The Automotive Exhaust Catalyst: Structure, Types, Function and Recycling Methods

A. Rabieifar[1,2,*], A. Shariatmadari[2], M. Ahmadi[2], H. Sabet[3,5], A. Askari[4,5]

[1]Department of Materials Engineering, ST.C., Islamic Azad University, Tehran, Iran.
[2]Centre of Applied Science & Technology Parts Makers of Fajr-e-Misagh,
University of Applied Science & Technology, Tehran, Iran.
[3]Department of Materials Engineering, Ka.C., Islamic Azad University, Karaj, Iran.
[4]Department of Mechanical Engineering, Ka.C., Islamic Azad University, Karaj, Iran.
[5]Institute of Manufacturing Engineering and Industrial Technologies, Ka.C., Islamic Azad University, Karaj, Iran.
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Abstract

The main function of automotive exhaust catalysts is to reduce the emission of toxic gases such as hydrocarbons, carbon monoxide, and nitrogen oxides, and convert them into carbon dioxide, nitrogen, and water vapor. Exhaust catalysts are divided into two types: two-way and three-way, which can be used in diesel and gasoline engines, respectively. The possibility of forming harmful compounds such as hydrogen sulfide and ammonia in these catalysts, as well as the possibility of increasing their temperature to more than the permissible limit (750˚C), leads to a reduction in the useful life and wear of the catalysts. Catalyst recycling is essential due to the use of platinum group metals (PGMs), as well as the growing market for these metals and their limited global resources. Catalyst recycling processes are divided into two categories: pyrometallurgy and hydrometallurgy.

Keywords: Exhaust Catalyst, Recycling, Platinum Group Metals.

1. Introduction

Since road transport accounts for more than 90% of all environmental pollution, it is crucial to develop solutions that reduce harmful substances in vehicle exhaust gases. The operation of most vehicle engines is based on the combustion of fossil fuels. Therefore, the primary solution to reduce the emission of toxic gases in the exhaust is to optimize the combustion process and simultaneously mitigate the toxic effects of incomplete combustion through chemical methods. Catalytic converters, which are installed in series with the exhaust pipe of gasoline vehicles, convert more than 90% of the hydrocarbons (HC), carbon monoxide (CO), and nitrogen oxides (NOx) emitted from the engine into carbon dioxide (CO2), nitrogen, and water vapor. Since catalytic converters were first installed on cars in 1974 (mandatory in Iran since 2002 according to the Euro 2 standard and testing for issuing technical inspection certificates since 2012 according to the Euro 4 standard), more than 12 billion tons of harmful exhaust gases have been prevented from entering the atmosphere. More than 96% of cars produced today are equipped with catalytic converters. The typical lifespan of a catalytic converter is estimated to be 80,000–100,000 km, depending on the engine's performance, volume, and fuel emissions. Used automotive catalytic converters contain precious platinum group metals (PGMs). Due to environmental constraints, these metals must be recovered from catalytic converters at the end of their life [1].

Despite their positive role in reducing exhaust emissions, catalytic converters hurt the environment due to the release of platinum metal particles.

The high cost of these metals has prompted extensive research into alternatives; however, there is currently no economical alternative with similar catalytic properties. Given that every vehicle produced requires a catalytic converter and the automotive supply and demand market is constantly increasing, the demand for platinum group metals, especially platinum, is expected to continue to increase.

A catalytic converter contains approximately 1 to 15 grams of PGMs. This sector is therefore considered the primary application area for metals in this group, including palladium, platinum, and rhodium [2]. These metals are critical to the development of sustainable technologies due to their exceptional catalytic properties. On the other hand, they are scarce, and therefore, their supply chain is hampered due to geopolitical implications. Therefore, the introduction of efficient and effective recycling technologies is essential to ensure the responsible and environmentally friendly use of these valuable resources [3]. Catalytic processes are utilized in various sectors, including power generation, transportation, and chemical production.

Their efficient recycling helps reduce energy consumption, lower greenhouse gas emissions, and minimize waste generation [4].

2. Structure and types of automotive catalytic converters

A catalytic converter is a component installed in the exhaust system of a vehicle. This component reduces

*Corresponding author
Email address: a.rabieifar@kiau.ac.ir

the activation energy required for a reaction, thereby increasing its rate of occurrence. After the reaction is complete, the catalytic converter retains its original chemical composition. It converts harmful agents, such as carbon monoxide, hydrocarbons (HCx), and nitrogen oxides (NOx), produced from incomplete combustion in the engine cylinders into nearly harmless substances, including carbon dioxide (CO_2), nitrogen (N_2), and water vapor (H_2O). This conversion is facilitated by metals such as platinum, rhodium, and palladium (in some applications, gold or cadmium) or a combination of them that possess catalytic properties. The catalytic converter consists of a steel shell (often made of stainless steel), a monolithic ceramic or metal core, or a substrate in the form of a honeycomb structure (Fig. 1), in which metals such as platinum, rhodium, palladium, and metal fibers are used. The honeycomb surface increases the amount of surface area available to the catalyst and is therefore referred to as the catalyst support. Additionally, the catalyst features a lambda probe, which is crucial for ensuring the correct fuel-to-air mixture. The main part of the catalyst is its active area, i.e., the ceramic or metal substrate, which is coated with a vapor deposition of platinum, rhodium, and palladium metals (Fig. 2). This coating, known as a wash coat, typically consists of a mixture of alumina, silica, aluminum oxide, or titanium dioxide, enhancing the efficiency of the catalytic converter while protecting the catalyst material. When the wash coat is added to the core, the surface becomes rougher and its surface area increases compared to the smooth core surface. As a result, more sites are available for the active precious metals to be located [5].

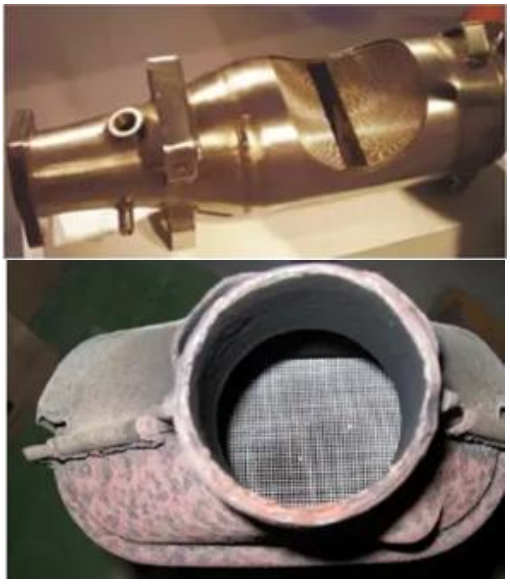

Fig. 1. Exhaust catalyst with metal core (left) and ceramic core (right) [6].

Catalysts (precious metals) are often added via a suspension solution to the wash coat and then applied to the core (Fig. 3). Platinum and rhodium combinations are used as reductive catalytic converters, and platinum and palladium combinations are used as oxidizing catalytic converters [2,3].

The exhaust catalytic converter requires a significant amount of heat to complete its chemical process, around 800-900 ˚C. For this reason, this component must be placed on the part of the exhaust that produces the most heat. On the other hand, another important feature of the catalytic converter is to help absorb heat, thereby reducing the temperature of the car engine. To achieve the goals mentioned above, the catalytic converter is installed at the end of the exhaust system on the engine side of the car, located next to the exhaust manifold outlet. This placement enables it to perform optimally under the specified conditions, particularly in environments requiring very high temperatures [2, 5].

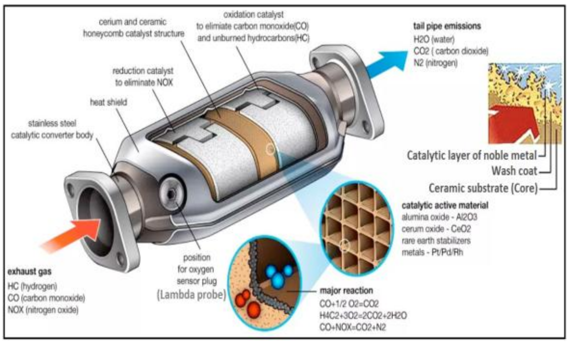

Fig. 2. A schematic of the various components of a automobile exhaust catalyst [7].

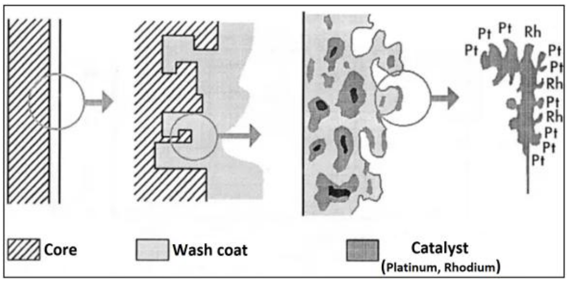

Fig. 3. A schematic of the settlement of the exhaust catalyst PGMs in the washcoat [8].

Catalytic converters are divided into two types: two-way and three-way. The two-way converter has two tasks simultaneously. Firstly, the oxidation of carbon monoxide to carbon dioxide occurred, and secondly, the oxidation of unburned or partially burned hydrocarbons (CXH2X+2) to carbon dioxide and water (a combustion reaction) took place. This type of catalyst is widely used in diesel engines, reducing emissions of carbon monoxide and hydrocarbons. The three-way catalytic converter, which is most commonly used in gasoline engines, performs three tasks simultaneously [8,9].

1. Reduction of nitrogen oxides (NOX) to nitrogen and oxygen;
2. Oxidation of carbon monoxide to carbon dioxide;
3. Oxidation of unburned hydrocarbons to carbon dioxide and water.

These three reactions occur when exhaust from an engine operating slightly above the stoichiometric point reaches the catalytic converter. In this case, the fuel-to-air weight ratio for gasoline is 1:14.6 to 1:14.8. This ratio is somewhat different for natural gas, LPG, and ethanol fuels. When using a three-way catalytic converter, one or more oxygen sensors are also used in the engine. If the fuel-to-air ratio is stoichiometric, the conversion of all three pollutants occurs according to the mentioned reactions, and the catalytic converter's efficiency is high. However, outside this stoichiometric ratio, the conversion efficiency drops rapidly. If more oxygen is available than required, the system is in an oxidizing condition. In this condition, the two oxidizing reactions of the catalytic converter (CO oxidation and hydrocarbons) are preferred. However, if there is more fuel, reducing NOX is preferred over oxidizing CO and HC. If the air-fuel ratio is low, three-way catalytic converters can store and consume oxygen from the exhaust gas stream. This event also occurs when the oxygen from NOX reduction is low [9]. The dependence of pollutant conversion on the air-fuel ratio in a catalytic converter is shown in Fig. 4.

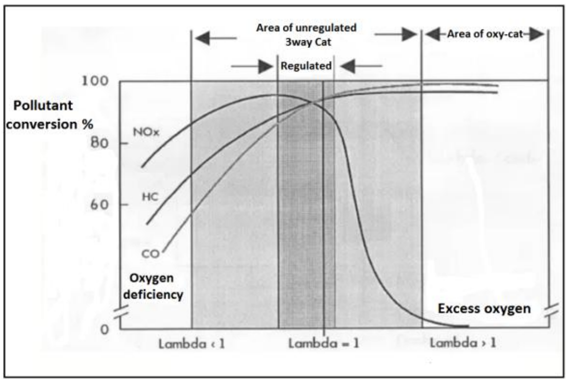

Fig. 4. Dependence of pollutant conversion on air-fuel ratio in exhaust catalysts [9].

Unwanted reactions also occur in three-way catalytic converters, such as the formation of hydrogen sulfide and ammonia. The formation of each of these can be limited by improving the wash coat and the precious metals used, but it is not easy to completely prevent the formation of these by-products [9, 10].

As mentioned earlier, two-way catalysts are used in diesel engines. The efficiency of these catalysts in oxidizing CO and converting it to CO_2, as well as oxidizing hydrocarbons and converting them to water and CO_2, is approximately 90%. However, these converters cannot reduce NOX compounds. To reduce NOx, the chemical composition of the exhaust gas must be changed first. In this regard, two techniques are used, including selective catalytic reduction (SCR) and NOX trapping. The exhaust gas of diesel engines contains a high percentage of soot and elemental carbon. Therefore, with the help of a diesel particulate filter (DPF), it will be possible to separate up to 90% of soluble organic substances such as soot. Instead of a catalyst, pyrolyzed ammonia from urea is also used to reduce NOX to nitrogen [10].

Temperature sensors are often used in two-way catalytic converters. These sensors operate when the catalytic converter temperature exceeds the safe limit of 750 °C (1380 F). Most catalytic converters are designed to be insensitive to temperature damage and can withstand temperatures up to 900 °C (1650 F). Temperature sensors are also used to evaluate the performance of catalytic converters. Two sensors, one upstream and one downstream, are used to monitor the temperature rise in the catalytic converter core. For every 1% CO in the exhaust gas stream, the exhaust gas temperature increases by 100 °C [10]. The dependence of the efficiency of exhaust catalysts on temperature changes is shown in Fig. 5.

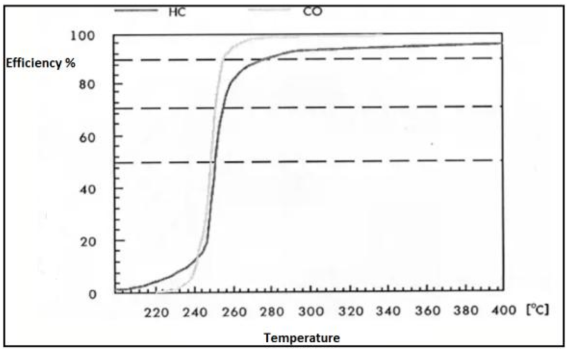

Fig. 5 Dependence of exhaust catalyst efficiency on temperature [10].

3. Damage to Automotive Catalytic Converters

Poisoning of the catalytic converter occurs when exhaust gas materials cover its surface. As a result, the catalytic converter cannot come into contact with the exhaust gas. The most important pollutant in this area is lead. Therefore, vehicle fuel should be as free as possible from high concentrations of lead. Another pollutant is phosphorus, which is not found in automotive fuel. Today, phosphorus is used as an anti-wear additive in engine oil. Suppose the engine is operated for a long time and under high load, due to the heating of the exhaust gas. In that case, pollutants on the surface of the catalytic converter may melt or sublimate. It is not possible to remove lead deposits due to its high boiling point. Any condition in which a significant amount of unburned or partially burned hydrocarbons reaches the catalytic converter will result in a substantial increase in its temperature, potentially causing substrate meltdown and catalytic converter inactivation. Vehicles equipped with OBD-II diagnostics are used to alert the driver to this condition. Another problem with stoichiometric catalytic converters (a 20:1 fuel-to-air ratio) is that, despite their low fuel consumption and carbon dioxide emissions, it is difficult to control NOX compounds in these converters. In addition, the production of large amounts of CO_2 as a greenhouse gas, as well as nitrous oxide (N_2O), which is 300 times more potent than CO_2, contributes significantly to global warming. On the other hand, the production of catalytic converters requires huge amounts of platinum and palladium reserves [11].

4. Methods for Recycling Used Catalysts

The operating temperature of around 250 °C is very important for catalysts. Metal catalysts reach this temperature faster than ceramic catalysts. The rapid heating rate is significant because a large amount of exhaust gases is produced when the catalyst reaches operating temperature. At very high temperatures, metal catalysts exhibit superior physical and chemical properties compared to their ceramic counterparts. The cylindrical and corrugated design enables them to withstand excessive vibrations and frequent temperature fluctuations more effectively. With stricter regulations on greenhouse gas emissions, the amount of platinum, palladium, and rhodium in automotive catalytic converters is increasing. A two-way catalyst contains 0.04 wt.% platinum and 0.015 wt.% palladium, while a three-way catalyst contains 0.08 wt.% platinum, 0.04 wt.% palladium, and 0.005–0.007 wt.% rhodium. Palladium and rhodium are of considerable economic importance. If properly managed, the collection and recycling of automotive catalysts will provide a significant secondary source of platinum, palladium, and rhodium [12]. The processing of spent catalysts can be classified into two main methods (Fig. 6).

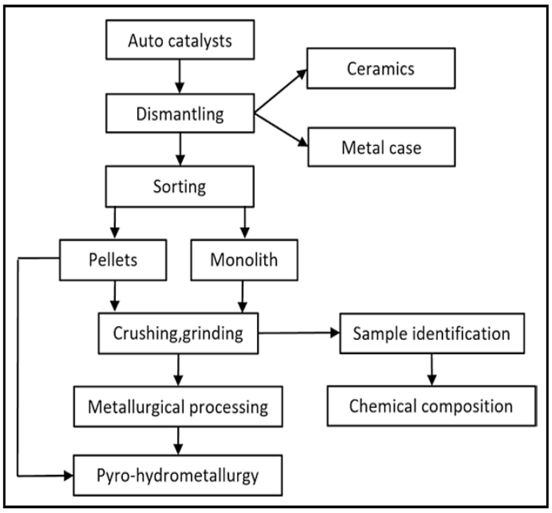

Fig. 6 Processing and recycling of used exhaust catalysts [13].

4.1. Pyrometallurgical Process

There are various approaches to recovering valuable metals from spent catalysts. Pyrometallurgical processes used in platinum metal recycling involve the use of collector metals, particularly iron and copper. This process can be divided into two components:

A: Plasma Melting: In this process, the catalyst is crushed, ground, and mixed with a mixture of Fe + FeO. It is then melted in a plasma furnace and heated to temperatures exceeding 2000 °C. Next, during the separation process, the molten slag is separated from the iron-rich phase, which contains the concentrated platinum group metals. The iron alloy is subsequently aerated and dissolved in an aqueous solution of sulfuric acid. While the filtration performs the neutralization, any un-dissolved platinum group metals are also filtered out [12, 13].

B: Melting in EAF (Electric Arc Furnaces: During the copper smelting process, the catalyst, after undergoing mechanical treatment including crushing and grinding, is combined with copper carbonate, silica, calcium oxide, and iron oxide in a special electric furnace at temperatures from 1600-1800 °C. This process results in the separation of slag and an alloy of platinum group metals with copper. The copper alloy is dissolved in an aqueous sulfuric acid solution using air as an oxidant. Copper carbonate is recovered by precipitation with soda ash, and the cooled solution is subjected to filtration. Today, in pyrometallurgical methods, special emphasis is placed on the melting method with copper as the collector metal and plasma melting using iron as the collector. To extract precious metals from an alloy, the use of a collector metal is necessary, making hydrometallurgical processes essential. Therefore, recycling methods are often used as combined processes. Fig. 7 shows the pyrometallurgical recycling process for platinum group metals [14].

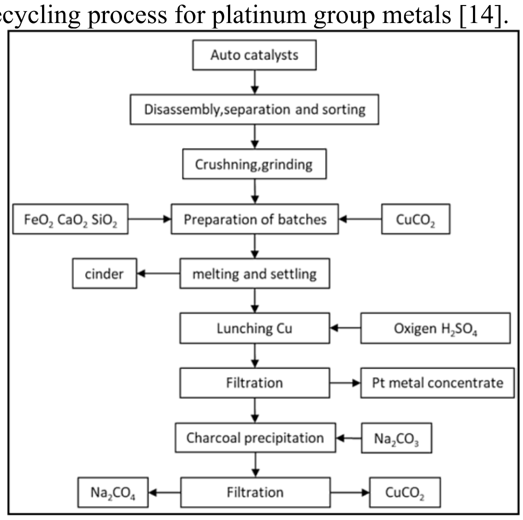

Fig. 7. Recycling of used exhaust catalysts through pyrometallurgy [13].

4.2 Hydrometallurgical methods

In these methods, platinum group metals are leached using salicylate acids, and the resulting leachate containing these metals is purified by chemical precipitation or cementation. The purified leachate is further processed into a metal form or converted into chemical concentrates of precious metals through alternative methods, such as adsorption, ion exchange, or extraction. However, disadvantages of these methods include loss of metal, especially rhodium, in the wastewater and the production of aluminum sulfate as a by-product [12, 13].

Catalyst purification in hydrometallurgical processes involves initial calcination at temperatures of about 500-600 °C. In this step, organic residues from gasoline and oils are burned. Carbon residues, if not burned, stick to the pores of the catalyst, preventing the penetration of leaching solutions and reducing metal yield. The purified catalyst pellets are then dissolved in sulfuric acid to produce an almost neutral solution. The leachate from the leaching of the catalysts is purified by cementation with aluminum in the presence of tellurium dioxide. The resulting aluminum sulfate is used in water purification. The solid phase from the cementation, along with insoluble residues from the initial leaching, is dissolved in a mixture of HCl + Cl_2. Platinum group metals are separated from this solution using sulfur dioxide, with tellurium as a collector. The filtered solution produces platinum group metals, and subsequent cooling produces lead chloride. Hydrochloric acid (HCl) is recovered in the process. The acid leaching purification method for the catalyst is shown in Fig. 8. Hydrometallurgical methods include acid leaching of the entire pellet or selective leaching of platinum group metals. Subsequently, the extract is subjected to refining and processing, which often involves pressure reduction or precipitation [13].

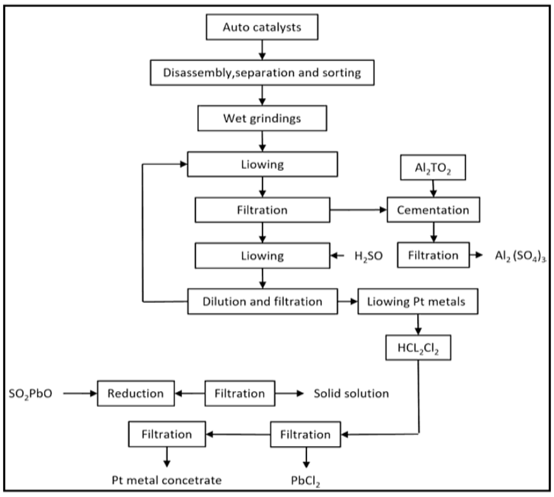

Fig. 8. Recycling of used exhaust catalysts through hydrometallurgy [13].

5. Classification and Estimation of Catalyst Life

To evaluate the performance and remaining life of catalysts, the characteristics of the monolithic ceramic substrate, including cell density, cell wall thickness, and wash coat thickness, is usually measured using OM images [15-17]. Additionally, the concentrations of platinum, palladium, and rhodium in the catalyst are calculated in ppm using XRF [17]. The catalysts used are divided into general categories. Those containing rhodium along with platinum and/or palladium are classified as three-way catalysts (TWC). Catalysts that mainly contain platinum and/or palladium are generally classified as diesel oxidation catalysts (DOC). Used catalytic converters with lower concentrations of platinum and/or palladium (depending on the vehicle model and year of manufacture) are referred to as dual-function catalysts [18].

6. Global Supply and Demand of Precious Metals

Platinum group metals (PGMs) are utilized in various sectors, including dentistry, jewelry, and the chemical industry. However, their main application is attributed to their catalytic properties. Approximately half of the platinum and palladium used is in automotive catalysts. Rhodium, on the other hand, is used primarily (about 80-90%) as an alloying element in the active layers of various catalysts. The significant amount of ore required (300-900 kg) to produce approximately 1 g of PGMs results in a substantial increase in their global price. Typically, ores containing Pt and Pd contain 5-10 times more than Rh and Ru ores and approximately 50 times more than Ir and Os ores [18, 19].

Furthermore, mineral reserves of PGMs are limited to two countries: South Africa and Russia. Therefore, the gap between the supply and demand of PGMs necessitates recycling catalysts from both environmental and economic perspectives. Hence, efforts are being made to improve catalyst efficiency through advances in washcoat technology, utilizing heavy oxides of rare earth elements, such as cerium. Rare earth elements, such as cerium oxide, increase the surface area and enhance the oxygen supply. However, the introduction of cerium as an additional critical element in catalytic converter recycling poses a major challenge, especially for conventional pyrometallurgical methods [19].

According to the US Geological Survey, in 2013, 95% of the world's PGMs reserves were in South Africa, 1.6% in Russia, and the remaining 3.4% were scattered across the globe, emphasizing the strong dependence on South Africa's supply. The costly processing of ores due to permafrost in Russian mines and deep mines in South Africa adds to this challenge. In Russian mines, PGMs are a by-product of nickel ore. Therefore, any attempt to increase PGM production requires an increase in nickel production, which leads to overproduction and, consequently, a decrease in the price of the primary product, nickel [20-27].

7. Conclusion

Exhaust catalysts consist of four main parts: a steel shell (often stainless steel), a monolithic ceramic or metal core, a lambda probe, a wash coat (usually a mixture of alumina, silica, aluminum oxide or titanium dioxide produced by dip coating), and precious metal particles of platinum/palladium with oxidizing ability and platinum/rhodium with reducing ability. Catalyst toxicity due to contact with lead, phosphorus, or unburned hydrocarbons leads to surface contamination of the catalysts and an increase in their temperature, meltdown of the monolithic core, and deactivation of the catalyst. Recycling of spent catalysts is carried out by pyrometallurgical methods, either by plasma melting and Fe-FeO mixture at temperatures above 2000˚C and dissolution in sulfuric acid, or by electric arc furnace with mixtures of copper carbonate, silica, and calcium oxide at temperatures of 1600-1800˚C and by copper collector. The most common hydrometallurgical method involves the leaching of PGMs in Salicylicacids and the purification of the resulting leachate through the cementation process.

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