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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 life-cycle 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