then be collected separately from ^{7}Li enriched liquid phase, and further processing through successive stages leads to increase isotope enrichment to a desired percentage. Lithium-isotope separation using horizontal molecular stills, demonstrated by Trauger et al., in which lithium evaporated from a heated plate and condensed on a nearby surface, with multistage configurations investigated for improving isotope separation [7]. The effectiveness of the distillation process depends strongly on the temperatures of evaporation and condensation, vacuum conditions, and the distance between the evaporating and condensing surfaces, since collisions of vapor atoms can reduce the isotope separation effect [6].

Scalability:(Scalable)

The scalability of the distillation process is primarily hampered by the need to maintain effective isotope separation across a multistage cascade at high operating temperatures and under high vacuum conditions. This process requires multistage cascade arrangement and high temperature ($> 500℃$) to achieve desired ^{6}Li concentration for fusion technology [7]. Trauger et al. conducted an experimental evaluations at the Oak Ridge Gaseous Diffusion Plant demonstrating the mechanical feasibility of continuous spiral-staged and horizontal multistage stills for liquid metals [7]. For scaling up to fusion blanket target requires precise vacuum-tight containment, extensive column infrastructure [6]. Hence, molecular distillation process is scalable for industrial purpose for lithium isotope separation.

Energy requirements: (Very High)

Since the distillation process relies on the continuous vaporization of liquid metallic lithium at high operating temperatures ($> 500℃$) under high vacuum, the energy requirement of molecular distillation is substantially very high. In spite of having relatively greater representative separation factor ($\alpha_{Distillation} = 1.06$) than most conventional processes, a large number of

cascade stages is required, which increases the thermal energy demand for repeated evaporation and condensation cycles across the system. Maintaining the high-vacuum conditions and required evaporation rates across continuous multistage stills demands substantial thermal and mechanical power input required for continuous multistage operation demonstrated by Trauger et al. [7]. Although, recent engineering findings suggest that the integration of the thermal duty of the distillation cascade directly with high-temperature thermal energy sources could mitigate the severe electrical power requirement for liquid-metal vaporization, the absolute thermal footprint of the distillation cascade remains exceptionally very high [6].

Engineering Feasibility: (Feasible)

Although large scale continuous operation presents significant technical challenges, the engineering feasibility of molecular distillation for lithium isotope separation has been demonstrated experimentally. Trauger el al. established the mechanical feasibility of continuous liquid-metal distillation using horizontal multistage stills under high vacuum conditions [7]. However, operating these systems requires addressing complex engineering demands, including precise thermal control to maintain liquid lithium above 500℃, high-vacuum sealing, and prevention of liquid-phase mass transfer resistance [7]. Moreover, multistage molecular stills require continuous liquid condensate transport and controlled liquid flow between stages. While modern concepts aim to streamline heat integration and continuous flow designs, the combination of extensive column infrastructure and high-temperature vacuum containment means that while molecular distillation is technically feasible, implementing it at industrial fusion-blanket scales requires substantial mechanical and thermal optimization [6].

Economic feasibility:(feasible)

In spite of having the relatively greater separation factor of molecular distillation process, the process requires multi-stage cascades with high-temperature structural materials and extensive vacuum equipments to achieve high enrichment level, leading to high fixed capital scaling [6]. Early experimental studies by Trauger et al. focused the need for precise still construction and materials capable of withstanding high temperature liquid metal operation [7] while subsequent liquid-metal distillation scaling studies for potassium isotopes by McGill et al. demonstrated engineering requirement for multistage operation [8]. contributing to significant upfront infrastructure costs. Although the initial cost is high, the distillation process has potential for economic feasibility at industrial scale due to its relatively high separation factor and multistage cascade system.

Environmental Impact: (Low)

Molecular distillation process presents an definitely favorable environmental profile compared with conventional chemical separation processes. The process, operating strictly through

physical phase changes under high vacuum, avoids toxic amalgam (Hg) media typical of industrial column exchange techniques [7]. Consequently, distillation process can avoid hazardous effluent generation, making it an sustainable environmental pathway providing high-temperature thermal inputs via clean energy sources [6].

Justification For Representative separation factor for distillation process ($\alpha_{Distillation} = 1.06$):

Molecular distillation depends on the differential free-evaporation rates of lithium isotopes under high vacuum. The ideal single-stage separation factor is theoretically governed by the square root of their atomic mass ratio, $\frac{M_{^7Li}}{M_{^6Li}}$;

$$\alpha_{Ideal} = \sqrt{\frac{M_{^7Li}}{M_{^6Li}}} = \sqrt{\frac{7.016}{6.015}} \approx 1.08 \text{ [6]}$$

Malyusov et al. reported the theoretical separation factor, $\alpha = 1.08 \pm 0.02$ at 500℃ temperature in an experimental single-stage evaporator [9]. However, the effective stage performance is reduced in intermolecular vapor collisions, engineering-scale distillation systems and multi-stage cascades, liquid-phase mass transfer resistance, and condensate re-evaporation. Trauger et al. conducted some trials on multi-stage high-vacuum distillation at the Oak Ridge Gaseous Diffusion Plant yielded operational stage separation factors of 1.052–1.064 across temperatures of $535 - 627℃$ [7]. Therefore, for this comparative study the separation factor, $\alpha_{Distillation} = 1.06$ is adopted as representative value of the distillation process.

4. COLEX Process

The column exchange (COLEX) process is one of the most well-known and historically significant methods for lithium isotope separation. To achieve isotope separation between a liquid lithium-amalgam phase and an aqeueous lithium containing solution phase, the COLEX process utilizes the selective partitioning of lithium isotopes [10].

Mechanism: (Chemical Exchange)

Fundamentally it relies on a chemical exchange reaction between lithium dissolved in an aqueous phase (such as LiOH aqueous solution) and lithium present in a liquid metal amalgam (Li-Hg). The conventional COLEX process is established based on equilibrium isotope exchange between lithium amalgam and a lithium-containing solution. In the classical amalgam-solution system, isotope exchange occurs at the liquid-liquid interface between lithium amalgam and lithium-containing solution. During equilibrium, ^{6}Li preferentially migrates into the amalgam

phase, whereas ^{7}Li remains relatively enriched in the solution [10, 11]. As the two phases interact, isotopic exchange occurs according to the equilibrium reaction:

^{7}Li(Hg) + ^{6}LiX(solution) ⇌ ^{6}Li(Hg) + ^{7}LiX(solution) [11]

where, X represents the counter-ion, such as OH^{-}, Cl^{-} or other suitable anion.

The process results in preferential enrichment of one isotope (Li-7) in the aqueous phase and the other (Li-6) in the amalgam phase. The Li-7 isotopes from Li-Hg liquid metal phase show a tendency to migrate into aqueous LiOH solution; on the other hand, Li-6 isotopes from LiOH aqueous solution phase demonstrate a tendency to partition into Li-Hg liquid metal phase.

Scalability: (Established)

For industrial scale lithium isotope production, historically the COLEX process has been established, and its minimal production cost is the main advantage for choosing this process [12]. In industrial counter-current production set up in COLEX process, aqueous LiOH flows upward and lithium-amalgam (Li-Hg) flows downward enriching ^{6}Li in amalgam phase and ^{7}Li in aqueous phase [12]. However, in implementation of COLEX process for large-scale lithium isotope production, substantial amount of mercury is to be handled and involves significant environmental and technical drawbacks, which have reduced its suitability for future deployment [12].

Energy Requirement: (High)

In industrial production, for the continuous circulation of lithium-amalgam (Li-Hg) and aqueous LiOH through counter-current separation stages, as well as for the electrolysis involved in lithium processing, the COLEX process requires a large amount of energy [12]. High electrical energy is consumed in the electrolysis used to produce lithium-amalgam in the COLEX process: about the energy of 46 KWh per kilogram of lithium metal is consumed, beyond the startup power required to melt the electrolyte [13]. One of the major technical limitation of the COLEX process is its high energy consumption, as identified by Badea et al., in spite of its historical low production cost and established industrial-scale operation [12].

Engineering Feasibility: (Established)

Historically it is proven that counter-current columns was used for lithium isotope separation for the COLEX process demonstrated industrial scale engineering feasibility [14]. In the established plant, various elements of the cascade system such as separation columns, electrolysis equipment, pumps, amalgam-making and decomposition equipment, and mercury-recovery systems with special pumping equipment are needed for circulating the large process flows through the systems [15]. Despite of operating COLEX process at production scale, the amalgam

decomposition issues, large mercury inventory, and extensive auxiliary equipment generated substantial engineering and operational challenges for future implementation [16].

Economic Feasibility: (Established)

For large-scale ^{6}Li production in an established plant, during the COLEX process development at Y-12 National Security Complex in the United States, the COLEX process was considered economically feasible, and its minimal production cost was one of basic advantages supporting its industrial application [12]. However, the economic attractiveness of the COLEX process is reduced as the large quantities of mercury is required for this process and the process is associated environmental issues [12].

Environmental Impact: (Very High)

Since large quantities of mercury are used in the lithium-amalgam phase with mercury vapors and mercury-containing waste generated during operation, the COLEX process has very high environment impacts [12]. Historically, in the COLEX process operation, the mercury is lost in buildings, equipments, surrounding soils and creates long-term environmental contamination and requires subsequent mercury-recovery with cleanup activities [17].

Justification for Representative Separation Factor for COLEX Process ($\boldsymbol{\alpha_{COLEX} = 1.055}$)

For the COLEX process, Palko et al. reported that when LiCl was dissolved in ethylenediamine (EDA), and propylenediamine (PDA), the lithium isotope separation factor was 1.054 and 1.057, respectively [10]. In addition, Okuyama et al. demonstrated that the separation factor varies under different temperatures and the reported range was from1.028 to 1.085 [11]. For the Li(Hg)-LiCl(DMSO) exchange system where DMSO (dimethyl sulfoxide) serves as the solvent for LiCl, Okuyama also reported an isotope separation factor of 1.056 ± 0.001 [11].

Therefore, based on the report of Okuyama et al. for the COLEX process $\alpha = 1.055$ was selected as a representative separation factor and adopted as the baseline value for the present comparative study.

5. Electrochemcal Process

Electrochemical lithium isotope separation is based on the slight difference in the electrochemical behavior of ^{7}Li and ^{6}Li during lithium-ion transport phenomena such as electrodeposition, and intercalation processes [18].

Mechanism: (Electrochemical Isotope Effect)

In electrochemical process, lithium isotopic fractionation arises from differences in ionic mobility, solvation characteristics, and electrode-electrolyte interactions, enabling preferential enrichment of one isotope under appropriately controlled electrochemical conditions [18]. In the electrolyte system, when an electrical potential is applied, Li^+ ions move toward the cathode, and reduction reactions take place.

$$Li^+ + e^- = Li \text{ [18] .}$$

The small mass difference between $^6Li^+$ and $^7Li^+$ produces slight differences in diffusion, ionic mobility, and electrochemical transport behavior [19, 20]. Although the electrochemical isotope effect is small in a single electrochemical stage, repeated enrichment through multistage cascade operation enables progressive lithium isotope separation.

Scalability: (Scalable)

In electrochemical process, repetitive multistage can potentially be arranged in a multicolumn cascade system, and so it has significant potential for scale up. Different scale up configurations of electrochemical process was studied by Acosta and Flexer and they showed that it is possible to extend this process through repeated stages. They also showed that at least 145 stages were required for 90% enrichment in ^6Li under their simulation conditions [21]. Although the electrochemical process is considered scalable for large scale lithium isotope separation [21], it remains at the development stage with challenges associated with the large number of stages, process optimization, operating conditions [18].

Energy Requirement: (Very High)

Large amount of electrical energy is required for electrochemical separation process, because the separation is driven by intercalation, insertion or electrochemical deposition with applied voltage [21]. Energy consumptions of approximately 2520-6640Wh g^{-1} lithium for the prospective large scale electrochemical methods was estimated by Acosta and Flexer. Their evaluation depended strongly on the electrochemical technique and operating conditions [21]. In electrodeposition technique for lithium, they estimated about 6200Wh g^{-1} lithium for enrichment to 90% lithium over 153 stages that correspond to an average energy consumption of approximately 40KWh kg^{-1} lithium per stage [21]. Hence, the energy requirement for electrochemical process is very high.

Engineering Feasiblity: (Feasible)

The engineering feasibility of the electrochemical lithium isotope separation process is supported by experimental operation with different cathode materials and solvents, with the isotope separation performance strongly influenced by the electrode-electrolyte combination and applied conditions [18]. In electrochemical process, lithium isotope separation can be performed using simple electrochemical cells in which lithium containing electrode, electrolyte, and controlled electrical potential are used to achieve isotope-selective deposition or intercalation. With different electrode substrate and electrolytes, Zhang et al. experimentally demonstrated lithium isotope separation using electrochemical deposition and intercalation [18]. Despite extensive experimental testing, the electrochemical process still requires further development of stable electrode materials, process control, inter-stage material transfer, continuous multistage operation, as industrial-scale implementation [21].

Economic Feasibility: (Feasible)

The economic feasibility of the electrochemical process is possible, because Acosta and Flexer showed an estimation with relatively low costs for prospective pilot or industrial scale production, with the calculated costs varying among the electrochemical techniques [21]. The total cost they calculated per gram was approximately $0.40-1.16 for the evaluated electrochemical insertion with graphite and gallium and electrodeposition methods showing lower estimated costs than several other methods [21]. However, the costs are estimated only for the electrochemical enrichment stages including related equipments, and the costs associated with drying, product conversion, reactant purification and other side processes are excluded. So, further economic assessment is required before industrial-scale implementation [21].

Environmental Impact: (Low)

The electrochemical process for lithium isotope separation is considered a more environmentally friendly alternative than the COLEX process, because it does not use mercury or mercury related compounds that could generate amalgam related wastes or mercury vapor [12]. In addition, lithium isotope separation is achieved through electrodeposition, electromigration, or intercalation rather than isotope exchange between amalgam phase and aqueous phase [12]. Although further optimization is required for minimizing the material consumption as well as energy related environmental impacts during large scale operation, the electrochemical process is considered to have a low environmental impact compared with the COLEX process [12].

Justification for the Representative Separation Factor for Electrochemical Process ($\alpha_{Electrochemical} = 1.040$)

In electrochemical process, the separation factor for lithium isotope separation can be changed and strongly depends on operation conditions [22]. Zhang et al. reported separation factor of around 1.04 and 1.05 when they used propylene carbonate (PC) and [BMIM][DCA], respectively

with LiBr4 as the supporting salt and graphite cathode [18]. Separation factor range of 1.023-1.031 was reported by Murali er al. with variation in electrode material [2]. Zhang et al. also reported that separation factor of 1.033 and 1.054 was obtained using gel electrolyte and ionic solvent, respectively [18]. Considering these reported values of separation factor, 1.04 was selected as the representative separation factor as the baseline value for the electrochemical process.

6. Ion-Exchange process

Based on the isotopic exchange equilibrium, lithium isotope separation by ion-exchange chromatography takes place between the resin phase and the solution phase. To achieve the desired isotope separation between ^{6}Li and ^{7}Li, the process exploits the slight isotopic exchange effect in ion-exchange equilibrium [23].

Mechanism: (Ion-Exchange)

In the mechanism of displacement chromatography, a lithium-containing band migrates through a cation-exchange resin, and the slight isotope-dependent difference in the exchange equilibrium causes ^{6}Li and ^{7}Li isotopes to fractionate along the migrating band. Using a porous ion-exchange resin, Araki et al. demonstrated this mechanism and observed ^{7}Li enrichment toward the front part of the lithium band, while the lighter ^{6}Li preferentially accumulated toward the rear portion of the band [23]. Suzuki et al. subsequently investigated this displacement-chromatography mechanism using a cation-exchange resin and confirmed lithium-isotope fractionation during band migration, with ^{7}Li enriched at the front boundary and ^{6}Li enriched in the rear portion; their study further developed the chromatographic configuration toward long-distance and automated operation [24].

Scalability: (Scalable)

Ion-exchange chromatography has demonstrated potential for scale-up because displacement chromatography can be implemented using established column-operation principles. Putra et al. demonstrated lithium isotope separation using a highly cross-linked cation-exchange resin and showed that the isotope separation coefficient increases with cross-linkage, although this improvement tends to saturate at very high cross-linkage [25]. More importantly, Suzuki et al. conducted an engineering-scale assessment using 1 and 4 m columns and estimated migration lengths of approximately 340, 1600, and 5800 m to obtain 20%, 50%, and 90% 6^{6}Li enrichment, respectively. They subsequently designed and fabricated a prototype automatic circular chromatographic system with boundary detection for long-distance migration [24]. Thus ion-exchange chromatography is technically scalable, although achieving high ^{6}Li enrichment requires very long migration distances and correspondingly large-scale chromatographic equipment [23].

Energy Requirement: (High)

Ion-exchange chromatography has a relatively low energy requirement as the separation can be achieved through ion-exchange equilibrium between the resin and solution phases without an energy-intensive driving force. A study, conducted by Araki et al., on displacement chromatography using porous ion-exchange resin specifically reported that the ion-exchange method does not consume a large amount of energy [23]. Despite the possibility that practical energy requirements can be increased with liquid circulation over the long migration distances and chromatographic operation at larger scales, the ion-exchange chromatography can still be considered a relatively low-energy separation process [23].

Engineering Feasibility: (Feasible)

Suzuki et al. conducted an engineering study using ion-exchange chromatography and investigated the detection of lithium adsorption-band boundaries for the development of an industrially relevant circular chromatographic system. Based on their experimental results, they designed and fabricated a prototype automatic circular chromatographic system equipped with a boundary-interface detector, demonstrating the feasibility of implementing long-distance migration in an automated configuration [24]. Araki et al. investigated the effect of chromatographic development speed on separation performance using porous ion-exchange resin and showed that increasing the development speed reduces separation performance because the length of height equivalent to a theoretical plate (HETP) increases with development speed, although higher development speeds can improve production rate [23]. Therefore, ion exchange chromatography is technically feasible for engineering-scale implementation, although optimization of development speed, separation performance, and equipment configuration is necessary for efficient large scale operation.

Economic Feasibility: (Feasible)

In economic perspective, ion-exchange chromatography has demonstrated potential for practical plant implementation for ^{6}Li production. Sujuki et al. demonstrated that achieving high isotope enrichment requires substantially extended chromatographic migration distances and considerable column infrastructure [24]. A recent simulation of a simulated moving-bed chromatographic system estimated production of 186 Kg/year of 90% 6Li using a 50 cm diameter, and 437 m long column, demonstrating the potential for high-throughput production while highlighting the scale-up challenge of chromatographic system [26]. Therefore, ion-exchange chromatography has promising economic potential for feasibility of ^{6}Li isotope production, although the capital and operating costs associated with large scale chromatographic equipment require careful optimization.

Environmental Effect: (Moderate)

Ion-exchange chromatography offers an environmental advantage over the conventional amalgam process because it does not require toxic mercury, and so it avoids the generation of mercury associated wastes and vapor for lithium isotope separation [25]. However, the use of regenerating chemicals and the resulting aqueous waste should also be considered in assessing its overall environmental impact. Therefore, ion-exchange chromatography is considered as a comparatively environmentally favorable, mercury-free alternative to the COLEX process with a moderate environmental impact [24].

Justification of Representative Separation Factor of Ion Exchange Process ($\boldsymbol{\alpha_{IEx} = 1.030}$)

In ion-exchange process, a maximum separation factor of 1.022 was obtained by Kakihana et al. using 0.1M LiOH solution in 20% acetone water and synthetic inorganic ion exchanger [27].

Kanzaki and Suzuki reported a $^{7}Li/^{6}Li$ isotope separation coefficient of 1.04 for cubic-$HNbO_3$, with a theoretical maximum of approximately 1.05 at 25℃ for completely dehydrated inorganic ion exchangers [28]. Using LiI of LiBr as the lithium salt with methanol or ethanol as the solvent, Fujine at al. reported a lithium isotope separation factor of 1.03-1.06 [29] Therefore, a separation factor of 1.030 was adopted as the representative value for our present comparative study for ion-exchange process.

7. Determination of Minimum Requirement of Theoretical Stages

Since we assume the lithium isotope separation process under ideal condition, the minimum theoretical stage requirement can be computed to achieve ^{6}Li enrichment from 7.5% to 60%. Using equation (2), we developed a python code for computational simulation in Google Colab, where, feed composition, $x_f = 0.075$; production composition, $x_p = 0.600$; and the selected values (Justified in each process description) of representative separation factors (α) are 1.060, 1.055, 1.040 and 1.030 for distillation process, COLEX process, electrochemical process and ion-exchange processes, respectively.

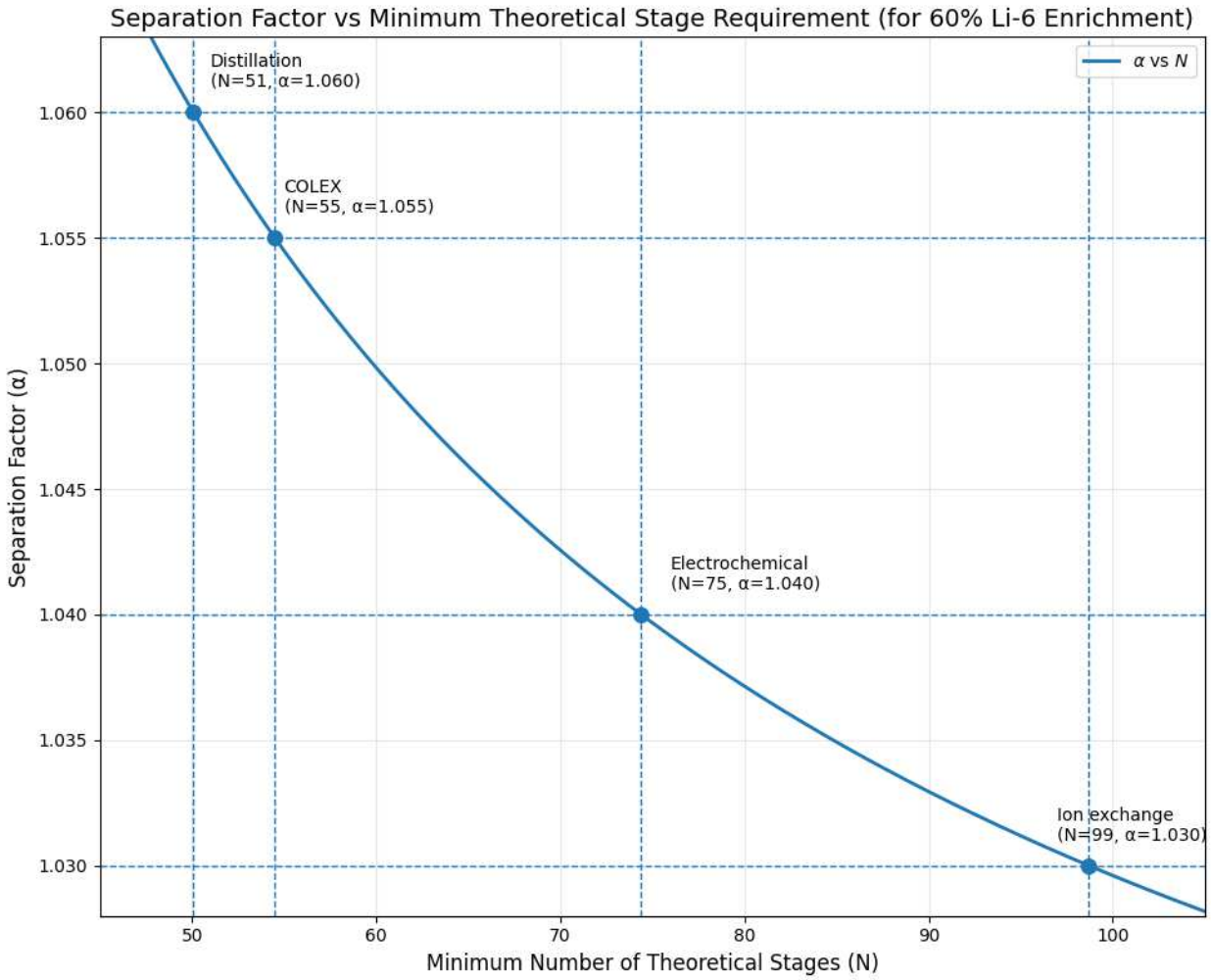


Figure 1. Relationship between representative separation factors and minimum theoretical stage requirements for ^{6}Li enrichment to 60% for the four separation processes.

All of the calculations are performed in computational simulation in Google Colab. The simulation code is provided in **Supplementary Code LHR2** in Section 10. The required continuous stage numbers were rounded to the next integer to result in minimum theoretical stage numbers.

Direct calculation can be performed using Equation (2), for example:

for distillation process, $N = \frac{ln\{\frac{x_p(1-x_f)}{x_f(1-x_p)}\}}{ln(\alpha)} = \frac{ln\{\frac{0.600(1-0.075)}{0.075(1-0.600)}\}}{ln(1.060)} = 50.1 \rightarrow 51$ (Rounded to the next integer stage)

Table 1. Calculated continuous and minimum integer theoretical stage requirements for ^{6}Li enrichment to 60% using representative separation factors for the four processes.

Process	Representative α	Continuous N	Integer N
Distillation	1.060	50.1	51
COLEX	1.055	54.5	55
Electrochemical	1.040	74.4	75
Ion-Exchange	1.030	98.7	99

From the Figure 1 and Table 1, it is observed that the processes of distillation, COLEX, electrochemical and ion-exchange with their corresponding representative separation factors of 1.060, 1.055, 1.040 and 1.030 require 51, 55, 75 and 99 integer theoretical stages, respectively.

It is remarkable that the process with a smaller representative separation factor requires a greater number of separation stages, while the process with a greater representative separation factor requires a smaller number of separation stages.

8. Results and Discussion

The comparative results of the current study are obtained from the process discussion and the computational simulation that presents the minimum required theoretical stages (N) based on representative separation factors (α) for each process. The results are presented in Table 2.

Table 2. Comparative evaluation of lithium isotope separation processes based on separation characteristics and minimum theoretical stage requirement.

Characteristics	Distillation	COLEX	Electrochemical	Ion-Exchange
Mechanism	Vapor-liquid equilibrium	Chemical exchange	Electrochemical isotope effect	Ion-exchange
Scalability	Scalable	Established	Scalable	Scalable
Energy Requirement	Very High	High	Very High	High
Engineering Feasibility	Feasible	Established	Feasible	Feasible
Economic Feasibility	Feasible	Established	Feasible	Feasible
Environmental Impact	Low	Very High	Low	Moderate
Representative Separation Factor (α)	1.060	1.055	1.040	1.030

Minimum Theoretical Stages (N)	51	55	75	99

Table 2 demonstrates that COLEX is an established industrial isotope separation process, but presents a very high environmental impact. Electrochemical process with higher theoretical stages and energy requirements offers low environmental impact, while ion-exchange separation has the lowest representative separation factor and highest stage requirement. Interestingly the distillation process offers the most balanced combination of separation performance and feasibility among the four processes, with lowest minimum theoretical stage requirement (N=51) and highest representative separation factor.

9. Conclusion

The study presented a comparative evaluation of the four lithium isotope separation processes: distillation, COLEX, electrochemical and ion-exchange. Each process discussed the seven characteristics of separation mechanism, scalability, energy requirement, engineering feasibility, economic feasibility, environmental impacts including separation performance in terms of the representative separation factor that was selected considering reported separation factors. Considering ideal process conditions, a computational simulation was performed to determine the minimum required theoretical stages using Fenske equation and the minimum stage numbers were also included in the comparative evaluation. The computational simulation presented that distillation, COLEX, electrochemical and ion-exchange processes required 51, 55, 75 and 99 theoretical stages, respectively to enrich ^{6}Li from 7.5% to 60%.

Overall, the study demonstrates that COLEX is an established lithium isotope separation process, but it faces many environmental issues, since it uses toxic mercury in its process. Electrochemical process offers mercury free isotope separation technique, but it requires very high energy and greater number of stages. Although the Ion-exchange process is scalable and has engineering feasibility, its separation performance is relatively low compared with other three processes, and it requires greatest number of stages (N = 99). While distillation process, with highest representative separation factor and lowest stage requirement, demonstrated the most balanced combination of lithium isotope separation performance and feasibility among the four processes considered.

10. Supplementary Code

The supplementary computational simulation code is in the Google Colab Notebook namely “Supplementary Code LHR2”. The link is

https://colab.research.google.com/drive/1KxvHaOSiNdgdSuzd4Z7oh93M3n2-CNZ3?usp=sharing

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