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Distribution Analysis of Precious Metal Catalysts Containing the Rare Precious Metal Palladium

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Keywords: catalytic converter, petroleum refining catalyst, chemical industry catalyst, palladium.

ABSTRACT

Precious metal catalysts are mainly used as catalytic converters in motor vehicles and as petroleum refining and chemical industry catalysts. Platinum (Pt) group metals (PGMs) are mainly used as active materials for catalysts, which contains palladium (Pd). This study used material flow as the framework theory and adopted methods such as review, telephone interviews, and manufacturer visits. The aim was to investigate the distribution of import, manufacture, export, and waste removal of related raw materials and products for precious metal catalysts and Pd contained in Taiwan. The results showed that in 2019, Taiwan imported 1,845,557.5 kg of various precious metal catalysts and exported 653,425 kg. The potential stock in the consumer market was 1,077,448 kg, the amount recycled was 1,077,448 kg, the waste volume was 1,587,229.3 kg, and the abandoned overseas treatment capacity was 259,997 kg. In addition, in 2019, the import volume of Pd contained in various precious metal catalysts in Taiwan was 1413.6 kg, the export volume was 536.4 kg, the potential stock in the consumer market was 411.08 kg, the waste to be recycled was 411.08 kg, the waste volume was 1,403.84 kg, and the overseas disposal amount was 130.93 kg.

The researcher found that Taiwan's annual disposal of waste precious metal catalysts abroad reached 259,997 kg and contained 130.9 kg of Pd. Since the average international Pd price of 2019 was US$1,536.7 dollars per troy ounce, if the waste precious metal catalysts processed outside the country had been domestically recycled and refined, it would have generated an additional profit of US$6,406,756.

This study revealed the material flows of precious metal catalysts containing the rare precious metal Pd. It provides the basis for resource recycling management in related industries and serves as a reference for the government to establish relevant policies.

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INTRODUCTION

Research Background and Motivation

Although reserves of rare precious metals are scarce, they are widely used in high-tech and green energy-related industries. Most advanced countries attach great importance to the extraction of rare precious metals from "urban mines", and recycle large amounts of gold, silver, platinum, etc. from secondary resources every year. According to statistics, it costs about US$40,000 to mine 1 ton of silver (Ag), but only US$1,400 to recover the same amount. It also costs US$250–300 to mine 1 ounce of gold (Au), but US$100 to recover 1 ounce (Shi, 2008).

However, the technical threshold for the recovery and regeneration of rare resources is relatively high, necessitating significant research and development technology and capital expenditure. Currently, in Taiwan, there is a lack of material flow distribution information for various rare precious metals, which increases the uncertainty of investment in recycling and causes relevant enterprises to stagnate. These resources require extensive and systematic investigation to improve the understanding of flow trends for scarce resources.

Research Purposes

Rare precious resources are important for enabling Taiwan to develop electrical machinery and optoelectronics industries and to revitalize traditional industries. At present, no research has considered the material flow analysis of Pd-containing precious metal catalysts. The aim of this study was to use material flow theory and methods, such as a literature review, telephone interviews, and manufacturer visits, to analyze the import, industrial application, waste, reuse, and export of precious metal catalysts. This enabled us to estimate the distribution of Pd in precious metal catalysts in Taiwan and to map the distribution of this rare resource. The study can support manufacturers' future flow management and provide a reference for resource recycling operators to establish successful plants and make suitable investments.

MATERIALS AND METHODS

Current Situation of Rare and Precious Pd

Pd is the forty-sixth element in the periodic table and one of the six platinum (Pt) group metals (PGMs) alongside Pt, rhodium (Rh), iridium (Ir), ruthenium (Ru), and osmium (Os).

According to Raymond et al. (2020), the Norilsk–Talnakh nickel–copper deposit in Siberia and the Bushveld igneous complex platinum-group metal deposit in South Africa are the two largest Pd deposits in the world, accounting for 46% and 39% of the known Pd deposits, respectively. At the end of 2018, These two deposits of Pd reserves totaled about 331 million troy ounces (about 10,300 tons) (Li WF, 2020).

Application of Precious Metal Catalysts

PGMs are mainly used as active materials for catalysts, the main applications of which include automotive catalytic conversion, oil refining, chemical, and pharmaceutical production. Different types of catalysts contain different types of precious metal, but precious metal catalysts mainly contain PGMs. mainly Pt, Pd, and Rh, although a few contain Au, Ag, Ru, and Ir. The concentrations of precious metals ranges from 200 ppm to 100%, and the various catalyst carriers differ. Basic information of various automotive, petroleum, and chemical catalysts is shown in Table 1.

Table 1. Catalysts for automotive, petroleum and chemical catalyst industry applications (source: Dong et al., 2015)

Application field		Catalyst support	Active components	PM contents	Lifespan/year
Automotive	Catalysts Diesel particulate filter	Pt-Rh；Pt-Pd-Rh；Pt Pt-pd	Cordierite monol ith ceramic pellets SiC or cordierite	0.1-0.3% 0.1-0.5%	＞10 ＞10
Oil-refining	Reforming Isomerisation Hydrocracking Gas to liquid	Pt; Pt/Re, Pt/Ir Pt; Pt/Pd Pd; Pt Co+(Pt; Pd; Ru; Re)	Al_2O_3 Al_2O_3, zeolites SiO_2, zeolites Al_2O_3, SiO_2, TiO_2	0.2–1%	1-12
Chemical engineering	Nitric acid H_2O_2 HCN PTA VAM EO KAAP	PtPdRh Pd Pt; Pt/Rh Pd Pd/Au Ag Ru	Gauzes $Al_2O_3Al_2O_3$ or gauzes Carbon granules Al_2O_3, SiO_2 Al_2O_3 Activated carbon	100% 0.3-0.5% 0.1%; 0.5% 0.2-0.3% 1–2% 10-15% –	0.5 1 0.2–1 0.5–1 4 1-5 1-3
Homogeneous	Oxo Alcohols Acetic acid	Rh Rh; Ir/Ru	Homogeneous	500–1000 g/t	1–5
Fine Chemicals	Hydrogenation Oxidation Debenzylation	Pd; Pd/Pt Ru; Rh; Ir	Activated carbon	2–10%	0.1–0.5

Catalytic Converters

Since vehicle exhaust emissions, due to incomplete combustion, contain carbon oxides (COx), hydrocarbons (HCs), and nitrogen oxides (NOx), air quality is rapidly deteriorating, and countries around the world have increasingly formulated emissions standards to limit exhaust gases. Of these, NOx and HCs are the most hazardous because they can easily generate photochemical fog after exposure to sunlight, causing serious air pollution and harming people's respiratory systems (Q. H. Li, 2003). At present, most catalytic converters on the market are three-way catalytic converters, which contain precious metals catalytic converters, which contain precious metals, such as Pd, Pt, and Rh.

Petroleum Refining Catalysts

The oil refining industry is by far the largest industry employing PGM catalysts and uses them in large quantities, particularly Pt and Pt/Rh catalysts for γ-Al_2O_3 substrates in recombination and isomerization processes. Some hydrocracking processes use zeolite catalysts with Pd, and many refineries employ Pd or Pd-Pt catalysts for hydrogenation, decyanation, oxidation, and various other processes (Brumby and Verhelst, 2005).

Chemical Catalysts

According to Hagelüken (2006), chemical catalyst applications include general chemicals such as formation of nitric acid, hydrogen peroxide, hydrocyanic acid, purified terephthalic acid (PTA), and vinyl acetate monomer (VAM). The types of these catalysts vary widely, ranging from Pt-Pd-Rh catalytic mesh (HNO_3; HCN-Andrussow-process) and high-surface palladium powder (H_2O_2-anthraquinone process) to various noble metal combinations on supports (such as oxidation aluminum extrudates) and silica orgraphitized carbon particles. The main metals used for support catalysts are Pd and Pt, with a typical PGM content range of

0.02-0.5%. Rh is mainly used in Pt catalytic mesh about 5-10% alloy. Ru on carbon in the KAAP process for ammonia synthesis from natural gas is a relatively new application. In fine chemicals and pharmaceuticals, Pd-activated carbon powders are most commonly used at typical 5% PGM loadings.

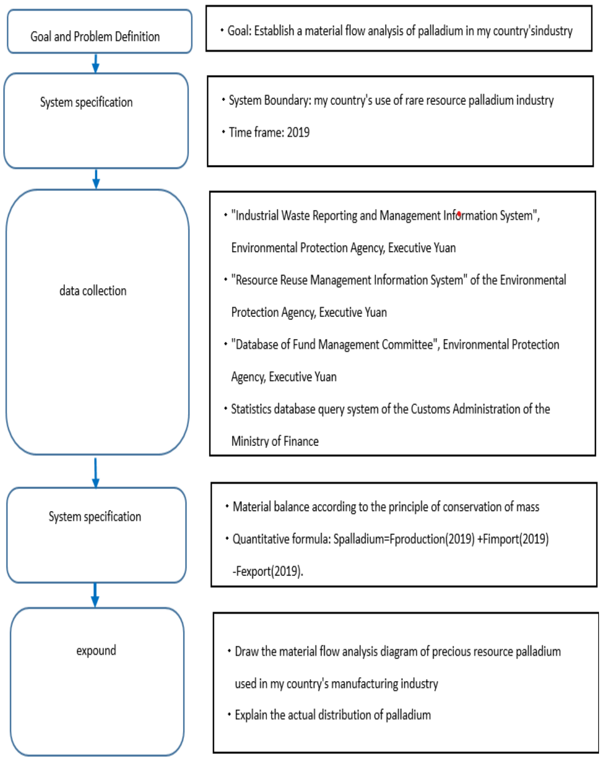


Figure 1. Material flow analysis step architecture

Substance Flow Analysis

Substance flow analysis (SFA) is a material flow analysis method for specific elements. According to Hendriks et al. (2000), the basic SFA includes defining the goals and problem, describing the system, collecting relevant data, conducting material balancing and expounding. Therefore, in this study, Therefore, in this study, we conducted a Pd material flow analysis according to these steps. The analytical process architecture is shown in Figure 1.

To assess the collected data, we used a quantitative estimation equation and employed the principle of mass conservation to conduct mass balancing according to mass flow input = mass flow output. The analog balancing basis was as follows:

SPd = Fproduction (2019) + Fimport (2019) – Fexport (2019).

where S = stock inventory (i.e., the potential waste in the system) and F = flow (i.e., the main input and output factor).

SPd represents the total flow of Pd metal in the system. From the perspective of mass balance, it is assumed that the total amount in the system is the waste potential. Fproduction (2019) represents the production volume of Pd products in the system in 2019, including the manufacturer's own production and commissioned production. Fimport (2019) represents the total amount of Pd imported into the system in 2019. Fexport(2019) represents the total amount of Pd leaving the system boundary in 2019, which includes the export of Pt metal products, and public institutions' or people's output of waste Pd metal. Pd products are mostly exported to other countries through direct exportation, although some of the total amount is scrap Pd exported overseas for processing after the product life cycle ends.

RESULTS AND DISCUSSIONS

Weight Ratios of Pd in various Products

To establish an analytical map of the distribution of Pd-containing products in Taiwan's precious metal catalysts, it was necessary to calculate the pure Pd content of the Pd-containing products. In this study, we used the percentage of Pd in each product as the basis for subsequent estimations.

This section explains the method for calculating the Pd-containing weight ratios of individual products, and Table 2 summarizes the Pd-containing ratios of various precious metal catalyst products.

According to Faisal et al. (2008), the amount of Pd in the catalytic converter of a general vehicle is 200 to 800 μg·g−1, and the average value is 500 μg·g−1, which is equal to 500ppm or 0.05%. Therefore, we used these values as the amount of Pd in automotive catalytic converters.

Based on interviews with representatives of relevant petroleum refining companies and recyclers, we found that the Pd content of petroleum refining catalysts was about 0.194 g/kg, which we took as the value for the Pd content of petroleum refining catalysts.

According to scholar Nicola Della Ca' (2021), Pd can facilitate countless organic transformations (alkylation, arylation, cyclization, hydrogenation, oxidation, isomerization, cross-coupling on both laboratory and industrial scales, cascade reaction, free radical reaction, etc.). Pd may be the most widely used and developed transition metal in catalysis, and natural alkaloids, biologically active compounds, pharmaceuticals, agricultural chemicals and special polymers can be effectively obtained through Pd catalysts. Therefore, we assumed that the most chemical catalysts would be Pd-containing catalysts. According to Hagelüken (2006), the content of Pd in most chemical catalysts is 0.02%-0.5%, so we took the average and estimated that the Pd content in chemical catalysts would be about 0.26%.

In addition, after an interview with the manufacturer, we learned that invalid Pd-containing petrochemical catalyst was imported by the manufacturer for the purpose of refining Pd. It's Pd content was 0.5%.

Table 2. Pd ratios of various precious metal catalysts (source: Faisal et al., 2008; Hagelüken 2006); related businesses)

Precious metal catalyst	Pd ratio	Reference
Catalytic converter	0.05%	Academic literature
Petroleum refining catalyst	0.0194%	Related businesses
Chemical industry catalyst	0.26%	Academic literature
Expired pd-containing catalyst	0.5%	Related businesses

Classification and Names of Imported and Exported Precious Metal Catalysts

Based on data drawn from of the statistical database of the Customs Administration of the Ministry of Finance (CAMF, 2020), we found that product number 38151200005 refered to catalysts with precious metals or precious metal compounds as active substances, such as automobile catalytic converters, petroleum refining catalysts and chemical industry catalysts. In addition, automobiles imported from foreign countries and exported by domestic automobile manufacturers are equipped with catalytic converters, which are also classified as precious metal catalysts.

Analysis on the Import Volume of Precious Metal Catalysts and Their Pd Content (Input Side)

The 2019 import volume of some precious metal catalysts was obtained from CAMF's (2019) database, while that of others was obtained from the Statistics Department of the Ministry of Economic Affairs (MoEE, 2020) and from interviews with industry representatives. In 2019, Taiwan's precious metal catalyst imports could be divided into three major channels: precious metal catalysts with product number 38151200005, totaling 1,523,287 kg; precious metal catalysts attached to foreign imported cars, totaling 322,270.5 kg; and invalid Pd-containing petrochemical catalysts listed under product number 71129990004, with a total import volume of 20,000 kg. Table 3 summarizes the import volumes of precious metal catalyst-related goods and their Pd content in Taiwan in 2019.

Table 3. Imports of precious metal catalysts and their Pd content in Taiwan in 2019.

Product classification	Product name	Import weight (Kg)	Pd content (Kg)
38151200005	Catalytic converter	382,500	191.25
	Petroleum refining catalyst	525,935	102.03
	Chemical industry catalyst	614,852	1,598.62

Table 3(cont.)

Import of complete vehicles	Catalytic converter	322,270.50	161.14
71129990004	Expired palladium-containing petrochemical catalyst	20,000	100
Total		1,865,557.50	2,153.04

After checking the manufacturer registration system of the International Trade Bureau (2019), it was found that the "catalyst using precious metals or precious metal compounds as active substances" with the product number of 38151200005 was divided into three categories, described as follows:

a) *Catalytic converters*

The weight of the catalytic converter used in a car is about 4–7 kg, with the carrier weighing about 1–3 kg. Therefore, the weight of the catalytic converter for each car is about 1.5 kg (Guanming, 2015). In this study, to conduct the relevant data calculation, we assumed that a car had a 1.5-kg automotive catalytic converter load. According to domestic car manufacturers, the catalytic converters for automobiles are imported from abroad, and then attached to automobiles in car factories. According to information from MoEE's (2020) Statistics Office, Taiwan produced about 255,000 vehicles in 2019. Calculated on the basis that one vehicle was equipped with a single catalytic converter, 255,000 catalytic converters (382,500 kg) were imported in 2019. Since each catalytic converter contained 0.05% Pd, 191.25 kg of Pd was imported for domestic car production in that year.

b) *Petroleum refining catalysts*

There are two main petroleum refineries in Taiwan operated by CNPC and Formosa Plastics, respectively. According to interviews with industry representatives, CNPC's import volume of precious metal petroleum refining catalysts in 2019 was about 239,890 kg. Calculated based on the fact that each one contained 0.0194 g% of Pd, about 46.55 kg of Pd was imported. Because the oil and petrochemical products produced by CNPC and Formosa Plastics were very similar, after consulting the Taiwan Stock Exchange Public Information Observatory (TWSE,2020), we found that CNPC's output of various oil and petrochemical products in 2019 was 25,285,728 kg, while Formosa Plastics' output of homogeneous products was 30,153,000 kg. Therefore, we estimated that Formosa Plastics imported about 286,045 kg of precious metals and petroleum refining catalysts in 2019. Calculated based on 0.0194 g% of Pd in each one, the imported Pd was about 55.51 kg. Therefore, in 2019, Taiwan imported about 525,935 kg of petroleum refining catalysts, which contained about 102.03 kg of Pd.

c) *Chemical industry catalysts*

According to CAMF statistics, the import volume of catalysts containing precious metals or precious metal compounds as active substances in 2019 was 1,523,287 kg. After deducting the automotive catalytic converters and petroleum refining precious metal catalysts for 2019, we calculated that the total import of chemical industry catalysts for the year was 614,852 kg. Since the average Pd content was 0.26%, about 1,598.62 kg of Pd was imported.

According to MoEE (2020) statistics, 214,847 vehicles were imported into Taiwan in 2019. Assuming that a car had about 1.5Kg of automotive catalytic converters, the total amount of automotive catalytic converters was 322,270.50 kg, containing about 161.14 kg of Pd.

According to CAMF statistics, product number 71129990004 referred to other products under Section 7112. After interviewing the manufacturer, we learned that it was an invalid Pd-containing petrochemical catalyst imported by the manufacturer for the purpose of refining palladium. According to CAMF data, the import volume in 2019 was 20,000 kg. Calculated based on 0.0194g% of Pd content, the imported Pd was about 100Kg.

The three pipelines together imported 1,865,557.5kg of precious metal catalysts into Taiwan, containing a total of 2,153.04 kg of Pd.

Analysis of Precious Metal Catalyst Manufacturing Volume and Pd Content (Manufacturing Side in the Economy)

After interviewing industry representatives, we discovered that there was no domestic manufacturer of precious metal catalysts for catalytic converters in Taiwan; catalytic converters for domestic vehicles were imported from abroad. To avoid confusion with the catalytic converters of imported vehicles, and to facilitate the mapping of the material flows, we listed catalytic converters integrated into domestic vehicles as domestic manufacturing. According to MoEE (2020) statistic, Taiwan produced about 255,000 automobiles in 2019. If each was equipped with a single catalytic converter, 255,000 catalytic converters were imported in 2019. The weight of the catalyst was about 1.5 kg, and we knew that about 382,500 kg of catalytic converters were imported in 2019. Calculated based on the 0.05% Pd each one contained, the imported Pd was about 191.25 kg, which we regarded as the amount of Pd used in domestic catalytic converter manufacturing.

Both petroleum refining catalysts and chemical catalysts were imported directly from abroad, so there was no domestic production.

Analysis of the Potential Stock of Precious Metal Catalysts (Market Consumption Side in the Economy)

According to MoEE (2020) statistics, Taiwan imported 18,615 vehicles in 2019. Calculated on the basis that each car contained a 1.5 kg catalytic converter, there were 27,922.5 kg of car catalytic converters. According to data drawn from the Environmental Protection Department's (EPD, 2019) pollutant release and transfer register system, the total amount of waste generated by the catalytic converter business in 2019 was about 16,555 kg. We based the estimation of the potential stock of catalytic converters at the consumer end of the market in 2019 on the import volume of catalytic converters in imported automobiles in 2019 + domestic assembly (manufacturing) volume in 2019 - vehicle export volume in 2019 - amount of industrial waste generated in 2019 (322,270.5 kg + 382,500 kg - 27,922.5 kg - 16,555 kg), which made a total of 660,293 kg, containing 330.15 kg of Pd.

The imported petroleum refining catalysts used in Taiwan were imported from abroad, and all were used domestically and were not exported overseas. Hence, the estimated potential stock of petroleum refining catalysts at the consumer end of the market in 2019 was equal to import volume of petroleum refining catalysts in 2019 - amount of industrial waste generated in 2019 (525,935Kg-108,780 Kg), making a total of about 417,155 kg, containing 80.93 kg of Pd.

The chemical industry catalysts used in Taiwan were imported from abroad, but some were re-exported overseas. According to information from CAMF's statistical database, 457,438 kg of precious metal catalysts were exported in 2019. After interviewing exporters, we learned that this amount included about 243,000 kg of waste vehicle catalytic converters. In addition, 16,555 kg of domestic automobile manufacturers' discarded catalytic converter business waste was treated overseas. The remaining 197,883 kg consisted of chemical industry catalysts. Therefore, the estimated potential stock of chemical industry catalysts at the consumer end of the market in 2019 was equal to the import volume of chemical industry catalysts in 2019 - the export volume of chemical industry catalysts in 2019 - the waste generation of chemical industry catalysts in 2019 (614,852 kg - 197,883 kg－752,587.34 kg). Because the value was negative, based on the law of conservation of mass, we assumed that the existing stock of chemical catalysts in Taiwan in 2019 was 335,618.34 kg. Therefore, there was no potential stock of chemical industry catalysts in the market in 2019.

Analysis of the Waste Volume of Precious Metal Catalysts and Their Potential to Be Recycled (Recycling and removing side in The Economy)

According to information from the EPD Resource Recovery Management Fund's management committee, the announcement recoverable system certification quantity of abandoned catalytic converter was 568,996.50 kg in 2019, containing about 284.35Kg of Pd. Based on data from the EPD's (2019) pollutant release and transfer register system, industrial waste catalytic converters were classified as D-2624 waste catalysts containing precious metals (Au, Ag, Pd, Pt, Ir, Rh, Os, and Ru). According to the EPD data, approximately 16,555 kg of waste was generated by the catalytic converter business in 2019, containing about 8.23 kg of Pd.

The potential amount of discarded automotive catalytic converters to be recycled refers to the catalytic converters imported and assembled in 2019 that are still used in the consumer market but have not yet entered the announcement recycling system. Since the service life of automotive catalytic converters is more than ten years (Dong et al., 2015), we assumed that the catalytic converters used in the vehicles on the market in 2019 had not yet reached the disposal stage, so no discarded vehicles enter the recycling system. Therefore, the potential amount of discarded catalytic converters to be recycled in 2019 was equal to the market potential stock in 2019(660,293 kg, containing 330.15 kg of Pd).

According to the EPD's (2019) pollutant release and transfer register system, petroleum refining catalyst business waste consisted of D-2624 waste catalysts containing precious metals (Au, Ag, Pd, Pt, Ir, Rh, Os, and Ru). According to the EPD data, the total amount of catalyst waste generated by the petroleum refining industry in 2019 was about 108,780 kg, containing about 21.1 kg of Pd.

The potential amount of discarded petroleum refining catalysts to be recycled referred to the petroleum refining catalysts imported in 2019 that were still in use in the consumer market and had not yet entered the recycling system. Since waste petroleum refining catalysts did not belong to the announced recovery items, the potential amount of discarded petroleum refining catalysts to be recycled in 2019 was equal to the market potential stock in 2019 (417,155 kg, containing about 80.93 kg of Pd).

Chemical industry catalyst waste was also classified as D-2624 waste catalysts containing precious metals. According to the 2019 EPD data, the total amount of chemical industry catalyst waste was about 752,587.34 kg, containing about 1,956.73 kg of Pd.

The potential amount of discarded chemical industry catalysts to be recycled refered to the chemical industry catalysts imported in 2019 that were still used in the consumer market and had not yet entered the announcement system for recycling. Since discarded chemial catalysts did not belong to the announced recovery items, the potential quantity of discarded chemical industry catalysts to be recycled in 2019 was equal to the market potential in 2019, with a zero value.

Analysis of the Export Volume of Precious Metal Catalysts (Output Side)

The export volume of precious metal catalysts was obtained partly from CAMF's (2019) statistical database, partly from MoEE's (2020) statistics, partly from EPD's (2019) pollutant release and transfer register system, and partly from interviews with industry representatives. The export of precious metal catalysts in Taiwan could be divided into three major channels: precious metal catalysts with export number 38151200005 (457,438 kg); exhausted chemical catalysts with export number 71129990004 (442 kg); and precious metal catalysts attached to exported complete vehicles (27,922.5 kg). Table 4 summarizes the export volume of precious metal catalyst-related goods from Taiwan in 2019 and their Pd content.

Table 4. Taiwan's precious metal catalyst exports and their Pd content in 2019.

Product classification	Product name	Export weight (Kg)	Pd (Kg)
38151200005	Abandoned catalytic converter	259,555	129.78
	Chemical industry catalyst	197,883	514.49
71129990004	exhausted chemical catalysts	442	1.15
Export of vehicles	Catalytic converter	27,922.50	13.96
Total		485,802.50	659.39

Based on data from the ITB's (2019) manufacturer registration system, we found that "catalysts using precious metals or precious metal compounds as active substances," with item number 38151200005, were divided into two categories, as follows:

a) Catalytic converters

According to the EPA's (2019) statistics on the quantity of recycled waste and containers, 379,331 vehicles were recycled in Taiwan. After deducting the export of 112,043 used cars that year, the remaining 267,288 vehicles were sent to used car recycling centers to salvage the catalytic converters. After interviewing the exporters, we learned that all the catalytic converters discarded in Taiwan were exported to foreign countries to extract precious metals, and they were exported under the title of "catalysts using precious metals or precious compounds as active substances" with item 38151200005. These included waste catalytic converters from discarded vehicles, and waste catalytic converters counted as business waste generated by vehicle manufacturers. According to interviews with exporters, the total export volume in 2019 was about 243,000 kg. In addition, according to data from the EPD's (2019) pollutant release and transfer register system, about 16,555 kg of waste catalytic converters were counted as industrial waste in Taiwan in 2019. The total of both types of waste was 259,555 kg, containing about 129.78 kg of Pd.

b) Chemical industry catalysts

According to CAMF's (2019) statistics, the total export weight of "catalysts using precious metals or precious metal compounds as active substances" with article number 38151200005 was 457,438 kg. Based on data from ITB's (2019) manufacturer registration system, we found that only automobile catalytic converters and chemical industry catalysts were included in the export data, and there were no petroleum refining precious metal catalysts. Based on this information, we knew that about 259,555 kg of waste catalyst converters were exported. The rest (197,883 kg) were all exported chemical industry catalysts containing about 514.49 kg of Pd.

According to MoEE (2020) statistics, 18,615 vehicles were exported in 2019. Based on a single 1.5-kg catalytic converter per vehicle, the total export of catalytic converters was about 27,922.5 kg, containing about 13.96 kg of Pd.

These two export pipelines together exported 736,663 kg of Taiwan's precious metal catalysts in Table 4 Taiwan's precious metal catalyst exports and their Pd content in 2019.

Precious Metal Catalysts and Contained Pd Flow Data

After collecting and analyzing the above data, we compiled the previously described data into a table for inputs (imports), manufacturing in the economy, market consumption in the economy, recycling, and removal in the economy, and a table for outputs (exports and overseas processing). They are shown in table 5.

Analysis of the Distribution of the Rare Precious Metal Pd in Precious Metal Catalysts

In recent years, the output of rare precious metal mines has steadily declined, and most developed countries regard precious metals as strategic resources. Because the mineral purity of rare precious metal waste is higher than that of primary minerals, it is referred to as "urban mines." Since Pd is used in many important basic and technological industries, and the price of Pd has greatly increased in recent years, the efficient recovery or reuse of rare precious metals not only provides a stable supply for domestic use but also avoids the risk of over-reliance on supplies from other countries.

SFA can be used to investigate the distribution of a substance in an environmental ecosystem, helping management decision-makers to formulate relevant management policies.

Due to the scarcity of Pd, its storage and distribution are limited to a few specific countries, so Taiwan is forced to rely on imports. Therefore, to ensure that this rare precious metal can stably meet industry needs at a reasonable price, a material flow analysis of Pd is vital.

In this study, based on the previously collected data, we conducted quantitative analysis to determine the distribution of precious metal catalysts and the Pd they contained in Taiwan in 2019.

According to Figures 2 and 3, there were three import pipelines for precious metal catalysts in 2019: "catalysts using precious metals or precious metal compounds as active substances" with import number 38151200005 (1,523,287 kg), catalytic converters attached to imported new cars (322,270.50 kg), and invalid Pd-containing petrochemical catalysts with import number 71129990004 (20,000 kg). Together, they contained a total of 2,153.04 kg of Pd.

The consumer market side of the economic system was divided into three categories: market potential stock volume of catalytic converters (i.e., the import volume of catalytic converters deducted from the domestic assembly volume, and then deducted from the export volume and business waste volume, totaling 660,293 kg); market potential stock volume of petroleum refining catalysts (i.e., the import volume of petroleum refining catalysts deducted from the business waste volume, totaling 417,155 kg); and market potential stock volume of chemical industry catalysts (i.e., the import volume of chemical industry catalysts deducted from the export volume and business waste volume, totaling 0 kg).

The recycling and removal stage was also divided into three categories. The first was the quantity of industrial waste of catalytic converters (about 16,555 kg), and the potential quantity of discarded catalytic converters to be recycled was equal to the market potential stock (660,293 kg), and the annual recoverable system audit and certification quantity of catalytic converters was 568996.5 kg. The second was the petroleum refining catalyst business waste volume (108,780 kg), and the potential quantity of discarded petroleum refining catalysts to be recycled equaled the market potential stock (417155 kg). The third was the chemical industry catalyst business waste volume (752,587.34 kg), and the potential quantity of discarded chemical industry catalysts to be recycled equaled the market potential stock (0).

The outputs were also divided into three categories. The first was "catalysts using precious metals or precious metal compounds as active substances" with export number 38151200005 (i.e., precious metal catalysts, 457,438 kg). The second was "invalid Pd catalysts" with export number 71129990004 (442 kg). The third was the export volume of new car catalytic converters (27,922.5 kg). Together, they contained 743.41 kg of Pd.

Table 5. 2019 Taiwan Precious Metal Catalyst and Its Contained Palladium Flow Data Collection at Various Stages.

Stage	Product name		Weight (Kg)	Data Source	Palladium Content (Kg)
Input side (import)	**1.**38151200005 Precious metal catalyst (1523287Kg)	**2.**Catalytic converter	382,500	Ministry of Finance Customs Department、Statistics Office of the Ministry of Economic Affairs	191.25
		3.Petroleum refining catalyst	525,935	Relevant industry, Data estimation	102.03
		4.Chemical industry catalyst	614,852	**1－2－3**	1,598.62
	5.Import volume of catalytic converter for vehicle		322,270.50	Statistics Office of the Ministry of Economic Affairs	161.14
	6.71129990004 Expired palladium-containing petrochemical catalyst		20,000	Ministry of Finance Customs Department	100
Economic system (manufacturing side)	**7.**Domestic assembly of automotive catalytic converter		382,500	Statistics Office of the Ministry of Economic Affairs	191.25
Economic system (market consumer side)	**8.**Market potential stock of catalytic converter		660,293	**2＋5－12－26**	330.15
	9.Market potential stock of petroleumrefining catalyst		417,155	**3－19**	80.93
	10. Market potential stock of chemical industry catalyst		0	**4＋11－21－25**	0
	11. Market stock of chemical industry catalyst in 2019		335,618.34	**21＋25－4**	872.61
Economic system (recycling and removal side)	**12.**Business waste amount of catalytic converter		16,555	Environmental Protection Department Pollutant Release and Transfer Register System	8.28
	13. Potential amount of discarded catalytic converters to be recycled		660,293	**＝8**	330.15
	14.2019 annoucement recoverable system certification quantity of abandoned catalyst converter		568,996.50	Environmental Protection Department	284.35
	15. Recycling amount of abandoned catalyst converter		400,932	**14-27**	200.47
	16. Storage Amount of abandoned catalytic converter		157,932	**14-27-22**	78.97
	17. Business waste amount of petroleum refining catalyst		108,780	Environmental Protection Department Pollutant Release and Transfer Register System	21.10
	18. Potential amount of discarded petroleum refining catalyst to be recycled		417,155	**＝9**	80.93
	19. Business waste amount of chemical industry catalyst		752,587.34	Environmental Protection Department Pollutant Release and Transfer Register System	1,956.73
	20. Potential amount of discarded chemical industry catalyst to be recycled		0	**＝10**	0
Output side (export and overseas processing)	**21.**38151200005 Precious metal catalyst (457438 Kg)	**22.**Abandoned catalytic converter	243,000	Related industry	18.28
		23. Business waste of catalytic converter	16,555	Environmental Protection Department Pollutant Release and Transfer Register System	8.28
		24. Chemical industry catalyst	197,883	**21－22－23**	514.49
	25.71129990004 Expired chemical catalyst		442	Related business	1.15
	26. Export volume of new car catalytic converter		27,922.50	Statistics Department, Ministry of Economic Affairs	13.96
	27. Amount of catalytic converters recovered for export of complete vehicle		168,064.50	Environmental Protection Department	84.03

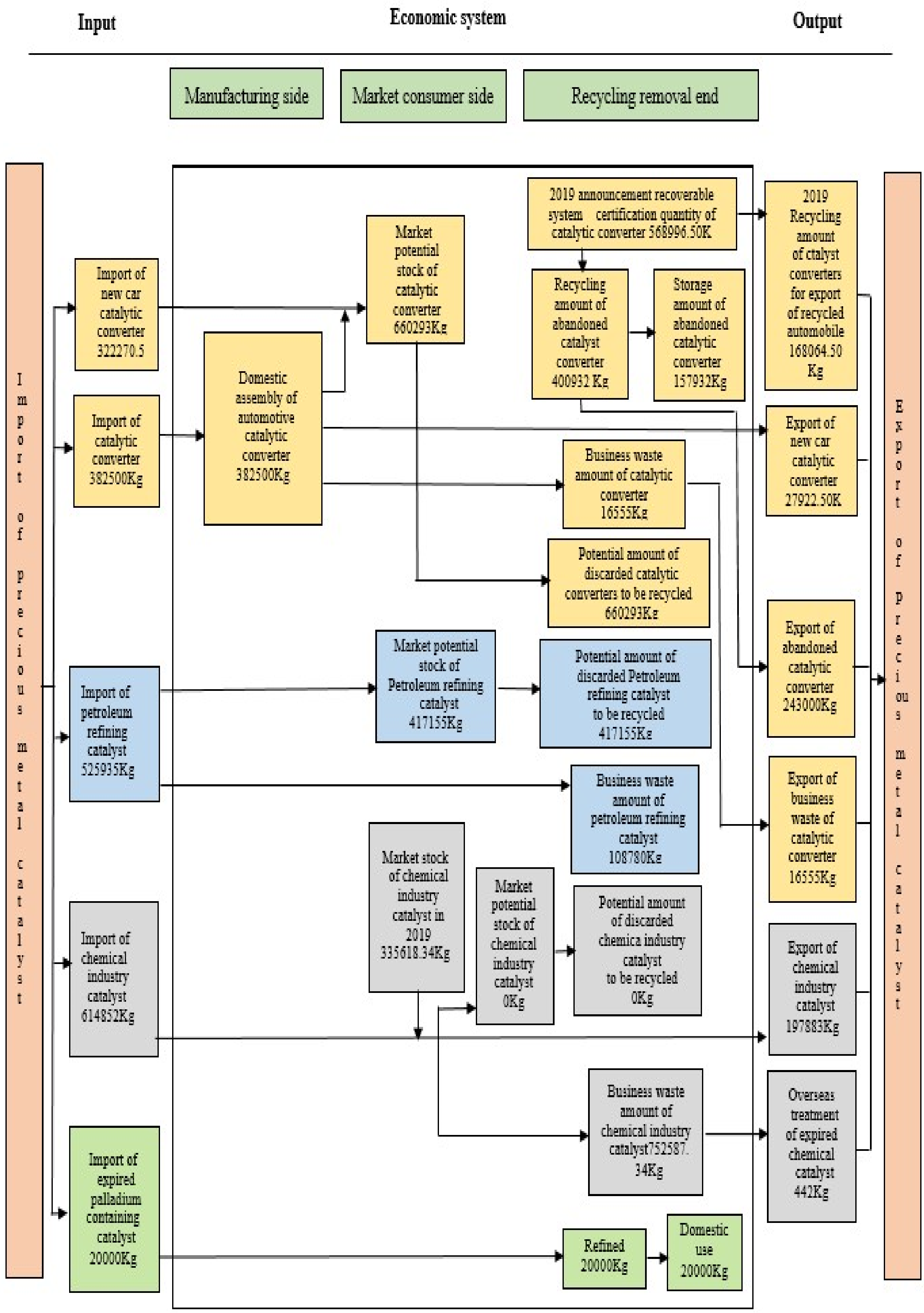
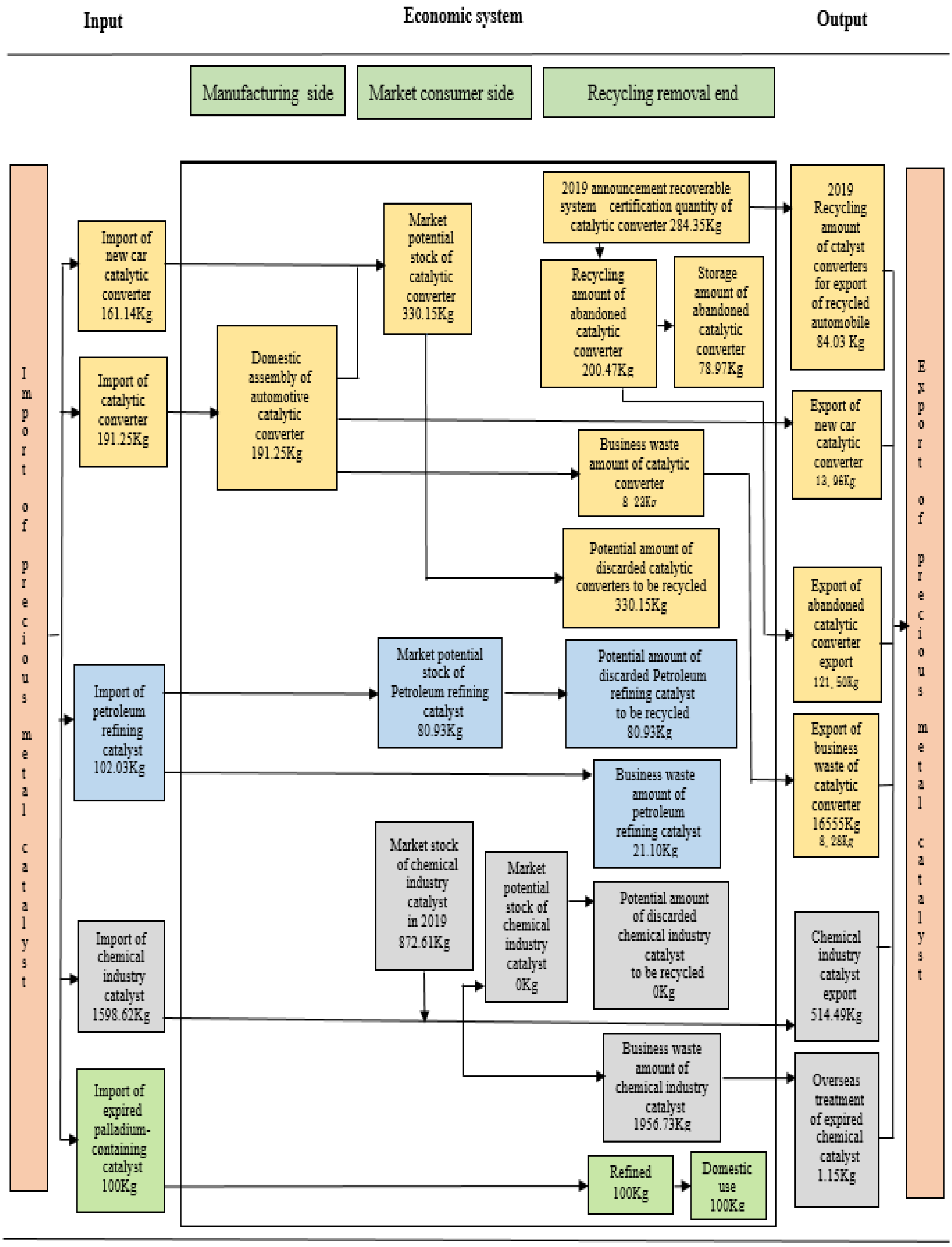


Figure 2. Mass flow analysis of precious metal catalysts in Taiwan in 2019.



Rare and precious metal palladium weight ratio: automotive catalytic converter: 0.05%, petroleum refining catalyst: 0.0194%, chemical industry catalyst: 0.26%, expired palladium-containing catalyst:0.5%

Figure 3. Mass flow analysis of palladium containing precious metal catalysts in Taiwan in 2019.

This study conducted mass balance for the material distribution of catalytic converters, petroleum refining catalysts and chemical industry catalysts in 2019 to confirm that the total input was equal to the total output:

a) *Catalytic converters*

Import of catalytic converter (322,270.5 Kg) + Import of new car catalytic converter (382,500 Kg) = Potential amount of discarded catalytic converters to be recycled (660,293 Kg) + Export volume of new car catalytic converter (27,922.5 Kg) + Export of catalytic converter buiseness waste (16,555 Kg)

b) *Petroleum refining catalysts*

Import of petroleum refining catalyst (525,935 Kg) = Potential amount of discarded petroleum refining catalyst to be recycled (417,155 kg) + Petroleum refining catalyst business waste (108780 Kg)

c) *Chemical industry catalysts*

Import of chemical industry catalyst import (614,852 Kg) + Chemical industry catalyst market stock (336,518.3 Kg) = Potential amount of discarded chemical catalyst to be recycled (0 kg) + Chemical industry catalyst business waste (752,587.3 Kg) + Export Chemical industry catalyst (197,883 Kg)

Analysis of the Disposal of Waste Precious Metal Catalysts in Taiwan

The preceding analysis shows that waste precious metal catalysts can be roughly divided into three categories: catalytic converters, petroleum refining catalysts, and chemical industry catalysts.

There are several regulations or policies to govern the disposal of waste precious metal catalysts in Taiwan. On March 28, 1997, Taiwan amended and announced Article 10-1 of the Waste Disposal Law (later revised to Article 16-1), The EPD successively established eight fund management committees, covering waste general articles and containers, waste motor vehicles, waste tires, etc. These committees provided guidance on and monitored the clearing of recyclable items and containers in various announcements. Later, the eight foundation management committees were transferred to the EPD and became the resource recovery management fund management association. This association annually announces the quantity of certified catalyst converters in waste recycling systems.

Waste precious metal catalysts produced by various business units count as industrial waste, the management of which is based on the Waste Disposal Law 1974. When a business starts operating, it should review the clearance plan previously submitted for approval in accordance with Article 31 of the Waste Disposal Law, and check whether it complies with Article 12 of the Implementation Rules of the Waste Disposal Law. The storage of industrial waste is regulated according to the Industrial Waste Storage and Clearance Treatment Methods and Facility Standards stipulated in Article 36 of the Waste Disposal Law. Article 6 regulates storage methods for general industrial waste, and Article 7 regulates storage methods for hazardous industrial waste. Article 28 of the Waste Disposal Law regulates the removal and disposal of industrial waste and its reuse. Industrial waste may be disposed of by self-removal and treatment, joint removal and treatment, entrusted removal and disposal, and other methods approved by the central competent authority (EPD). The waste baseline information and clean-up flow must accord with Article 31, which states that business units shall provide industrial waste disposal plans, declarations, and delivery forms. In addition, transportation equipment should have a real-time tracking system (GPS). For the proper clean-up of industrial waste, Articles 36 and 42, respectively, stipulate the storage and cleaning methods for industrial waste and the management of private clearing and disposal institutions.

For the export and import of industrial waste, Article 38, Paragraph 1, of the Waste Disposal Law clearly stipulates that “the import, export, transit, and re-export of industrial waste shall be approved by the municipal or county (city) competent authority before it can be carried out; hazardous industrial wastes should first be approved by the central competent authority.” However, this is not the case for the “industrial waste required for industrial materials,” according to the EPD, as announced by the central competent authority (EPD) after consultation with the target industry competent authority (MoEE). “Industrial waste required for industrial materials” comprises 15 major types, including waste wood, waste plastics, waste copper, and waste catalysts. The import and export of these industrial materials do not require approval documents. Precious metal catalysts are one of these 15 major types of industrial waste announced by the EPD. They can be imported and exported without permission, according to industrial materials regulations.

For more than 20 years, Taiwan has achieved remarkable recycling results. Precious metal catalyst and its contained Pd flow data in Taiwan is shown in Table 5. The 2019 recoverable system certification quantity of abandoned catalyst converters was 568,996.50 kg, and the recycled quantity of abandoned catalyst converters was 400,932 kg. In addition, according to industrial waste management information system data, 53,000 kg of disposed precious metal catalysts were recycled in 2019, and the storage difference between 2018 and 2019 was 609,000 kg. However, 753,000 kg of disposed precious metal catalysts were recycled in 2018, and the storage difference between 2017 and 2018 was -356,000 kg. This phenomenon may be due to Pd recycling manufacturers deciding that because the annual average price of Pd had risen from US$869 per ounce in 2017 to US$1,028 in 2018, they should quickly extract Pd from the precious metal catalysts in stock to make more profit. However, in 2019, the price of Pd rose even further, so they became reluctant to sell it and preferred to store precious metal catalysts and wait for better prices before processing them.

The recovery of precious metals from spent catalysts has advantages, including a simple process, a short production cycle, and low investment and environmental impact compared with mining production. However, the concentration of PGMs in spent catalysts varies from type to type, and catalyst carriers for different applications are also diverse. Among recycling technologies, the most practical concentrate precious metals using pyrometallurgical or hydrometallurgical processes. Pyrometallurgical processes are mainly divided into two methods: metal trapping methods and matte trapping methods.

Iron (Fe) is one of the most promising collectors due to its low-costing and strong chemical affinity with PGMs to form a solid solution. Plasma melting technology is most extensively used method for recycling PGMs from spent catalysts, concentrating PGMs in the molten the iron phase. The Fe-PGMs alloy and slag can be separated easily because of their large densities difference (6.0~7.0 g/cm3 versus 3.0~3.5 g/cm3). Through industrial test on plasma melting iron capture technology, therecovery rates of Pt, Pd and Rh were over 98%, 98% and 97%, respectively(He et al., 2016). Plasma melting technology has the advantages of high recovery rate, short process, high efficiency, no generation of wastewater and gas. However, it is high energy consumption and works at high temperature (> 2,000 °C). The high temperature can reduce silicon dioxide into monatomic silicon and form ferrosilicon alloy. However, the ferrosilicon alloy is extremely difficult to dissolve, and resulting recovery of PGMs challenging. To solve this problem, Dong et al. (2014) investigated PGMs recovery by the iron trapping method based on solid state. The results showed that Pt, Pd, and Rh recovery rate were 98.6%, 91.7% and 97.6%, respectively when the reducing temperature was only 1,220 °C. They found PGMs transformed to atomic state or atomic cluster and bonded with free electrons in iron metal during reduction process. The iron atoms kept together and formed crystal nucleus particle, and the crystal grain grew up, after which the iron containing PGMs can be recovered by ball grinding-magnetic separation. Another method uses nickel and sulfur as collectors to smelt spent catalysts in the presence of sodium carbonate (Na_2CO_3) and sodium tetraborate ($Na_2B_4O_7$) at 1,050°C for 30 min, achieving Pt, Pd, and Rh recovery of 90%, 93%, and 88%, respectively (You et al., 2016).

Hydrometallurgical processes use selective leaching reagents such as cyanide, aqua regia, and strong acids. Oxidizing agents such as hydrogen peroxide (H_2O_2), oxygen (O_2), chlorine (Cl_2), and iodine (I_2) are also added to increase leaching efficiency. The recovery rate of PGMs can be heightened by cyanide leaching, but it requires a high temperature, and cyanide is highly toxic. This situation is similar to aqua regia leaching. High leaching efficiency can also be achieved using environmentally friendly leaching agents such as hydrogen chloride (HCl) / sulphuric acid(H_2SO_4) + H_2O_2/copper (II) ($Cu2^+$) under optimal conditions. In general, the recovery rate for Pd is 95–99% (Dinga et al., 2019).

Almost all spent automobile catalytic converters in Taiwan are exported overseas for recycling. When we consulted precious metal catalyst recycling and reprocessing manufacturers, they explained that car recycling companies could not guarantee the content of precious metals in automobile catalytic converters, thus causing instability in the amounts of precious metals recycled and refined by manufacturers. Due to cost considerations, almost all spent automobile catalytic converters in Taiwan are exported to foreign countries for processing and refining precious metals.

Taiwan's precious metal resource recycling market is quite small, and it is difficult to achieve mass production on a large economic scale. In recent years, many aspects of domestic industry have moved abroad, and structural reforms have resulted in a lack of precious metal resources and materials. The main potential risk of disposing of spent precious metal catalysts containing Pd abroad is the low utilization rate of related equipment in Taiwanese recycling centers, potentially leading to higher operating costs. Another potential risk is that Pd is an important strategic reserve metal in many countries, and the disposal of large amounts of waste precious metal catalysts overseas may lead to insufficient domestic reserves for the development of important national industries.

Due to the low natural abundance of Pd and spent hydroprocessing catalysts have been classified as hazardous wastes by the Environmental Protection Agency in the USA, it is common for Pd to be recovered and refined from different types of spent catalysts worldwide. Pyrometallurgical processes require large amounts of energy and investment and cause environmental pollution. In contrast, hydrometallurgy facilitates small-scale processing at the expense of mild reaction conditions and low energy consumption. Therefore, hydrometallurgical treatment is usually chosen as the preferred recovery method. Hydrometallurgical techniques are based on either support dissolution or precious metal dissolution. The first technique requires large amounts of reagents, while the second is based on aggressive reagents and, sometimes, high-pressure leaching, which is complicated and produces significant quantities of wastewater containing environmentally unsafe, highly corrosive, and costly chemical solvents. Therefore, reducing environmental pollution may be the only potential benefit of disposing of waste precious metal catalysts containing Pd abroad.

The current status of automobile recycling and clearance is overseen by the Recycling Management Fund Management Committee, and the relevant recycling system processors are responsible for the recycling and clearance of the waste vehicle channel. In addition, the public may engage with the waste motor vehicle reporting mechanism to ensure a complete recycling system. Overall, the system has achieved commendable results.

To actively promote the reuse of industrial waste, the EPD comprehensively revised the Waste Disposal Law in 2001. The MoEE officially became the management unit for recycling industrial waste. Under the relevant laws and regulations on waste management in Taiwan, the various channels for industrial waste recycling include publicized reuse, licensed reuse, public and private waste treatment for waste reuse, and a joint cleaning and removal system for waste reuse. The MoEE's Industrial Bureau regards the resource recycling industry as a complete industry; actively promotes, guides, and assists its growth and development; and aims to increase the number of manufacturers investing in resource recycling year on year. In 2019, 1,725 manufacturers contributed to various resource regeneration pipelines, and the number increased to 1,958 in 2022. At present, there are four relatively large-scale precious metal catalyst processing and recycling companies: Sus Recycle Technology Inc., Super Dragon Technology Inc., Solar Applied Materials Technology Inc., and Taiwan Tanaka Kikinzoku Kogyo Inc. Of these four companies, Solar Applied Materials Technology Inc. is the largest, with sound operations, excellent technology, and internationally recognized certificates.

CONCLUSIONS AND SUGGESTIONS

According to statistics from Johnson Matthey, the world's largest processor and distributor of PGMs, the Pd shortage in 2019 was 950,000 ounces, compared with 208,000 ounces in the same period in the preceding year (W. Li, 2020). Clearly, Pd demand exceeded annual production, and Pd is not easily extracted from raw ore as a by-product of mining, so prices have increased in recent years. This research focused on the material distribution of Pd contained in precious metal catalysts in Taiwan in 2019. The results showed that in 2019, the import volume of various precious metal catalysts in Taiwan was 1,845,557.5 kg, the export volume was 653,425 kg, the potential stock of the consumer market in the economic system was 1,077,448 kg, the waste to be recycled was 1,077,448 kg, and the waste volume was 1,587,229.3 kg, the abandoned overseas treatment capacity was 259,555 kg. In addition, in 2019, the import volume of Pd contained in various precious metal catalysts in Taiwan was 1,413.59 kg, and the export volume was 536.42 kg. The potential stock in the consumer market in the economic system was 411.08 kg, the waste to be recycled was 411.08 kg, the waste volume was 1,403.84 kg, and the abandoned overseas treatment capacity was 130.93 kg. This research found that Taiwan's annual waste of precious metal catalysts in 2019 was 259,997 kg, including 130.93 kg of Pd. Based on the average international Pd price of US$1,536.74 per troy ounce, if the waste precious metal catalysts processed overseas can be stored in domestic recycling and re-refining centers, it will create an additional profit of US$6,469,626.

Although there are many precious metal recycling centers in Taiwan, most of them are small- and medium-sized enterprises, which are often unable to effectively recycle waste containing precious metals due to financial and technical problems. However, the distribution of various types of waste containing precious metals in Taiwan is unknown, so recyclers are afraid to invest in the research and development of appropriate technologies. As a result, the purity and quality of precious metals cannot be certified by international organizations. Taiwan should follow the example of Japan and other advanced countries by establishing a rare precious metal material flow database to grasp the import, export, and market flows of various rare precious metals, and it should establish a national reserve stock to reduce domestic industrial production risks caused by major international events.

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貴金屬觸媒及所含稀貴金屬鈀物質流佈分析之研究

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摘 要

貴金屬觸媒主要包括機動車輛的觸媒轉化器、石油煉製觸媒及化工觸媒。。本研究針對台灣貴金屬觸媒及其所含鈀金進行相關原料及產品進口、製造、出口以及廢棄物清除之流布調查，結果顯示 2019 年臺灣各式貴金屬觸媒進口量共 1,845,557.5 Kg，出口量為 653,425 Kg，經濟體系內之消費市場潛在存量為 1,077,448 Kg，廢棄待回收量為 1,077,448 Kg，廢棄量為 1,587,229.3 Kg，廢棄境外處理量為 259,555 Kg。另外，2019 年台灣各式貴金屬觸媒中所含鈀金之進口量為 1413.59 Kg，出口量為 536.42 Kg，經濟體系內之消費市場潛在存量為 411.08 Kg，廢棄待回收量為 411.08 Kg，廢棄量為 1403.84 Kg，廢棄境外處理量為 130.93 Kg。本研究發現若能將境外處理之廢棄貴金屬觸媒在國內回收再提煉，以當年度國際鈀金價格平均 1,536.74 美元/盎司計算，將可多創造出 6,469,626 美元之利潤。

本研究揭示了貴金屬催化劑及其所含稀有貴金屬鈀的物質流向。除了為相關回收行業的資源回收管理提供依據外，還可以為政府制定相關政策提供參考。