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Original Article

Economic evaluation of thorium oxide production from monazite using alkaline fusion method



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ABSTRACT

Monazite is a phosphate mineral that contains thorium (Th) and rare earth elements. The Th concentration in monazite can be as high as 500 ppm, and it has the potential to be used as fuel in the nuclear power system. Therefore, this study aimed to conduct the techno-economic analysis (TEA) of Th extraction in the form of thorium oxide (ThO₂) from monazite. Th can be extracted from monazite through an alkaline fusion method. The TEA of ThO₂ production studied parameters, including raw materials, equipment costs, total plant direct and indirect costs, and direct fixed capital cost. These parameters were calculated for the production of 0.5, 1, and 10 ton ThO₂ per batch. The TEA study revealed that the highest production cost was ascribed to installed equipment. Furthermore, the highest return on investment (ROI) of 21.92% was achieved for extraction of 1 ton/batch, the ROI was decreased to 5.37%. This is mainly due to a significant increase in the total capital investment with increasing ThO₂ production cost was achieved for 1 ton ThO₂/batch equal to 335.79 \$/Kg ThO₂.

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1. Introduction

In recent years the ever-increasing world population resulted in a significant demand for energy to cater to diverse domestic and commercial needs. Based on an illustrative report published by the United States Department of Energy (US-DoE), the energy consumption was expected to double by 2050, in particular for commercial applications [1,2]. One of the major sources of energy with the potential to fulfill this demand is nuclear energy technology. Although uranium fuel technology still is of great attention for power industries, several challenges, including the shortage of uranium reserves and lack of waste storage, have raised global concerns about the further use of standard uranium-based nuclear reactors [3]. Nonetheless, recent studies revealed that Thorium-232 could be an efficient and safe fuel alternative to uranium to generate nuclear power for commercial purposes [4–8]. It should be noted that the abundance of thorium in nature is three or four times higher than uranium [4]. Moreover, compared to uranium fuels, thorium fuels produce much less plutonium and minor actinides; therefore, induced radiotoxicities are lower by more than two orders of magnitude [9]. These are the main advantages that persuaded researchers to consider replacing uranium with thorium for future nuclear energy technologies.

The thorium-based nuclear reactors can provide clean and environmentally friendly energy for the sustainable development goal of energy sources for the future generation. Thorium (²³²Th) as a fertile isotope with thermal neutron capture in the reactors transmutes to a fissile ²³³U nucleus, as expressed in Eq. (1). The fissile ²³³U forms a new nuclear fuel, obtained either as a fission product during nuclear reaction or may be removed chemically from spent fuel, then forms a new nuclear fuel [3].

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(1)

Thorium can be found in the form of monazite ore mixed with other rare earth elements (REEs) [10]. Given greater attention to employing thorium as a promising fuel reactor, particularly for Generation-IV nuclear reactor, more efforts have been undertaken to explore and extract thorium deposit resources [11–13]. For the past ten years, several researchers have expended much effort to develop an economical process for recovering thorium, rare earths, and uranium from monazite sands [14-17]. Currently, the conventional extraction and recoverable process of high purity thorium and REE metals from monazite ore consists of complex energy-intensive technologies including physical beneficiation, leaching, purification, and separation into individual compounds [18–23]. Among the hydrometallurgical techniques, the application of sulfuric acid, nitric acid, hydrochloric acid, and alkaline reagents have been studied by researchers for the extraction and recovery of Th and REE metals from monazite [24,25]. Apart from the mentioned industrially practiced methods, the literature has also proposed methods such as high-temperature reduction [26,27], roasting [28], mechano-chemical decomposition [29], and other independent methods [30,31].

The conventionally adopted hydrometallurgical techniques for monazite decomposition are leaching with sulfuric acid and alkaline leaching with sodium hydroxide. The acid leaching route is performed at a temperature of 230 °C, wherein the REEs are converted to sulfates and remain in the solution. However, the reaction of sulfuric acid with a majority of impurities in the monazite resulted in the low purity of final products [32]. Hence, the remaining thorium and uranium in the leach residue make this process less environmentally friendly [33]. As a result, recently, the alkaline digestion method with sodium hydroxide was investigated for monazite decomposition. In general, the alkaline method employs sodium hydroxide, and the operating temperature was around 160 °C, where the dosage of sodium hydroxide was 50 wt% [34]. Nonetheless, a recent study by Galvin and Safarzadeh [35] studied the application of potassium hydroxide for thorium extraction from monazite by alkaline fusion method.

Though there are numerous studies about the extraction of Th from its resources, such as monazite, it is essential to study the economic analysis of Th production to realize whether it is costeffective. A previous study by Salehuddin et al. [15] on the economic analysis of thorium extraction from monazite using the acid leaching method. However, the current research has provided the techno-economic evaluation of thorium extraction in the form of thorium oxide (ThO₂) from monazite ores through an innovative alkaline fusion method. The main highlight of this study is the method adopted in extracting the thorium from monazite. Although purely hydrometallurgical and pyrometallurgical methods have already been established for extracting the desired elements from monazite, the alkaline fusion method will have the upper hand over the existing methods, with improved decomposition efficiency with economically viable processing infrastructure. The method overcomes a major drawback of the conventional acid leaching of monazite, i.e., the loss of the phosphate, which may have potential economic value. However, sodium hydroxide decomposition of monazite allows for the recovery of phosphate as tri-sodium phosphate, which can then be used to produce fertilizers or as a feed chemical for other processes. The efficiency of extracting thorium through the alkaline method is much higher compared to the acid method.

SuperPro Designer (1), a process simulator software, is used for analyzing the techno-economic feasibility of large-scale chemical processes. In the present study, the software has facilitated comprehensive process calculations and optimization of thorium extraction from monazite using the alkaline fusion method. With built-in mathematical models, the software has been effectively exploited to present the techno-economic analysis of producing thorium with less technical knowledge, time, and process data. This approach is expected to impact product and process performance directly and ultimately bring about corporate profitability. Thus, the computer-aided tool will be utilized to conduct the economic analysis to determine the influential factors on the production costs to ensure a low-cost Th extraction process.

2. Materials and method

Thorium can be extracted by decomposing monazite using the alkaline fusion method. Fig. 1 shows the process flow diagram (PFD) to extract thorium from monazite and ThO₂ production. In order to determine the commercialization potential of this process, it is essential to evaluate the economic performance of the proposed process. Hence, techno-economic analysis (TEA) is performed for the production of ThO₂ from monazite. The techno-economic analysis evaluates the production cost, technology, and scale. The total investment cost of a production plant is a function of certain factors such as equipment, production scale, raw materials, operation cost, installation, electricity, and auxiliary facilities. Overall, the TEA represents a thorough assessment of the production expenses [36]. For this purpose, the commercial process simulation software SuperPro Designer V8.5 was employed. With this software, economic evaluation can be automatically performed based on the mathematical model built by the user [37]. The main cost elements include raw materials purchase, energy consumption, equipment capital investment, and waste treatment system [38]. The total production costs were calculated for different production scales, i.e., 0.5, 1.0, and 10.0 tons of the final product (ThO₂ powder) for a cost comparison. Besides the system's main product, the waste stream, such as gas emissions, aqueous and solid residues, could also have economic values depending on the waste treatment, as seen in Fig. 1. The produced trisodium phosphate (Na₃PO₄) from the water leaching step (P-3) can also be recycled to be used as a fertilizer [39].

2.1. Thorium laboratory scale extraction process

The alkaline fusion method with sodium hydroxide (NaOH) was employed to extract thorium from monazite [29]. The advantage of the alkali-based decomposition method is that it is a mature process with simple equipment and a high grade of integrated utilization [40]. Table 1 presents the chemical composition of the monazite obtained from the Malaysian peninsula. The detailed characterization of the monazite has been studied by Udayakumar et al. [10]. Firstly, monazite is ground below 50 μ m and mixed with NaOH as described in procedures P-1 and P-1/2, respectively. In the next step, the mixture is reduced to 350 °C for 4 h (procedure P-2/ V-101) according to Eq. (2). The fused sample is containing



Fig. 1. Thorium oxide process flow diagram from monazite with alkaline fusion method.

 Table 1

 Chemical composition of monazite

Element	Weight Percent (%)	Element	Weight Percent (%)
Al	0.25	Ce	36.48
Si	2.05	Pr	2.84
Р	27.27	Nd	17.26
K	0.00	Sm	2.17
Ca	0.92	Gd	1.38
Ti	0.37	Hf	0.00
Mn	0.08	W	0.48
Fe	0.44	Pb	0.16
Ni	0.06	Th	6.58
Y	2.11	U	0.34
Zr	1.18	Na	0.00
Sn	0.41	Mg	0.00
La	13.37	Trace elements (Cu, Ag, Ba, Bi, Eu, Tb, Dy, Ho, Er, Tm, Yb, Lu, Ta, Tl, Rh, Pd)	~1.2

insoluble hydroxide residue of rare earth elements, including thorium hydroxide (Th(OH)₄) and water-soluble Trisodium phosphate (Na₃PO₄), and other impurities. Moreover, during the reduction process, the minerals of Fe, Al, Ti, and Si can also be dissolved in NaOH solution, making it easy to separate them with rare earth hydroxides, thorium, rather than being able to be separated in their solid-state. In order to remove the phosphate products, the fused sample is leached with water at 80 °C for 1 h (procedure P-3). Throughout this process, the Na₃PO₄ is dissolved in water, and is separated from Th(OH)₄ with filtration process at filter unit P-4. Next, the filtered residue is dried at 80 °C overnight (P-5).

$$Th_3(PO_4)_4 + 12NaOH \rightarrow 4Na_3PO_4 + 3Th(OH)_4$$
(2)

The hydroxide residue can be leached in nitric, sulfuric, or hydrochloric acid to prepare rare earth solutions for separation processes [41]. Of all the acids, hydrochloric acid (HCl) was found to exhibit consistently positive results of better dissolution behavior in the literature [25]. Therefore, the filter cake after the drying process was leached with HCl 6 M at 90 °C for 90 min in leaching vessel P-6. At this step, the thorium hydroxide is converted to thorium tetrachloride (ThCl₄) as Eq. (3). Following the acid leaching, filtration is performed to separate the ThCl₄ from other REE residues (P-7). Later, the ThCl₄ needs to transform to insoluble thorium oxalate (Th(C₂O₄)₂) at precipitator (vessel P-8) through reaction with oxalic acid (C₂H₂O₄) as Eq. (4). The obtained product is then subjected to filtration at procedure P-9. As the final step, the Th(C₂O₄)₂ filter cake is calcined at 1000 °C in a furnace to convert into ThO₂ as the final product (Eq. (5)). The processing time for the respective procedures in the simulated model is summarized in Table S1 in supplementary materials.

$Th(OH)_4 + 4HCl \rightarrow ThCl_4 + 4H_2O \tag{1}$	3))
	-,	,

$$ThCl_4 + 2C_2H_2O_4 \rightarrow Th(C_2O_4)_2 + 4HCl$$
(4)

$$Th(C_2O_4)_2 \rightarrow ThO_2 + 2CO_2 + 2CO \tag{5}$$

3. Results and discussion

The production cost of the plant takes into consideration the fixed capital, material, wastewater treatment, disposal, as well as utility expenses. The construction period for the plant was assumed to be six months, and the total project lifetime was set for 15 years. The overall process parameters and the number of batches for different plant capacities on a yearly basis is sammurized in Table S2 in supplementary materials. As shown in Table S2, the overall batch time increases with the production capacity. This leads to a lower number of batches/years. In this study, the economic evaluation was carried out for a single batch cycle with a related recipe batch time.



Fig. 2. Equipment price distribution for different production scale of ThO₂.

3.1. Raw material and installed equipment costs

The material consumption for the designed process is estimated based on the stoichiometric amounts from Eqs. (2)-(5). The number of required materials (bulk materials and heat transfer agents) with their unit price in the economic evaluation for the extraction process of ThO₂ from monazite at different production scales is provided in supplementary materials in Table S3. The annual cost of each material can be calculated by multiplying the related unit price by the yearly amount that is utilized in a process. Besides, the details of the equipment and their purchase costs used in the economic evaluation of the production process are summarized in Tables S4 and S5, respectively, in supplementary materials. The SuperPro Designer software has built-in models that estimate the cost for each required equipment based on the provided size variable [37]. According to Table S5, on increasing the mass production, the total equipment price was significantly increased from \$3,886,000 for 0.5 ton ThO₂ per batch to \$36, 488, 000 for 10 ton ThO₂ per batch. Fig. 2 shows the total cost for each equipment at different mass production of ThO₂ per batch. As seen in the figure, the highest equipment cost is related to the furnace (reactor) for the fusion process of monazite, which is almost about \$23, 596, 000 for the production of 10 ton ThO₂ per batch. The other equipment price is almost below \$3,000,000. It should be emphasized that the capacity of the equipment varies according to the production throughput.

3.2. Capital costs analysis

The major components of the economic analysis were estimated by the simplified framework introduced by Peters et al. [42] for economic analysis of industrial chemical projects. In this method, the costs are estimated as a percentage of the equipment purchase cost (for capital costs) or the costs of raw materials and consumables (for operating costs). Generally, the fixed capital costs are divided into two categories as Eq. (6), i.e., total plant costs (TPC) and general expenses such as contractor and contingency fee (CFC) [37]:

$$DFC = TPC + CFC$$
(6)

Where DFC is direct fixed capital costs. TPC and CFC are also can be expressed as Eqs. (7) and (8), respectively:

TPC = TPDC + TPIC(7)

CFC = Contractor fee + Contingency fee (8)



The percentage coefficient of direct plant cost factors.

Factor	Name	Percentage
A	Piping	35% PC
B	Instrumentation	40% PC
C	Insulation	3% PC
D	Electrical facilities	10% PC
E	Building	45% PC
F	Yard improvement	15% PC
G	Auxiliary facilities	40% PC

Where TPDC is total plant direct cost and TPIC is total plant indirect cost.

The direct plant cost includes total equipment purchase cost (PC), installation, process piping, instrumentation, insulation, electrical, buildings, yard improvement, and auxiliary facilities (including waste treatment and disposal, emission control, and other related equipment). Meanwhile, indirect plant cost includes engineering and construction. The PC can be expressed as Eq. (9):

PC = listed equipment purchase costs + unlisted equipment purchase costs (9)

Unlisted (skids, storage units, etc.) and consumable equipment purchase cost were assumed as 20% of the total equipment purchase cost (PC) in SuperPro Designer software. Other factors in the direct plant cost were calculated from the PC, as shown in Table 2 [37].

The indirect plant cost parameters can be calculated from direct costs (DC) and indirect costs (IC). DC can be defined as below:

$$DC = PC + installation costs + A + B + C + D + E + F + G$$
 (10)

The installation cost is the total installation cost for the listed and unlisted equipment. The installation cost of unlisted equipment was considered as 50% of the unlisted equipment purchase price. Therefore, the engineering fee and consultation fee can be calculated as 25% DC and 35% DC, respectively. Moreover, the other costs (OC) related to CFC can be defined as follow:

Contractor's Fee = 5% (DC + IC)	(11)
------------------------------------	------

Contingency Fee = 10% (DC + IC) (12)

According to the purchase price of the total equipment, Table 3 summarized the capital fixed costs for the different mass production of ThO₂ (the more details provided in Table S6 in supplementary materials). For further calculation of the capital investment, parameters considered were included startup and validation, maintenance, insurance, local tax, and factory expenses, following the economic evaluation with SuperPro Designer v8.5 [37]. As witnessed in Table 3, with an increase in mass production per batch, the total direct and indirect plant expenses also increased. However, considering the weight of the ThO₂ per batch, the total direct and indirect plant costs decreased from 25,586 USD/ kg (12, 793, 000 USD) and 15,352 USD/kg (7,676,000 USD) for 0.5 ton/batch of ThO₂ to about 11,820 USD/kg (118, 199, 000 USD) and 7092 USD/kg (70, 919, 000 USD) for 10 ton/batch of ThO₂, respectively. Moreover, Tables 4 and 5 listed the quantity of required materials and their prices, as well as utility consumption, respectively, for different production scales of ThO₂ plant. It is evident that the highest factor is related to construction costs for all the mass productions, and the minimum price is the electrical price. The insulation price stands as the second-lowest place, and equipment purchase costs are the second-highest factor.

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Table 3

Fixed capital estimate summary for ThO₂ plant at different batch mass production in USD.

Parameter	0.5 ton	1 ton	10 ton
Total Plant Direct Cost (TPDC)	12,793,000	19,145,000	118,199,000
Total Plant Indirect Cost (TPIC)	7,676,000	11,487,000	70,919,000
Contractor's Fee & Contingency (CFC)	3,070,000	4,595,000	28,368,000
Direct Fixed Capital Cost (DFC)	23,540,000	35,228,000	217,486,000

Table 4

Materials annual cost for ThO₂ plant at different batch mass production in USD.

Material	0.5 ton	1 ton	10 ton
Hydrochloric acid (HCl 37%)	239,458	433,900	919,069
Monazite	124,163,433	224,985,436	476,554,167
Oxalic Acid (C2H2O4)	146,335	265,161	561,653
Sodium Hydroxide (NaOH)	33,258	60,264	127,648
Water	14	25	54
Total price (\$)	124,582,498	225,744,788	478,162,591

NOTE: Bulk material consumption amount includes the material used: Raw Material, Cleaning Agent and Heat Transfer Agent (if utilities are included in the operating cost).

Table 5

Process utilities cost summary for ThO2 plant at different batch mass production.

Parameter	Unit Cost (\$)	0.5 ton		1 ton		10 ton	
		Amount	Cost (\$)	Amount	Cost (\$)	Amount	Cost (\$)
Std Power (Kwh) Steam (MT) Chilled Water (MT) Steam (High P) (MT) TOTAL (S)	0.100 4.200 0.400 20.000	1,116,143 198 10,956 8	111,614 831 4382 157 116,985	2,022,462 359 19,851 14	202,246 1506 7941 285 211,978	4,283,889 760 42,049 30	428,389 3191 16,819 604 449,003

Table 6

Executive economic summary for Ti powder plant at different batch mass production.

Parameter	0.5 ton	1 ton	10 ton
Total Capital Investment (\$)	36,055,000	57,554,000	272,050,000
Operating Cost (\$/yr)	129,377,000	233,038,000	520,541,000
Revenues (\$/yr)	134,050,000	242,900,000	514,500,000
Cost Basis Annual Rate (kg MP/yr)ª	383,000	694,000	1,470,000
Unit Production Cost (\$/kg MPr)	337.80	335.79	354.11
Unit Production Revenue (\$/kg MPr)	350.00	350.00	350.00
Gross Margin (%)	3.49	4.06	- 1.17
Return On Investment (%)	18.38	21.92	5.37
Payback Time (yr)	5.44	4.56	18.61

^a MP = Total Flow of Stream "ThO₂"

3.3. Unit production cost of ThO₂

The economic evaluation was performed for three different mass productions of the final product with SuperPro Designer V8.5 software, and the final results are summarized in Table 6. The selling price of the final ThO₂ was considered as 350 USD/kg. The results showed that the maximum return on investment (ROI) is 21.92%, which was obtained for 1 ton ThO₂ production per batch. However, it has to be noted that the ROI decreases with further increase in production scale to 10 ton ThO₂, i.e., 5.37%. Overall, the unit production cost of ThO₂ was calculated to be in the range of 337.80–354.11 USD/kg. In the previous work by Salehuddin et al. [15], where the economic evaluation of acid leaching was investigated, it was reported that the unit production cost was in the range of 501.18–553.17 USD/kg ThO₂. Hence, it can be concluded that the alkaline fusion process is economically more profitable.

In order to study the effect of an increase in annual throughput on the economic evaluation factors (such as profit, ROI, unit costs, etc.), the batch size was increased from 0.5 ton to 10 ton, and the results are shown in Fig. 3(a) and (b). It can be seen from Fig. 3(a) that the revenues and net profits increase linearly with the plant capacity. This indicates that the plant should be run at the highest capacity. However, examining the economic factors used to evaluate the profitability of investment, namely the Return on Investment (ROI) and the Internal Rate of Return (IRR), it was observed that the value of these factors does not vary linearly with batch size (Fig. 3(b)). It shows that the investment is more profitable if the batch size is in the range of 1500-2500 kg/year since the ROI and IRR are higher in this range. This is mainly due to a significant increase in the total capital investment by increasing the production scale of ThO₂. It is evident from Table 6 and Fig. 3 that with the production scale-up from 0.5 ton to 10 ton ThO₂, there is a sharp increase in the total capital investment rather than the net profit. This means that the revenues rise faster than the expenses because production is increased by utilizing larger vessels, but not more vessels, and the cost of a vessel with respect to its size is not linear [43]. Beyond the point of 2000 kg/year, there is a need to install multiple equipments (which is similar to building another plant), and they no longer have those benefits.



Fig. 3. Change in the economic indices with increasing the batch size: (a) revenue and net profit, (b) ROI and Internal rate of return (IRR).

3.4. Sensitivity analysis

The economic evaluation for the ThO₂ scale-up suggested that the 2000 kg ThO₂ per batch has the maximum ROI and IRR values. Hence, to investigate the model's uncertainty, the Monte Carlo simulation was applied for 2000 kg ThO₂ scale up. This simulation is a useful tool to statistically analyze the risk contributed due to the uncertainties in the economic evaluation [44,45]. The uncertainty and sensitivity analysis was done using the SuperPro Designer software and Crystal Ball add-in function in Microsoft Excel software [37]. To conduct the Monte Carlo simulation, the assumptions were specified as input variables with Crystal Ball. These input variables were used to perform several simulations in the component object module (COM) function of SuperPro Designer [37]. Via these simulations, it is possible to determine which input variables have a significant effect on the economic model described for the process. In the current study, the decision variable is the unit production cost of the ThO₂ (\$334) for 2000 kg/batch scale-up, and 5000 trials were performed to obtain a low mean standard error. The dependency of this decision variable was investigated as a

function of production costs (utilities, consumables, and raw materials, which have the highest portion of production costs), as well as the duration of operations, as summarized in Table S1 in supplementary materials. The distribution models of the input variables as chosen assumptions were set to a normal distribution with a 10% standard deviation for the costs and triangular distributions for the duration of operations. The likelihood of the unit production cost of the ThO₂ is presented in Fig. 4 in the form of the probability distribution diagram, which indicates that the distribution curve followed the normal distribution, and the certainty of occurrence of the production goal is 81.98%. According to Fig. 4, the mean and median values of unit production cost calculated with sensitivity analysis were observed to be less than 10% of the production goal (i.e., 334 \$/kg). The sensitivity analysis was performed by applying the Crystal Ball, and the sensitivity diagram is presented in Fig. 5. It is evident that the cost of monazite is the most significant input variable, which resulted in the extreme fluctuation in the unit production cost of ThO₂. Moreover, the duration of the thorium oxalate reaction (P-8) stands as the second-highest impact on the unit production costs due to its longest process time. Therefore, any



Fig. 4. Probability distribution of the main product cost (5000 trials). Mean = 363.47, Median = 363.24, Standard Deviation = 35.88, Mean Std. Error = 0.51, Range = 300-400.



Fig. 5. Contribution of uncertain parameters to the variance of the unit production cost.

delay in the step P-8 could be compensated by increasing the minimum cycle time and increasing the unit production cost.

4. Conclusion

The techno-economic analysis of ThO₂ production from monazite using the alkaline fusion method was carried out for different production scales, including 0.5, 1, and 10 tons. The results showed that the total capital investment cost for 0.5, 1, and 10 ton of ThO₂ production are \$36, 055, 000, \$57, 554, 000, and \$272, 050, 000, respectively. According to the justified factors, the total revenue per year for 0.5, 1, and 10 ton ThO₂ were determined to be \$134, 050, 000, \$242, 900, 000, and \$514, 500, 000, respectively. Furthermore, the total direct fixed capital costs were successfully estimated to be \$23, 540, 000, \$35, 228, 000, and \$217, 486, 000, respectively. The calculation of the return on investment and payback time indicated that the highest ROI of 21.92% was achieved at 1 ton per batch ThO₂ production in 4.56 years payback time. It was proven that the commercialization of ThO₂ production from monazite with alkaline fusion method is economically feasible with a reasonable return. In the current cost estimation method, appropriate and relatable information on the process parameters and infrastructure has been considered. The specific costs involved in the process are orientational, wherein the level of error is about 5–10%. However, the costs can be sufficient to be relied upon for process panning at this stage, as the objective of this estimation is not to determine the actual cost but to assess the expenditure and select appropriate and favourable variants of the process, parameters, and methods. Overall, the TEA of extracting thorium from monazite has helped reveal the technical and economic advantages of the alkaline fusion method. Computer-aided tools have aided in conducting the techno-economic analysis of producing thorium, with less technical knowledge, time, and process data.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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