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Whole-Process Pollution Control for Cost-Effective and Cleaner Chemical Production—A Case Study of the Tungsten Industry in China

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ABSTRACT

In this research, a methodology named whole-process pollution control (WPPC) is demonstrated that improves the effectiveness of process optimization. This methodology considers waste/emission treatment as a step of the whole production process with respect to the minimization of cost and environmental impact for the whole process. The following procedures are introduced in a WPPC process optimization: ① a material and energy flow investigation and optimization based on a systematic understanding of the distribution and physiochemical properties of potential pollutants; ② a process optimization to increase the utilization efficiency of different elements and minimize pollutant emissions; and ③ an evaluation to reveal the effectiveness of the optimization strategies. The production of ammonium paratungstate was chosen for the case study. Two factors of the different optimization schemes—namely the cost-effectiveness factor and the environmental impact indicator—were evaluated and compared. This research demonstrates that by considering the nature of potential pollutants, technological innovations, economic viability, environmental impacts, and regulation requirements, WPPC can efficiently optimize a metal production process.

1. Introduction

With the rapid development of modern industry, consumption of primary energy and resources has resulted in severe pollution and the emission of huge amounts of industrial waste, especially in developing countries. In the mainland of China, the total industrial output increased by 20.52% from 2011 to 2015. Within the same time frame, the emission of pollutants, including industrial wastewater, waste gas, and solid waste, increased significantly due to inefficient pollution control (according to the China Statistical Yearbook on Environment, 2012–2016). In 2015, for example, the discharged amounts of industrial wastewater, material contributing to chemical oxygen demand (COD), and ammoniacal nitrogen (NH_3-N) were 1.816×10^{10}, 2.56×10^6, and 1.963×10^5 t, respectively, mainly from primary metal production (see Tables S1 and S2 for details). Furthermore, a supply shortage of materials and metals is occurring and is driving the definition of critical materials/metals [1], most of which are rare metals of strategic importance, including tungsten (W), magnesium (Mg), niobium (Nb), indium (In), and rare earth metals [2]. The production of these materials can result in a huge amount of pollutants, since their concentration in minerals is normally very low. For example, China supplies 84.6% of the world market of primary rare earth elements (REEs) [3] and 83% of primary tungsten materials [4] (in fact, more than 80% of the critical materials defined by the European Union are supplied by industries in China [5]), and these processes are partially responsible for the environmental issues currently present in China. Associated human health problems have already been observed [6]. Consequently, new environmental regulations and standards have been issued to limit pollution discharge, starting in 2015. However, these measures have substantially increased the pollution treatment-related costs for companies. Therefore, cost effectiveness via waste reduction is a key aspect in solving these environmental problems and ensuring the sustainability of China's current industry.

Considering the entire life-cycle of a metallic material (e.g., rare metals), the profit obtained from processing the mineral-hosted metal into intermediate products is lower than that from the

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process of converting the metal into high-value products. The narrow profit in preparing an intermediate product is very sensitive to new investments in waste treatment and the implementation of new facilities. Therefore, it is imperative to reduce the pollution level while simultaneously considering the process profit in order to ensure its sustainable development. Cleaner production, which is an initiative or principle that minimizes waste and emissions while maximizing the product output, is one possible method to reduce the pollution level. The atomic efficiencies of the main elements in the product are primarily taken into account during the implementation of cleaner production. A variety of methodologies have been developed for the process optimization of an individual step, including automatic parameter identification with computational simulation [7], response surface methodology, and a central composite design with a series of experimental data [8–11]. In practice, these methodologies can be integrated into a two-layered system that consists of real-time optimization and model predictive control to achieve unit optimization [12]. Global optimization has been used to achieve the optimization of a whole production process by integrating a range of technical indices from different unit processes [13]. To improve the efficiency of global optimization, such as the correlation among different unit operations, data-driven hybrid optimization methods have been developed and applied in mineral-processing plants [13,14]. However, in such optimization approaches, ① the treatment of waste and emissions is usually not included during process optimization including global optimization [15], and ② the process cost is usually not involved as a key factor associated with environmental impacts to evaluate the effectiveness of process optimization.

Tungsten is a strategic/critical metal with a wide range of applications in hard alloys, catalysts, energy storage, and electronic materials [16–18]. However, its primary production from tungsten minerals is very energy intensive and is associated with significant environmental impacts including solid waste, gas, and liquid emissions [19–21]. In this research, tungsten is therefore taken as a representative metal for a case study. The stage of mineral concentrate processing into ammonium paratungstate (APT), which has the greatest environmental impact [18], was chosen as the specific case. The production of APT and the subsequent production of tungsten powder are complex enough to represent the economic and environmental features of other metals. APT is an important intermediate product in the preparation of most tungsten alloys and chemicals, and is produced from tungsten minerals using a hydrometallurgical process. According to an estimation by the US Geological Survey, more than 85% of primary tungsten is produced in Asia [4], as shown in Table S3. The production of APT involves several chemical steps that are associated with the emission of pollutants, including hazardous solid waste with W, arsenic (As), chromium (Cr(VI)), and lead (Pb); wastewater with heavy metals and NH_3-N; and waste gas composed of sulfur oxides (SO_x) and ammonia (NH_3). In China, more than 80% of tungsten is obtained from the mineral scheelite ($CaWO_4$). The traditional process has resulted in severe facility corrosion and environmental pollution due to the use of hydrochloric acid [21]. Although the main process of tungsten mineral treatment is currently based on decomposition with sodium sulfate, sodium hydroxide, or sodium carbonate, the process still results in large amounts of solid, liquid, and gaseous wastes [20,22].

For APT production, one significant development in $CaWO_4$ processing is the replacement of traditional hydrochloride acid leaching with sodium hydroxide decomposition [17,20], which promotes cleaner production [23]. This kind of process optimization was based on an innovation in a specific step—such as leaching, separation, or product conversion—during APT production. For example, ion-exchange technology can be used to replace solvent extraction for extracting tungsten compounds from solution after sodium hydroxide leaching, thus fulfilling the principles of cleaner production [24,25]. Use of a new technology for process optimization is sometimes promoted by new environmental regulations, as has been reported in an industrial investigation of this field [26]. However, as mentioned earlier, the process profit and the waste/emission treatment step are not directly implemented in such an optimization.

In this research, a strategy is proposed that considers the process cost and the material efficiencies—especially these with significant environmental impacts—of the whole process, including the treatment of waste and emissions. Waste/emission treatment is considered as a step within APT production during process optimization and analysis, instead of being investigated separately. By further defining two factors—namely, the cost-effectiveness factor and the pollution level (environmental impact indicator), the proposed strategy is compared with a process optimization that uses only the basic principles of cleaner production.

2. Methodology

2.1. Concept and principles of whole-process pollution control

The process from a resource to a product or intermediate product usually includes conversion, separation/purification, and product preparation, while waste/emission treatment is generally dealt with as a separate stage and is not included in the production process. As mentioned above, the proposed strategy integrates waste/emission treatment and suggests the concept of a "whole process." This strategy takes material efficiency, cost efficiency, and the environmental impact of the whole process into account. Whole-process pollution control (WPPC) is therefore defined as a process optimization method based on identification of the footprints of elements or compounds that potentially present high environmental hazards or impacts using the principles of lifecycle analyses. WPPC takes a further step to implement cleaner production into waste/emission treatment in order to achieve materials/cost/environmental efficiency optimization in the whole process. Minimization of the comprehensive cost of the whole process is achieved by comprehensively integrating the principles of hazardous reagent substitution, atom economic reaction, green separation, reagent recirculation, waste/emission treatment, system optimization, and other technologies, through which national/local/industrial environmental regulations can be fulfilled (see Fig. 1).

WPPC involves the following procedures:

(1) Material and energy flow investigation and mapping to understand the most significant steps for optimization based on a systematic understanding of the distribution and physiochemical properties of potential pollutants, including their transition routes, reaction mechanisms, toxicity, and so forth, throughout the whole process.

(2) Stepwise process intensification and technology innovation to achieve high utilization efficiency of different elements and to minimize pollutant emissions.

(3) System integration and optimization of cost evaluation to determine optimization procedures, considering comprehensive cost minimization under the up-to-date discharge standards of environmental pollutants.

In WPPC, the footprint of an element or compound is monitored and evaluated based on different processing schemes, starting from resources and ending with a corresponding product. As shown in Fig. 1, the application or effectiveness of WPPC optimization requires active feedbacks from the materials and energy flow in different steps, including waste/emission treatment. This involves identifying an optimum process with a low

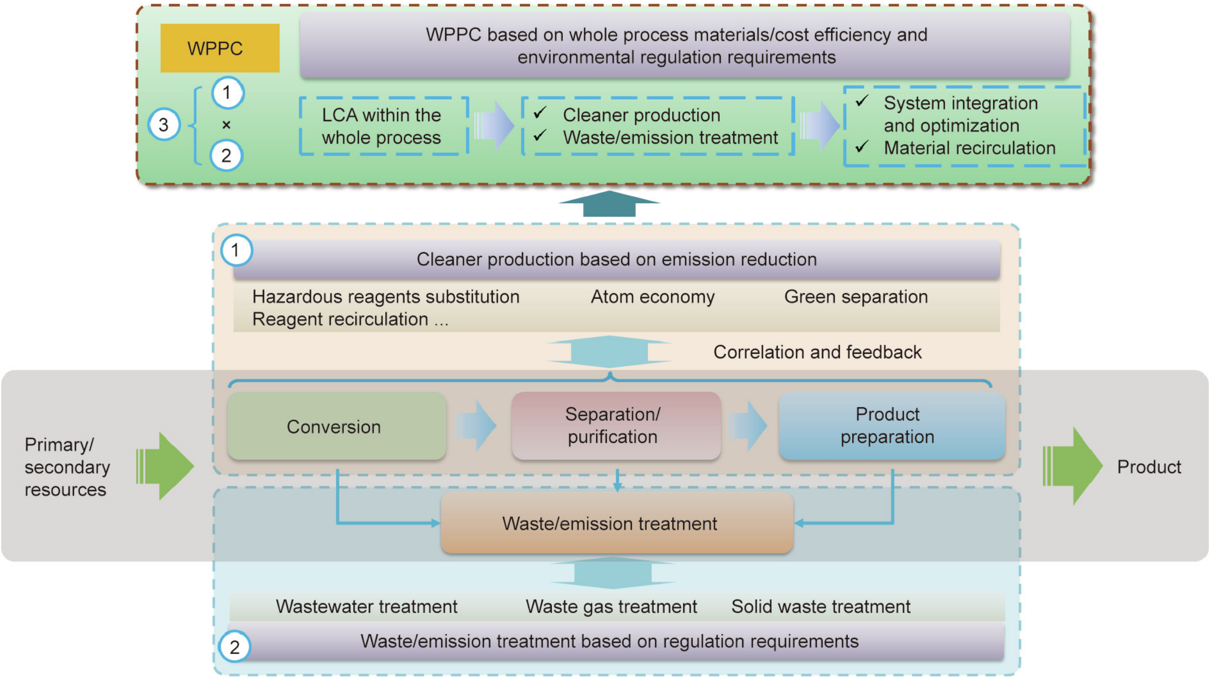

Fig. 1. Principles of WPPC.

comprehensive cost and low environmental impact, especially when new environmental regulations are implemented. The comprehensive cost of the whole process includes the operation cost and waste/emission treatment cost, and can also be extended to include the cost of potential environmental impacts with respect to regulations.

In WPPC optimization, life-cycle assessment (LCA)—specifically of the production process—is important in realizing effective process optimization. The LCA process employs the same principles as cradle-to-grave analyses [25,27], and its inputs and outputs must be predefined. The corresponding materials and energy flows are directly correlated with the optimization of a single step and of the whole process. Application of LCA is helpful in understanding the effectiveness and potential environmental impacts of the optimization of a step of the process. By combining the materials and energy flow information, materials cost, energy cost, and environmental impact cost can be identified for further WPPC evaluation.

As shown in Table 1 and Fig. 2, WPPC implements the principles of cleaner production along with waste/emission treatment in order to optimize a whole production process [28,29]. Consequently, this kind of optimization can increase the materials/cost/environmental efficiencies of the whole process.

2.2. Evaluation methodology

To evaluate the effectiveness of process optimization using the two strategies described above, two parameters are defined: the operational cost-effectiveness factor and the potential environmental impact. The operational cost for APT production including waste/emission treatment is considered, while the asset cost (including land, facilities, maintenance and other factors) is disregarded. Fig. 3 presents the principles by which the operational cost is evaluated. The main difficulties in implementing these principles involve identifying the appropriate factors and correlations to provide a quantitative evaluation. This evaluation of the entire process cost, C_W, is carried out via the following equation:

$$C_W = \sum_i \omega_i C_i = \sum_i \omega_i \left[\sum_j \left(c_{ej} + c_{mj} \right) \right] \tag{1}$$

where ω_i is the correlation factor of a single step in the whole process, C_i is the cost or normalized cost of the specific step, c_{ej} and c_{mj} are the cost related to energy consumption and materials consumption in the specific step j (conversion, separation/purification, product preparation, and waste treatment).

In Fig. 3, blue dashed arrows indicate the possible existence of energy or material exchanges among different steps. Recirculation of materials within the process may significantly reduce the energy and material costs in a specific step, and thus further improve the effectiveness of the process.

2.2.1. Cost-effectiveness factor of each step

As shown in Fig. 3, the whole process, from raw materials to product/intermediate product, can be divided into four steps:

Table 1
Differences between WPPC and traditional pollution-control methods.

	Waste/emission treatment	Cleaner production	WPPC
First time in history	1960s	1970s–1980s	This research
Ranges	Waste/emission treatment	Production process	Production process and waste/emission treatment
Objectives	Satisfaction of regulation requirements	Emissions reduction in production process	Minimization of the entire process cost and satisfaction of regulation requirements
Methods	Wastewater treatment, waste gas treatment, and solid waste treatment	Atom economy, hazardous reagents substitution, green separation, etc.	LCA, cleaner production, waste/emission treatment and system optimization, etc.

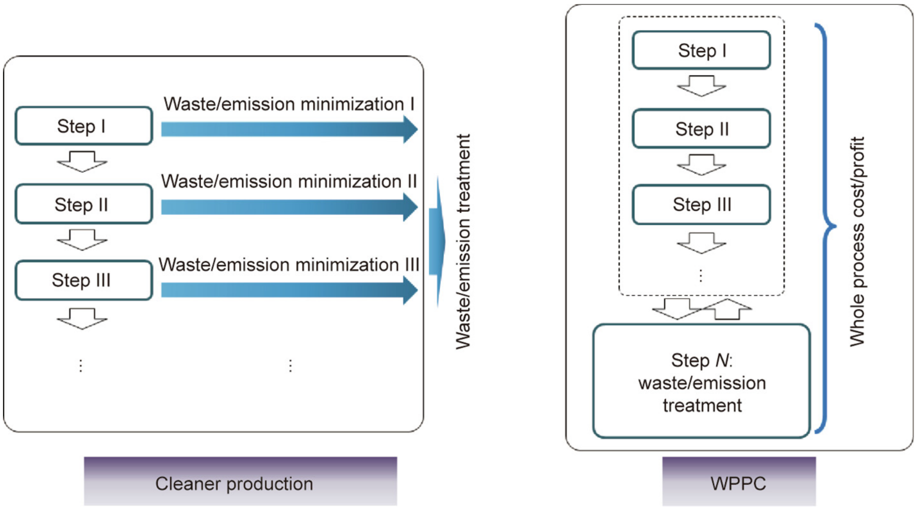

Fig. 2. Comparison of cleaner production and WPPC.

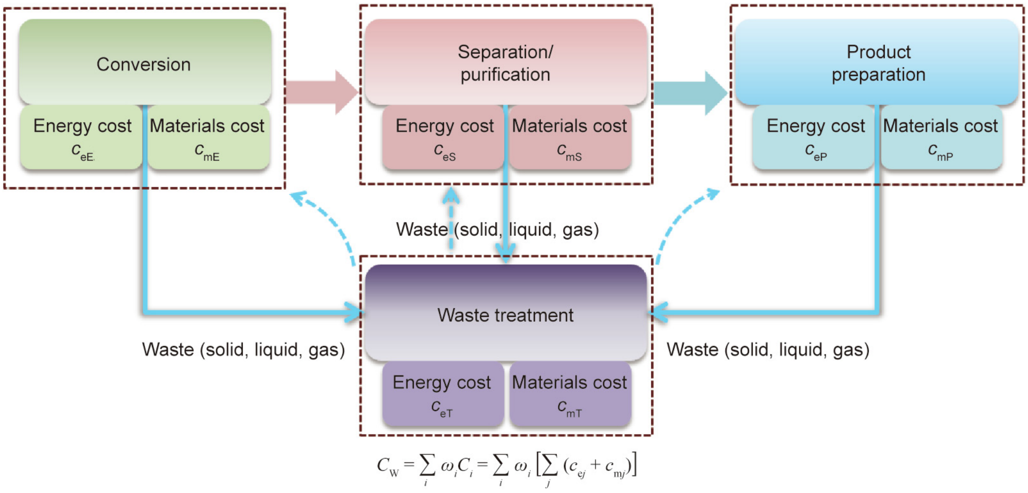

Fig. 3. Diagram of indices and important components in each step for evaluating the entire process cost.

conversion, separation/purification, product preparation, and waste treatment. By mapping the energy and material distribution/flow in each step, the consumption details can be identified for each step of the process. The comprehensive cost is then determined using Eq. (1). During conversion, extraction of the target metals from the corresponding mineral/waste material occurs; this process may include several operation steps based on the process design. In separation/purification, impurities are removed from the target materials. If hydrometallurgical leaching is applied to process the raw materials, solvent extraction or ion exchange could be used in this step. If smelting of the raw materials is applied, purification should be conducted based on high-temperature refining or electrochemical refining. After purification, the product preparation varies depending on the form of the product. The waste treatment step requires significant attention in order to lower the environmental impacts of current primary metal production. The cost can fluctuate significantly in this step, even in the same area, depending on the regulations and management efficiency [25]. The cost indicator of a specific step such as leaching/smelting, C_E, can be calculated by summarizing the cost (including materials and energy) of all the steps in the section. The difference in the cost indicator provides a guideline for evaluating the significance of the process optimization of a specific step, where a decrease in value indicates an improvement in the efficient utilization of materials and energy.

2.2.2. Determination of correlation factors

The correlation factors shown in Eq. (2) indicate the relative importance of different sub-steps during a specific step or throughout the whole process. The correlation factors for unit operation steps are not easily determined, depending on the available experimental data. For simplification, they are usually considered to be equally important, with a correlation factor ω_i of unity. For each step, ω_i is determined based on the following principles:

(1) If recirculation of materials and/or energy exists, ω_i is less than unity, and the value is calculated by integrating the recirculation ratio in the specific step.

(2) If recirculation of the materials and/or energy of the whole process is observed by considering the inputs and outputs of the process, a decrease in all of the correlation factors is expected.

$$\omega_i = \frac{1}{1 + \eta_i} \tag{2}$$

where η_i is the recirculation ratio of energy/materials during a specific step.

2.2.3. Potential environmental impact

During primary metal production, which is usually a metallurgical/chemical process, the environmental impact normally comes from the emission of solid, liquid, and gaseous wastes; furthermore, it is not only determined by the concentrations of hazardous/dangerous components, but also by the volume of the waste streams. To evaluate the potential environmental impact during APT production using different technologies (i.e., P1, P2, and P3), an environmental impact indicator can be defined as follows:

$$\mathrm{PEI_W} = \sum_i \omega_i \mathrm{PEI}_i = \sum_i \omega_i \left(\sum_k x_k m_k \right) \tag{3}$$

where PEI_i is the environmental impact indicator of a single step in the whole process, k indicates solid, liquid, or gaseous waste, and x_k and m_k are the concentration and amount of a hazardous compound in a waste stream, respectively.

With PEI_W, it is possible to understand the destination of hazardous elements in a process and the impact of the process on the environment. The waste or emission usually requires specific treatment or to be landfilled by certified companies.

3. Case study

To evaluate the applicability of the WPPC method, a WPPC assessment was used to analyze the effectiveness of producing APT from tungsten minerals via different processes. Considering the concept of WPPC, the process optimization must optimize the materials and energy flow of the whole production process—that is, from mineral to APT, in this case. As environmental regulations become more stringent, reducing waste generation is highly favorable, and WPPC for process optimization can be important. If an optimization fulfills the concept of WPPC and uses the WPPC assessment method, the optimized process can be called a WPPC process.

Three types of processes were considered for evaluation in this research: P1, a traditional process prior to optimization (a roasting and solvent-extraction process); P2, a process that is optimized using the principles of cleaner production (a pressure-leaching and ion-exchange process); and P3, a process that is optimized using the WPPC strategy (a pressure-leaching and material-recirculation process). During the analyses, the comprehensive cost was based on experiments and investigation in the APT plant of Jiangwu H.C. Starck Tungsten Products Co., Ltd. in Jiangxi Province, China (raw data for calculation is given in Table S4). In this plant, a new technology incorporated with the WPPC strategy by considering NH_3 recycling within the APT production process was recently developed to minimize wastewater emission and reduce NH_3 consumption [30].

In the WPPC strategy, the following procedures were adopted:

(1) The roasting step was optimized by implementing pressure leaching, while the experimental conditions were modified to ensure reusability of waste/emissions.

(2) Solvent extraction was replaced with ionic exchange, while the experimental conditions were modified to ensure reusability of waste/emissions.

(3) Pollutant-containing waste/emissions were reused or recirculated as much as possible.

3.1. Identification of the main pollutants

As mentioned above, it is important to track the distribution of pollutants in order to facilitate the WPPC strategy for process optimization. The compositions of the minerals, residue, and wastewater during the traditional process were specifically analyzed, and are given in Table S5. It was found that Cr, copper (Cu), zinc (Zn), mercury (Hg), nickel (Ni), As, and Pb were likely heavy metal pollutants in the minerals and were concentrated in the residue after leaching. NH_3-N is the main pollutant in the wastewater and may be associated with heavy metals.

Fig. 4 describes the three aforementioned processes for APT production. P2 is optimized in line with the principles of cleaner production, with new technological implementation in comparison with P1. It focuses on improving the yield of W, without considering the treatment of hazardous elements. It should be noted that the implementation of new technologies inevitably increases the investment in the process; therefore, such implementation must sometimes be motivated by new environmental regulations. P3 involves optimizing the whole process based on the WPPC strategy. This optimization not only considers requirements from new environmental regulations, but also considers the cost reduction of the whole process, including waste treatment. As shown in Table S3, the main hazardous elements in the solid waste are heavy metals including As, Ni, Zn, and so on. Molybdenum (Mo) and tungsten are extracted as much as possible. The main pollutants in the wastewater are heavy metals and NH_3-N.

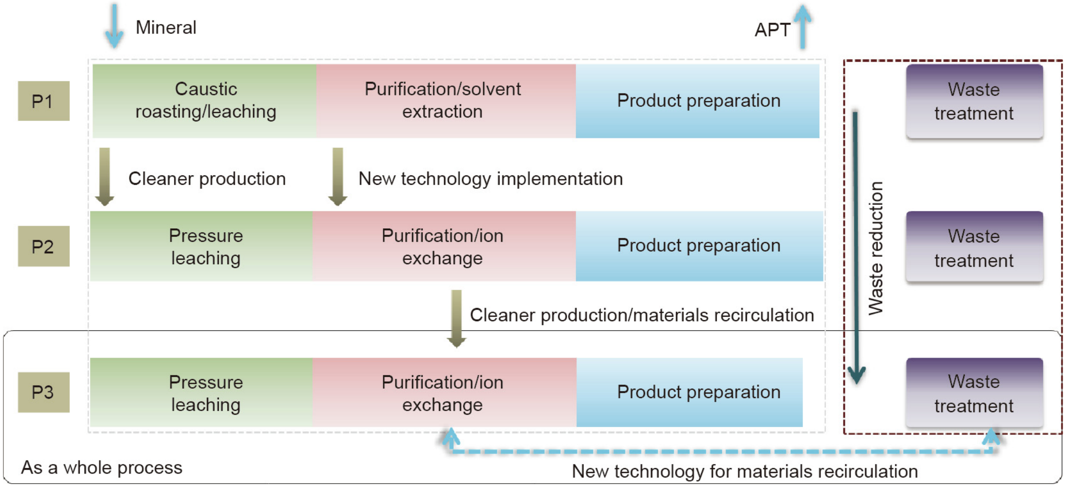

Fig. 4. Comparison of different technologies for APT production.

3.2. Stepwise operational cost of different processes after optimization

The APT production can be divided into four steps: a leaching/extraction step, purification step, product preparation step, and waste treatment step, as shown in Fig. 4 and Fig. S1. The processing costs for APT production were obtained from three identical tungsten production lines in the Jiangxi region of China. Process optimization was implemented, and cost details were obtained after stable operation of the new process for at least six months (an average of one month of operational data is used for this research).

Fig. 5 provides the results. The cost of the leaching/extraction step can be ranked as P3 ≈ P2 > P1 without considering the reduction of mineral consumption (Fig. 5(a)), since both energy and chemical consumption will be increased when the pressure-leaching technology is implemented. In the purification step, solvent extraction is already a mature technology and has been used in APT production in China since the 1940s [31]. Before the solution can be processed via solvent extraction, purification needs to be applied, usually via precipitation of impurities, such as silicon (Si) and As. The main advantages of solvent extraction are its relatively low running cost and mature operation. However, the need for further treatment of the residual solution to reduce the environmental impact has been a significant concern in recent years. Ion-exchange technology was developed to improve the tungsten recovery rate and potentially reduce the wastewater emission. This technology involves adding an anion/cation exchange resin to separate the tungsten from the crude Na_2WO_4 solution so that either Na^+ or WO_4^{2-} is adsorbed/exchanged by the resin. Due to its relatively high energy and chemical consumption, the cost of the ion-exchange step is greater than that of the traditional solvent-extraction technology in P1 (Fig. 5(b)). In P3, the cost can be significantly decreased by chemical recirculation from the waste treatment stage (according to the principles of WPPC) in comparison with P2, although the cost of the purification step in P3 is still higher than that of P1. For the product step (Fig. 5(c)), the cost of P1 is inevitably higher than those of P2 and P3. It is notable that additional crystallization and separation is required in P1 (Fig. S1). Based on the overall cost of the three steps excluding waste treatment, which can be called the operational cost, the ranking of the operational cost is P2 > P3 > P1 (Fig. 5(d)). This result indicates that the operational cost will be high if waste/emission treatment is not included in the whole process of APT production during optimization, even though the extraction selectivity of tungsten is significantly improved in P2. Based on site investigations of different companies using P1 and P2, we determined that the operational cost increase is the main reason why P1 has not been fully replaced, even though P2 features a high tungsten recovery selectivity.

3.3. Cost-effectiveness factor for different processes

In order to evaluate the cost-effectiveness factor for the different processes, C_W is calculated according to Eq. (1). For P1, C_W is determined as follows:

$$C_{WP1} = \omega_{L1}C_{L1} + \omega_{Pu1}C_{Pu1} + \omega_{Pr1}C_{Pr1} + \omega_{Wt1}C_{Wt1} \tag{4}$$

Since material recirculation/reuse was not considered, the recirculation ratio is zero for the leaching step, whereas it is 0.35 in the purification step, which involves solvent regeneration and water recirculation. In this case, $\eta_{L1} = 0$ and $\eta_{Pu1} = 0.35$. The correlation factor ω of each step and the cost-effectiveness factor C_{WP1} can be calculated.

For P2 and P3,

$$\begin{aligned} C_{WP2} &= \omega_{L2}C_{L2} + \omega_{Pu2}C_{Pu2} + \omega_{Pr2}C_{Pr2} + \omega_{Wt2}C_{Wt2} \\ C_{WP3} &= \omega_{L3}C_{L3} + \omega_{Pu3}C_{Pu3} + \omega_{Pr3}C_{Pr3} + \omega_{Wt3}C_{Wt3} \end{aligned} \tag{5}$$

where $\eta_{L2} = \eta_{L3} = 0.13$ due to the regeneration of leaching medium, as listed in Table 2.

Since measuring the recirculation ratio of different materials in accordance with their specific costs is a complex and not practically applicable task, the values given in Table 2 are average recirculation ratios of the corresponding materials; their costs are considered to be the same for the sake of simplicity.

Fig. 6 compares the cost-effectiveness factor of different processes, with a focus on the fractions of different steps. For P2, the cost-effectiveness factor is still high, since both pressure leaching and ion exchange will increase the energy and chemical

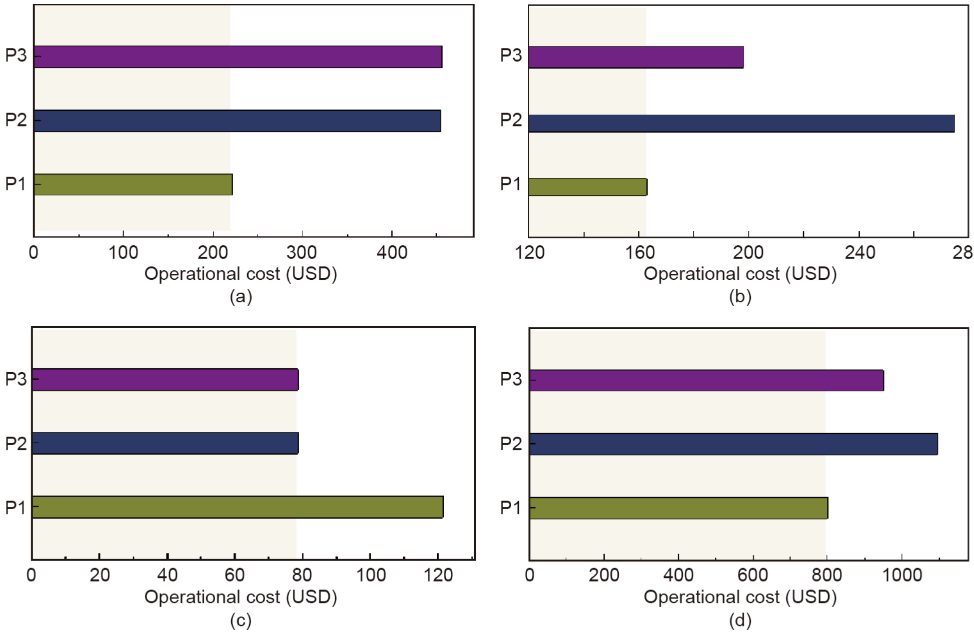

Fig. 5. Cost details of individual steps in different processes. (a) Leaching/extraction step; (b) purification step; (c) product step; (d) operational cost without considering the reduction of mineral consumption.

Table 2
Experimental results for the recirculation ratio and correlation factors of different APT production processes.

Process	Variable	Variable value of different production processes			
		Leaching	Purification	Production	Waste treatment
P1	η_1	0	0.35	0.30	0
	ω_1	1.00	0.74	0.77	1.00
P2	η_2	0.13	0.46	0.30	0
	ω_2	0.88	0.68	0.77	1.00
P3	η_3	0.13	0.46	0.30	0.40
	ω_3	0.88	0.68	0.77	0.71

consumption; however, it is lower than that of P1. Since material recirculation is integrated during cost-effectiveness analyses, the factor is a more suitable marker for reflecting the advantages of a process than the exact cost shown in Fig. 5. Relatively low process cost is that the reason why P1 still exists, especially in places where the environmental regulations are not well implemented. The main environmental impacts from APT production include waste gas (including NH_3 and NO_x), wastewater (containing NH_3, Cl^-, NO_3^-, SO_4^{2-}, and acids and heavy metal ions), and solid waste (including As_2O_5, MoS_3, $Ni(OH)_3$, P_2O_5, and other compounds). Moreover, a significant amount of wastewater with a high COD is generated in the solvent-extraction step when P1 is used for APT production. In P3, the waste/emission treatment step is included within the whole process of APT production according to the principles of WPPC, and significant improvement is evident, especially when comparing the costs of the purification step and the waste treatment step. The reduction in the waste treatment cost is an indication that the environmental impact can be decreased if WPPC is applied for process optimization.

Fig. 6(d) shows a breakdown of the cost-effectiveness factor for different steps. It is clear that C_{WP} cannot be significantly reduced by only introducing cleaner production technologies. By integrating the concept of WPPC, which systematically tracks the material flow of pollutants and increases their circulabilities within the process, the whole process cost can be significantly reduced.

To evaluate the whole-process environmental impact of different processes for APT production in a quantitative manner, the environmental impact indicator was calculated and then compared with that of the traditional salt roasting-solvent extraction process. Based on Eq. (3), the environmental impact indicator for different processes was evaluated using the following ratio:

$$\mathrm{PEI}_{\mathrm{W1}} = \frac{\mathrm{PEI}_{\mathrm{P}i}}{\mathrm{PEI}_{\mathrm{P1}}} \tag{6}$$

As shown in Fig. 7, the environmental impact indicator decreased slightly when ion-exchange technology was implemented into the APT production to replace the salt roasting-solvent extraction process. However, large amounts of wastewater are still generated, especially in the process of resin/membrane regeneration, although waste gas is significantly reduced in comparison with P1 and P2. In the case of P3, in which WPPC principles were introduced for process optimization, the environmental impact indicator greatly decreased (Fig. 7), indicating the importance of inter-circulation and optimization of the flow of materials and energy throughout the whole process. The environmental impact aligns with the cost-effectiveness factor for waste treatment, and it is important to identify the amount of waste material generation during a specific process. In WPPC optimization, it is critical to consider both the environmental impact reduction and the operational cost reduction in order to account for the full decrease in the comprehensive cost of the whole process. Figs. 6

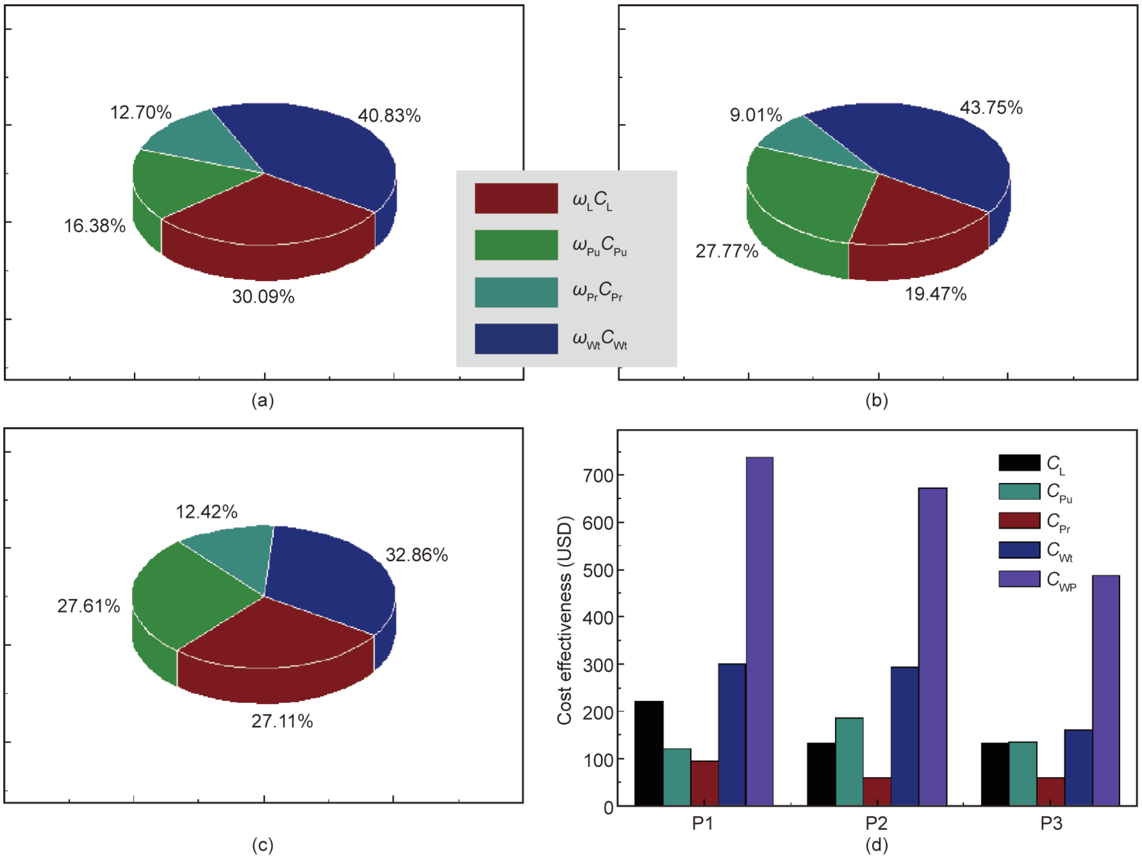

Fig. 6. Comparison of the cost-effectiveness factor. Fractions of different steps in (a) P1, (b) P2, and (c) P3; (d) details of the cost-effectiveness factor.

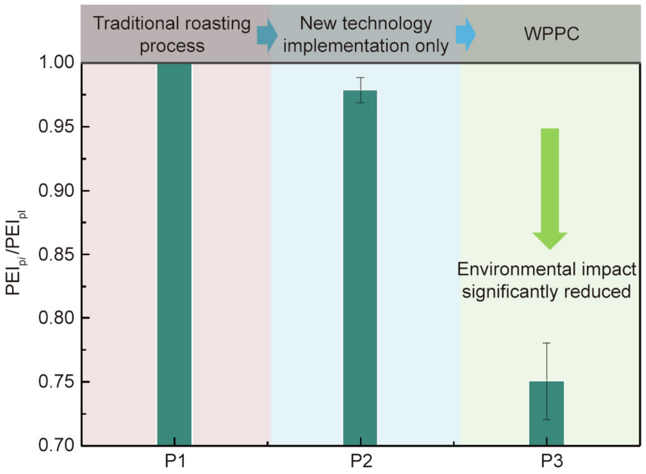

Fig. 7. Whole-process environmental impact indicators of different processes in APT production.

and 7 suggest that WPPC is an effective methodology for the process optimization of a typical hydrometallurgical process. The model developed in this research can be used to evaluate the effectiveness of such a process.

3.4. Discussion

Given the technologies in common use in primary metal production at present, waste generation in the form of three main waste streams (gas waste, wastewater, and solid waste) is inevitable, since the concentrations of rare metals in minerals are usually very low. Cleaner production has been promoted in order to improve material efficiency and recovery as products during process optimization. However, this strategy only focuses on the production stage, as shown in Fig. 2. In the case of APT production, the whole process cost is scarcely influenced by cleaner production, although the W recovery rate is significantly improved (Fig. 5). When WPPC is used as the strategy for process optimization, however, the recirculation of pollutants and associated materials can be promoted; as a result, the whole-process cost, C_P, is considerably reduced. Thus, it is clear that when waste/emission treatment is considered as a step of production (where a "whole process" represents APT production plus waste/emission treatment), process optimization can be stimulated to be more practically viable. In this case, industrial sustainability is likely to be more easily ensured through WPPC than through the implementation of cleaner production.

As another possible stream during production, storage, and transportation, waste chemicals represent a significant environmental hazard. The rare metals found in natural minerals are commonly associated with heavy metals or radioactive metals, such as thorium (Th) in rare earth minerals and As in tungsten minerals, which can become concentrated in waste materials after the target metals are extracted. These hazardous materials can migrate over long distances via wind or groundwater. Inter-city migration of hazardous materials has also been frequently reported, especially in regions with heavy industrial activities [32,33]. These environmental issues can result in health problems, as has been emphasized in recent years [34]. Thus, minimizing the adverse effects of primary metal production and developing a method to ensure the sustainability of the associated industries are of great importance. Cleaner production methods that optimize typical metal production processes have been proposed in order to address these issues. The principles involved in these methods, which include green chemistry, closed-loop techniques, and an atomic economy, have been well accepted and are even applied in the production of organic materials. However, the integration of these methods into mineral processing or the production of intermediate metal products has encountered great difficulties. In the current technological situation, it is not possible to extract all of the elements from minerals to produce additional associated materials. Waste materials—and especially wastewater—require proper treatment, as new environmental regulations are being implemented in China. The main challenges for optimizing metal production processes, among others, are related to the following three driving forces:

(1) Driving force from industry. It is often difficult for a company to realize investment in new facilities or a new technology in a production line. Although the technology may be innovative, the risk of potential profit loss must be considered.

(2) Driving force from the public. Process optimization can sometimes be driven by a newly issued regulation that requires reductions in emission levels for waste materials, or limits the landfilling of certain waste streams. Public awareness is also an important driving force for process optimization that reduces the environmental impact around a plant that produces critical metals.

(3) Driving force from technology improvement. As previously mentioned, ion-exchange technology can significantly improve tungsten extraction selectivity in comparison with the solvent-extraction process. Cleaner production also provides important principles for process optimization; some researchers have even proposed ways to utilize tungsten mineral fully. However, these suggestions are still in the exploratory stages. It is difficult to integrate industrial application with cleaner critical metal production.

Therefore, we propose a methodology for process optimization that involves increasing these driving forces in order to address the challenges described above.

4. Conclusions

This research demonstrated and systematically investigated a WPPC method, and proposed the concept and principles of WPPC. To demonstrate the effectiveness of the WPPC optimization strategies, comprehensive cost and environmental impact were quantitatively evaluated by considering factors such as facility/technology innovation related to the utilization efficiency of different elements in the minerals/raw materials, material and energy flows throughout the whole process, and waste treatment technologies/landfilling cost in view of regulation requirements. The effectiveness of producing APT from tungsten minerals via different processes was discussed. The results show that the cost cannot be significantly reduced by only introducing cleaner production technologies. By integrating the concept of WPPC, which systematically tracks the material flow of pollutants and increases their circulabilities within the whole process, the whole-process cost-effectiveness factor, C_{WP}, and the environmental impact indicator, PEI, can be significantly reduced. This research shows that by considering technological innovation, economic viability, and environmental impact in combination with regulation requirements, WPPC can be more efficient than cleaner production in optimizing a primary metal production process.

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Compliance with ethics guidelines

Hongbin Cao, He Zhao, Di Zhang, Chenming Liu, Xiao Lin, Yuping Li, Pengge Ning, Yi Zhang, and Zhi Sun declare that they have no conflict of interest or financial conflicts to disclose.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.eng.2019.01.010.

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