IMPROVEMENT OF THERMAL EFFICIENCY OF THE MHD PUMP FOR THE RECOVERY OF PLATINUM FROM SPENT CATALYTIC CONVERTERS IN CONTEXT OF ITS COMMERCIALIZATION

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Abstract

An increase in the consumption of platinum in the automotive industry is related to the growing number of cars manufactured. Modern and well-used equipment of this type which is fitted as standard in cars should work for at least $8 \cdot 10^7$ - $9 \cdot 10^7$ metres. When worn, they are considered as waste being a valuable source of platinum-group metals (platinum, palladium and rhodium) which act as catalysts in catalytic converters. The management of spent catalytic converters helps to reduce the amount of waste disposed as well as to lower the emission of pollution to the air in relation to the processes involving obtaining such metals in the production technologies from raw materials. Various hydrometallurgical, pyrometallurgical or mixed methods are used for the recovery of noble metals from spent catalytic converters. The purpose of the article is to present an experimental method for the recovery of noble metals from spent catalytic converters. This method involves flushing of spent catalytic converters with a liquid collector metal with the use of a magneto-hydro-dynamic pump for the intensification of the process and is applied to recover platinum from catalytic converters by melting them (collecting) in the liquid metal. In the paper the optimization of the operation of the system for the recovery of platinum-group metals which is essential in the light of later commercialization of the solution is proposed.

Keywords: Spent automotive catalysts, PGM recovery, thermal efficiency

1. INTRODUCTION

An increase in the consumption of platinum in the automotive industry is related to the growing number of cars manufactured. The catalytic converters that are fitted as standard in cars should work for about $8 \cdot 10^7$ - $9 \cdot 10^7$ metres. In practice, they are used much longer. After this time, they should be disposed properly. Spent catalytic converters constitute a valuable source of platinum-group metals (platinum, palladium and rhodium), that is metals which acts as catalysts. A proper management of spent catalytic converters allows reducing the amount of waste deposited. Moreover, the level of emission into the atmosphere is lower during the processes of obtaining metals from waste materials, in comparison to the technologies of obtaining them from raw materials. Scheme of catalytic converters is shown on **Figure 1**. Furthermore, recovery of platinum-group metals is beneficial in view of their limited global resources as well as costs related to their obtaining [1]. Various hydrometallurgical, pyrometallurgical or mixed methods are used for the recovery of these metals from spent catalytic converters that are commonly applied in the automotive industry [2-4]. The most common pyrometallurgy method is the collector metal method, using copper or nickel lead in this role [5].

In the paper an experimental method for the recovery of platinum-group metals from waste generated in the automotive industry (spent catalytic converters) involving flushing of spent catalytic converters with a liquid metal, with a lead as a “collector metal” with the use of a magneto-hydro-dynamic (MHD) pump for the intensification of the process is presented. This method allows for the recovery of platinum from catalytic converters by collecting them in the liquid metal. The various stages of handling of spent automotive catalysts by using the mhd device are shown in **Figure 2**.

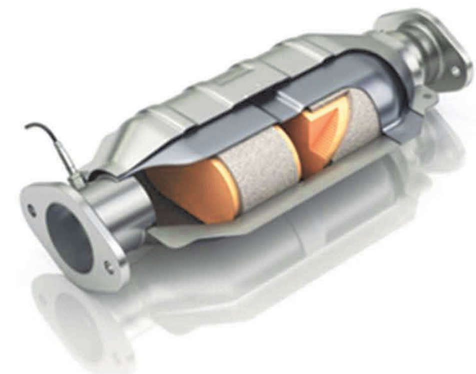

Figure 1 Scheme of auto catalytic converters

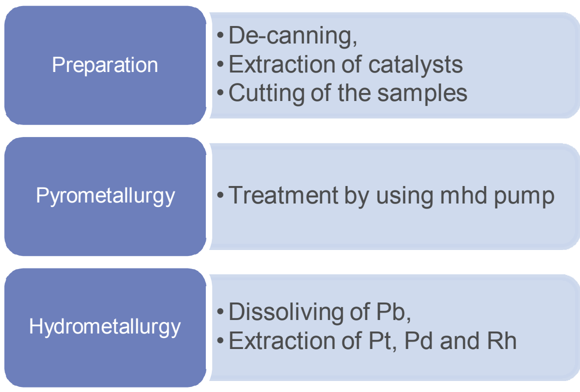

Figure 2 The various stages of handling of spent automotive catalysts by using the mhd device

When considering the possibilities of commercialization of the presented solution, it was attempted to describe theoretically this issue, based on the methods of numerical computational fluid dynamics and electromagnetic phenomena [6], and also an energy balance was developed for this process [7]. The operation of the equipment was optimized by increasing its thermal efficiency, which is crucial in view of the profitability of the whole process of recovery of platinum-group metals from spent catalytic converters.

2. DESCRIPTION OF MHD PUMP OPERATION

The equipment for the recovery of platinum from spent catalytic converters with the use of an electromagnetic field is shown in **Figure 3**. The principle of operation of the MHD pump (magnetohydrodynamic pump) for the recovery of platinum consists in placing liquid metal in an annular channel around which there is an inductor wound on the core producing a vortex field (**Figure 4**) having an axis coincident with the axis of the ring. The rotating electromagnetic field produces eddy currents in the liquid metal that interact with the electromagnetic field of the inductor, and the Lorentz force is generated causing whirling motion of the metal. Placing catalytic converters in the so produced flow of liquid metal allows melting platinum, palladium and rhodium from their capillaries, and the continuous motion of metal significantly intensifies the process of rinsing out. The solution is designed for using of the same metal for flushing a large number of catalytic converters results in the appearance of platinum in the solution and then, in an increase of the concentration of platinum to the values which guarantee the profitability of their extraction from the liquid metal. The application of a closed circuit of lead allow to limit the negative impact of the process on the atmosphere [4].

The main advantages of this method is that can be used both type of catalytic converters applied in cars: on metallic and ceramic carriers, and the operating temperature is much lower than in case of conventional methods. The method does not require grinding of spent catalytic converters before the process of rinsing platinum out from them.

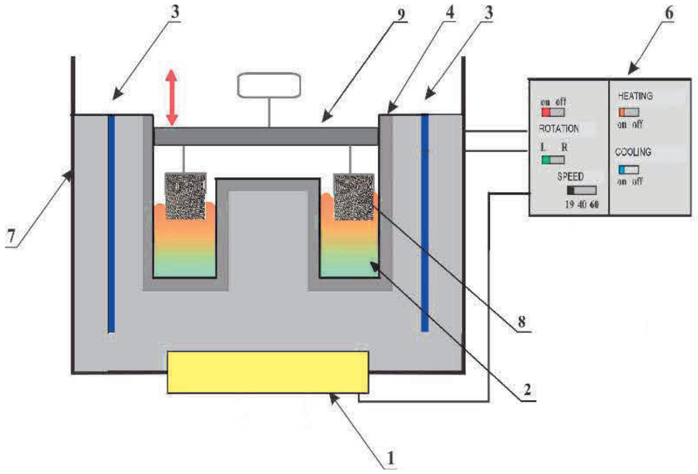

Figure 3 Scheme of installation for recovery of platinum from waste generated in the automotive industry
1 - cooling, 2 - liquid metal, 3 -winding, 4 channel, 5 - thermal insulation, 6- controller, 7 - casing, 8 - catalyst, 9 - cover

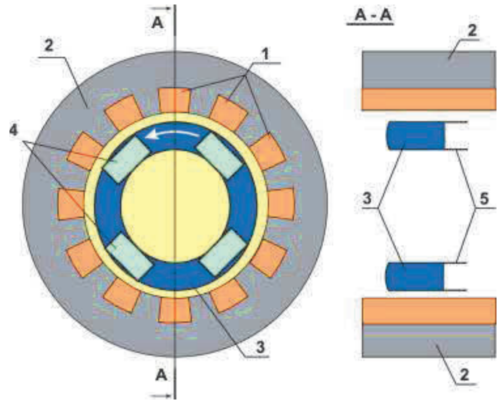

Figure 4 Scheme of MHD pump for metals recovery from spent catalyst carriers:
1 - winding; 2 - magnetic core; 3 - liquid metal; 4 - catalyst carriers; 5 - channel

3. ESTIMATING THE ENERGY EFFICIENCY OF THE MHD PUMP

The equipment for the recovery of platinum from spent catalytic converters with the use of MHD pump is powered by electricity. The parameters for presenting device and process are presented in **Table 1**.

Table 1 Parameters of the device using magnetohydrodynamics for the recovery of precious metals from used car catalysts

Parameters	**Parameter value**
Power - electricity	Power demand P = 100 W
Range of Temperature	Ambient Temperature $T_1 \approx 293$ K
	Working Temperature $T_2 \approx 673$ K
Time	The sample of catalysts are faded every t = 300 s
Mass	**Catalytic converter** - average mass of one sample m = 0.24 kg
	Platinum - average content of Pt in one sample $4.3 \cdot 10^{-6}$ kg
	Lead - average content of Pb circulating in the channel 50 kg

It has been assumed in the calculations that the catalyst matrix is composed of Al_2O_3 and it is an inert in this process. Based on the computations made with the application of the HSC Chemistry 6 software, a correlation of physical increase of enthalpies relevant for Al_2O_3 and Pt was determined:

$$\Delta i(T) = \int_{T_1}^{T_2} (a + bT + cT^2 + dT^3 + eT^4) dT, \quad \text{(J/kg)} \tag{1}$$

The coefficients of the correlation equation are shown in **Table 2**.

Table 2 Coefficients of the correlation equation (1)

No.	Chemical unit	*a*	*b*	*c*	*d*	*e*
		J/(kg·K)	J/(kg·K^{-2})	J/(kg·K^{-3})	J/(kg·K^{-4})	J/(kg·K^{-5})
1	Pt	131.8	0.033	$-3·10^{-5}$	$5·10^{-8}$	$-4·10^{-11}$
2	Al_2O_3	707.9	2.05	-0.003	$2·10^{-6}$	$-2·10^{-9}$

The heat absorbed by a portion of the batch during heating up was calculated as a sum of products of masses and the physical increases of specific enthalpies from the initial temperature T_1 to the temperature of the lead bath T_2. An average power value P_2 was assumed in the energy balance, and it was determined empirically with consideration of the duration of the periods between the changes of a new portion of the batch as well as the flow of energy needed to heat up the crucible in order to maintain the desired temperature. The heat loss to the surroundings through the mirror of molten metal that occurred mainly as a result of convection and radiation was also calculated separately [7]. The heat loss to the surroundings Q_e (**Eq. (2)**) was determined based on the developed energy balance, and afterwards, it was compared with the heat loss determined in the paper [7] in order to adjust the balance [8]:

$$Q_e = P \cdot t - m[g \cdot \Delta i_1(T) + (1-g) \cdot \Delta i_2(T)], \quad \text{(J)} \qquad (2)$$

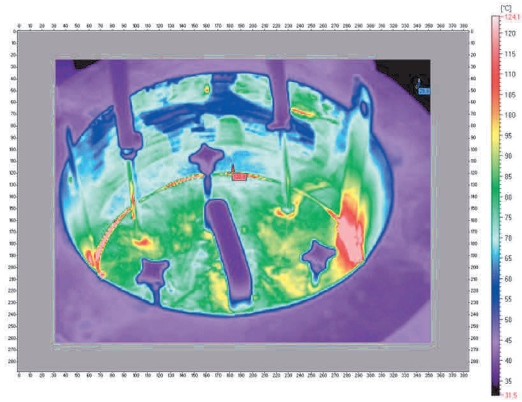

Figure 5 Side view of MHD pump [9]

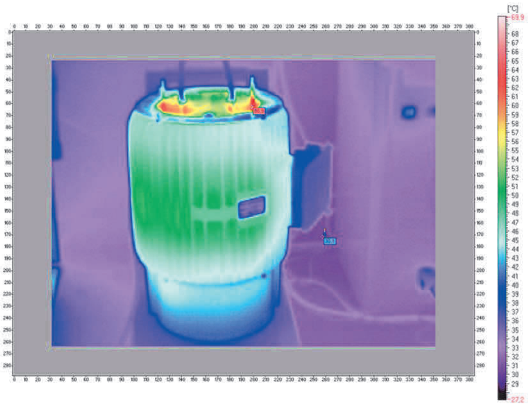

Figure 6 Top view of MHD pump [9]

To protect the surface of the lead bath against oxidation, argon with a mass flow of $9.91·10^{-6}$ kg·s^{-1} was forced into the space between the mirror of the molten metal and the screen. This provided a slight overpressure of the inert gas above the surface of metals. The flowing argon was heated from the ambient temperature to the temperature close to the temperature of the mirror of the lead bath. The enthalpy flow leaving the space of the equipment with heated argon was negligible, approximately 2 W [6]. The most intensive heat loss in MHD pump was through the top cover, as shown in **Figures 5** and **6** presenting an infrared image.

A high-temperature plate made of a ceramic fibre with a coefficient of thermal conductivity λ_i = 0.05 W/(m·K), which is characterized by high thermal and chemical resistance, was used as the insulating material. The cover was made of austenitic steel with a coefficient of thermal conductivity λ_c = 15 W/(m·K), diameter d_c = 0.29, surface A_c = 0.066 m^2, and thickness $\delta_c = 2·10^{-3}$ m. Firstly, the coefficient of heat transfer α_1 from the lead bath to the inner cover surface was to be estimated. Due to a significant share of radiation in the heat loss to the surroundings, besides the convective coefficient of heat transfer α_c, also the radiation coefficient of heat transfer α_r was determined. The measured temperature of bath was $T_2 \approx 673$ K, and the temperature of the cover was $T_3 \approx 353$ K. The ratio of radiant energy exchange for two parallel planes was calculated based on the knowledge of the emissivity for the lead bath ε_1 = 0.28 and the emissivity of the cover ε_2 = 0.35 [8]. Comparing the heat flow flowing to the cover as a result of convection (Newton's equation) with the heat flow flowing as a result of radiation (Stefan-Boltzmann law), a radiation coefficient of heat transfer was calculated and it was α_r = 6.4 W/(m^2·K). Due to the extremely slow flow of argon, an algorithm for natural convection was used in the calculations, and an average temperature in the convection flow was T_{m1} = 513 K, the determined convection coefficient of heat transfer was α_c = 11.1 W/(m^2·K), while the total coefficient of heat transfer from the lead bath to the internal cover surface was α_1 = 17.5 W/(m^2·K). A natural convection was present at the outside of MHD pump, and an average temperature of air in the convection flow was T_{m2} = 323 K. The calculated convection coefficient of heat transfer from the outer cover surface to the air α_2 = 5.8 W/(m^2·K).

Loss of heat through the cover as a result of heat transfer was estimated based on the following equation:

$$\Delta Q_{BC} = Q_{BC}(\delta_i = 0) - A_C\, t \left(\frac{T_2 - T_1}{\frac{1}{\alpha_1} + \frac{\delta_C}{\lambda_C} + \frac{\delta_i}{\lambda_i} + \frac{1}{\alpha_2}} \right), \qquad \text{(J)} \qquad (3)$$

where the thickness of the insulation δ_i was assumed in the range 0 - 0.05 m, and the maximum thermal efficiency of the equipment constituted the optimization criterion. Loss of heat for one run cycle (Q_e = 236 kJ) and the thermal efficiency of MHD pump (η = 28 %) were previously determined [6]. The following dependencies were applied for current calculations of the efficiency of the equipment

$$\eta = \left(1 - \frac{Q_B - \Delta Q_{BC}}{P\, t} \right) 100\,\% \qquad (4)$$

The calculation results of the equipment efficiency in the function of the thickness of the insulation layer are shown in **Figure 7** [9]. The maximum thickness of the insulation to which it is worth to insulate the cover of MHD pump is δ_i = 0.03 m, and the thermal efficiency of the equipment reaches the limit value of η = 36 %. The above-specified actions allowed increasing the efficiency of the pump by 8% in comparison to the initial state, and further thickening of the insulating layer is unprofitable.

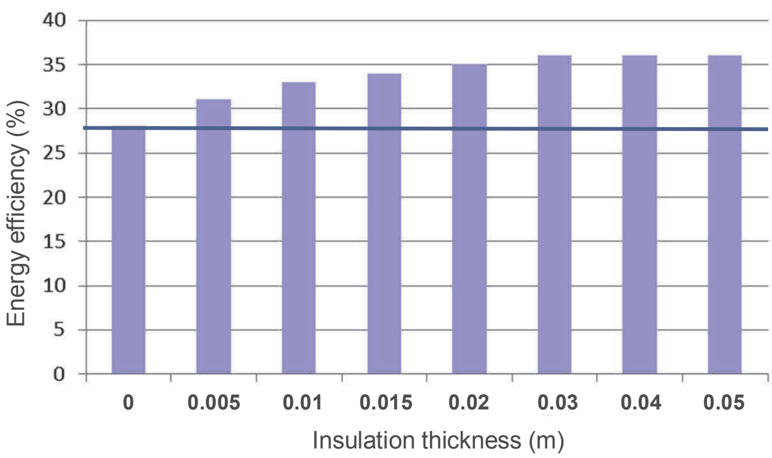

Figure 7 The effect of increasing the thickness of the insulation on the process efficiency

4. CONCLUSIONS

1) Recovery of valuable secondary raw materials, such as noble metals (platinum, rhodium or palladium) used for production of catalytic convertors is particularly important due to the reduction of the amount of waste and protection of environment.

2) The use of a magneto-hydro-dynamic pump for the recovery of noble metals is currently an experimental, innovative and unique method in the world.

3) An attempt to increase the thermal efficiency by insulating the cover of the pump was undertaken. As a result of the presented calculations, it was found that insulating the cover of MHD pump with an insulating layer with a thickness of 0.03 m will allow achieve the maximum thermal efficiency of the equipment of 36%, which is higher by 8% in comparison to the initial state.

REFERENCES

[1] HAGELÜKEN, C. Recycling the platinum group metals: a European perspective. *Platinum Metals Review*, 2012, vol. 56, no. 1, pp. 29-35.

[2] YOO, J. S. Metal recovery and rejuvenation of metal-loaded spent catalyst. *Catalyst Today*, 1998, vol. 44, pp. 27-39.

[3] POŚPIECH, B. Hydrometalurgiczne technologie odzysku metali szlachetnych i nieżelaznych ze zużytych katalizatorów. *Przemysł Chemiczny*, 2012, vol. 91, no. 10, pp. 1000-1003. (in Polish).

[4] OLEKSIAK, B., SIWIEC, G., BLACHA-GRZECHNIK, A., WIECZOREK, J. The obtained of concentrates containing precious metals for pyrometallurgical processing. *Metalurgija*, 2014, vol. 53, no. 4, pp. 605-608.

[5] RUMPOLD, R., ANTREKOWITSCH, J. *Recycling of platinum group metals from automotive catalysts by an acidic leaching process.* The Southern African Institute of Mining and Metallurgy Platinum, 2012, pp. 695-714

[6] FORNALCZYK, A., GOLAK, S., PRZYŁUCKI, R. Investigation of the influence of supply parameters on the velocity of molten metal in a metallurgical reactor used for platinum recovery. A*rchives of Civil and Mechanical Engineering*, 2015, vol. 15, no. 1, pp. 171-178.

[7] GIL, S., BIALIK, W., SATERNUS, M., FORNALCZYK, A. Thermal balance of the magneto-hydro-dynamic pump for recovery of platinum group metals from spent auto catalysts. *Archives of Metallurgy and Materials*, 2016, vol. 61, no. 1, pp. 253-256.

[8] KOSTOWSKI, E. *Przepływ ciepła*. Wydawnictwo Politechniki Śląskiej, Gliwice 2000. (in Polish).

[9] FORNALCZYK, A. *Analiza możliwości wykorzystania magnetohydrodynamiki do intensyfikacji odzysku platyny ze zużytych katalizatorów samochodowych*. Wydawnictwo Politechniki Śląskiej, 2016. (in Polish).