Recovery of Platinum and Palladium from Scrap Automotive Catalytic Converters

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Abstract

The hydrometallurgical method was used to platinum and palladium leaching with aqua regia solution (3HCl: HNO_3). The leaching experiments were designed to obtain the optimum conditions by using Taguchi method with 16 experiments at three different factors (time, temperature and solid to liquid ratio), and each factor has four different levels. In this study, leaching the powder sample of catalytic converter that contains platinum and palladium was conducted on the basis of the formation of chloro complexes platinum and palladium ($PtCl_6^{2-}$, $PdCl_4^{2-}$) with different concentrations in the acidic solution. The optimum conditions for leaching process are (time = 120 minutes, temperature = 100°C, and solid to liquid ratio = 1/20).The best efficiency for platinum and palladium leaching at these conditions were 97.58%, 93.23%, respectively. After leaching process at optimum conditions, platinum was firstly precipitated in the form of $(NH_4)_2PtCl_6$ (ammonium chloroplatinate) by adding (5ml/l) of ammonium chloride (NH_4Cl) as a precipitant agent .Then, palladium was precipitated in the form of $(NH_4)_2PdClO_3$ (palladium ammonium chlorate) by adding (5g/l) of sodium chlorate ($NaClO_3$) as a precipitant agent to the remaining solution after the removal of platinum. The best efficiency for platinum and palladium recovery at optimum leaching conditions were 91.94%, and 98.82%, respectively.

Keywords: *Aqua regia, catalytic converter, PGMs, Platinum, Palladium, recovery, catalytic converter, 3HCl:HNO₃.*

1. Introduction

Commitment to the European standards to keep pollution-free environment, all cars manufactured after 1993 should be equipped with catalytic converter. Catalytic converters Contain precious metals like Pt, Pd and Rh, which work on the oxidation (CO), and (HC), and the reduction of (NO_x) to lower the required levels of emissions [1].The formation of catalytic converter carrier vary depending upon engine capacity and kind of used fuels [2].Recovery of platinum group metals from secondary sources was an important mean of making materials economically in addition to environmental considerations. Recycling of automobiles catalysts to recover platinum, palladium, and rhodium had received a lot of

attention in last few years; there are many works provision for the extraction of precious metals by using different ways. platinum group metals are extracted from scrap by melting them with collector metal like copper, or leaching in strong acidic solutions .The extraction methods can be classified as hydrometallurgical, and pyrometallurgical [3,4,and 5]. About 125,000 kg of platinum, palladium, and rhodium was recovered around the world from new and old scrap in 2015; 55,000 kg was recovered from spent catalytic converters in the United States. 25% of Palladium replaced platinum because it is less expensive in most gasoline engines catalytic converters, the proportion can be as much as 50% in some applications [6].The carrier is made up of a ceramic or metallic substances as shown in

Fig.(1).The used type of catalytic converters for the cars are honeycomb cordierite skeleton type $(2MgO-2Al_2O_3-5SiO_2)$,which is coated with(platinum, palladium, and rhodium) with a density of 60–120 cells/cm^2 and the thickness of the walls between the cells is about 150 µm . The internal surface of the cell is coarse and it is covered with high porosity wash coat about 50-200 µm that made up of about 90% γ-Al_2O_3 and a mixture of basic added minerals are oxides of Cerium, Zirconium, Lanthanum, Nickel, Iron and alkaline earths that helps improving the stability of catalytic converter [7].
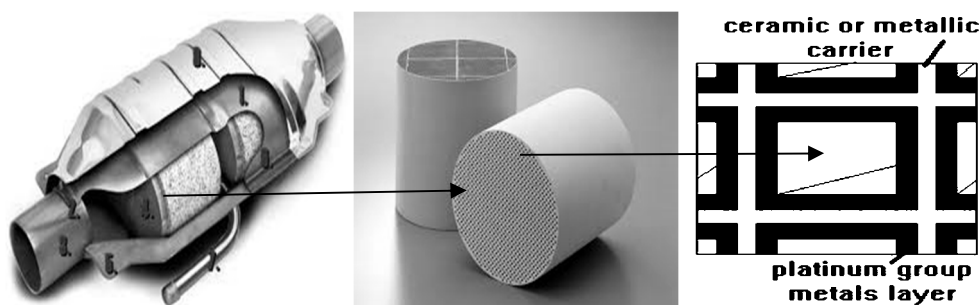

Fig. 1. Carrier auto catalytic converter with the honeycomb structure [8].

The amount of PGMs in catalytic converter is 2 to 6 gram per car and from 6 to 30 gram per sports cars where the exhaust gases are in large quantity. The proportion of platinum to palladium depends on the level of emission control, metals prices, and the type of car. Platinum is used in catalytic converters for diesel engines, while platinum and palladium are used in vehicles powered by gasoline [9].

The total content of platinum group metals in secondary sources is many times higher than in the ore and the benefit from these sources is very attractive .Concentrations of PGMs are to be different in catalysts depending on the manufacturer, the age and the ability of the catalyst .The concentration of PGMs in the Earth crust are measure by micro grams per kilogram. Concentration of PGMs in ores are (Ruthenium = 0.1 µg/kg); (Rhodium = 0.06µg/kg) ;(Palladium= 0.4µg/kg) ;(Osmium and Iridium = 0.05µg/kg); and (Platinum=0.4µg/kg).While, the concentration of Platinum in a new automotive catalytic converter is about 0.3 to 1.0µg/kg, Palladium from 0.2 to 0.8µg/kg and Rhodium from 0.05 to0.1µg/kg [5, and 10].

Auto catalysts recycling, platinum, palladium, and rhodium recovered were about 1.16(Moz), 3.15(Moz), and 3.4(Moz) respectively in 2015. Fig. (2) shows the recovery of Pt , Pd ,and Rh during the years from (2010 to 2015) [11].
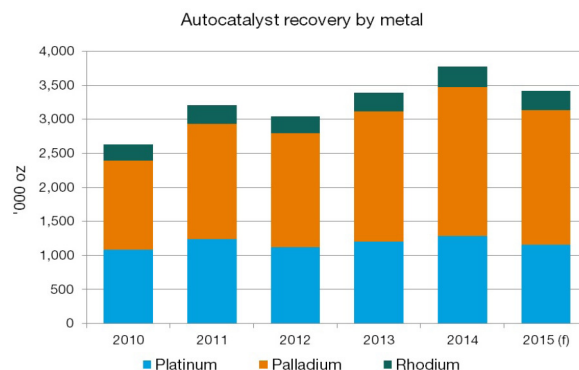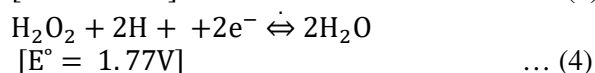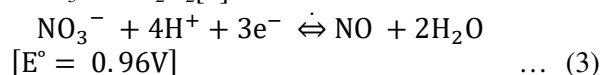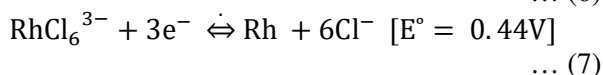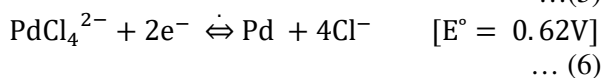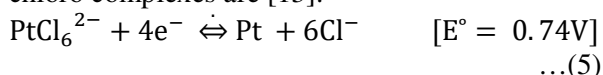

Fig. 2. Recovery of Pt, Pd, and Rh from auto catalysts during years from2010 to 2015 [11].

When PGMs dissolved by aqua regia, forming Cl_2 and NOCl, which has a high oxidation energy and high concentration of chloride ions act as complex agent. Based on the reactions described in (1), and (2) [12].

$$HNO_3 + 3HCl \Leftrightarrow NOCl + Cl_2 + 2H_2O \qquad ... (1)$$

$$NOCl + H_2O \rightleftharpoons HNO_2 + HCl \qquad ... (2)$$

PGMs are very resistant to acid dissolving .Chloro complexes ($PtCl_6^{2-}$, $PdCl_4^{2-}$, $RhCl_6^{3-}$)were formed when Pt,Pd, and Rh leach in acidic chloride solutions, which required to oxidizing agent with a reduction potential >0.74 V. Thus, better oxidizers can be used as oxidizing agents are HNO_3 and H_2O_2[2]:

$$NO_3^- + 4H^+ + 3e^- \Leftrightarrow NO + 2H_2O$$
$$[E° = 0.96V] \qquad ... (3)$$

$$H_2O_2 + 2H + 2e^- \Leftrightarrow 2H_2O$$
$$[E° = 1.77V] \qquad ... (4)$$

Electrode potentials for the half reactions of chloro complexes are [13]:

$$PtCl_6^{2-} + 4e^- \Leftrightarrow Pt + 6Cl^- \quad [E° = 0.74V] \quad …(5)$$

$$PdCl_4^{2-} + 2e^- \Leftrightarrow Pd + 4Cl^- \quad [E° = 0.62V] \quad …(6)$$

$$RhCl_6^{3-} + 3e^- \Leftrightarrow Rh + 6Cl^- \quad [E° = 0.44V] \quad …(7)$$

Leaching of platinum and palladium by aqua regia solution is a redox reaction according to reactions (8), and(9)[14,and 15].
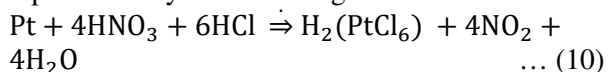
$$3Pt_{(s)} + 18HCl_{(ac)} + 4HNO_{3(ac)}$$
$$\Leftrightarrow 3[PtCl_6]^{2-}{}_{(ac)} + 6H^+{}_{(ac)} + 4NO_{(g)} + 8H_2O \quad …(8)$$

$$3Pd_{(s)} + 12HCl_{(ac)} + 2HNO_{3(ac)}$$
$$\Leftrightarrow 3[PdCl_4]^{2-}{}_{(ac)} + 6H^+{}_{(ac)} + 2NO_{(g)} + 4H_2O \quad …(9)$$
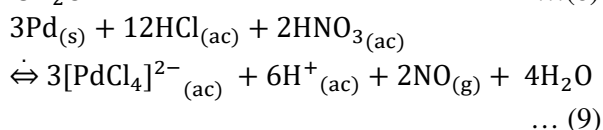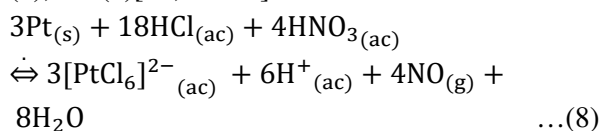
There are two stages to precipitate platinum and palladium [16]:-

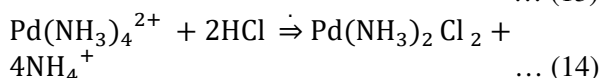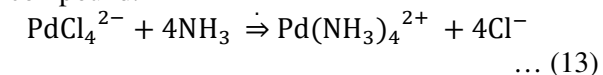1- Leaching platinum by aqua regia was represented by the following reaction:

$$Pt + 4HNO_3 + 6HCl \Rightarrow H_2(PtCl_6) + 4NO_2 + 4H_2O \quad …(10)$$

Platinum is precipitated as ammonium hexachloroplatinate $(NH_4)_2(PtCl_6)$ by adding ammonium chloride to the platinum chloride solution according to the following reaction:

$$H_2(PtCl_6) + 2NH_4Cl \Rightarrow (NH_4)_2(PtCl_6) + 2HCl \quad …(11)$$
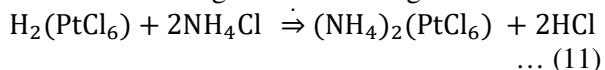
Ignition of ammonium hexachloroplatinate $(NH_4)_2(PtCl_6)$ is carried out by placing it into a glazed refractory evaporating dish and heat strongly to obtain metal called platinum sponge as represented by the following reaction:

$$(NH_4)_2(PtCl_6) \Rightarrow Pt + 2NH_3 + 2HCl + 2Cl_2 \quad …(12)$$
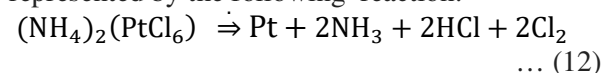
2-Finally, palladium is precipitated as diamine palladium (II) chloride by addition ammonia to the palladium chloride solution. Then HCl is added to be precipitated to a yellow salt as shown in the reactions below. A sponge of palladium metal is obtained by ignition of the palladium compound.

$$PdCl_4^{2-} + 4NH_3 \Rightarrow Pd(NH_3)_4^{2+} + 4Cl^- \quad …(13)$$

$$Pd(NH_3)_4^{2+} + 2HCl \Rightarrow Pd(NH_3)_2Cl_2 + 4NH_4^+ \quad …(14)$$

The aim of this study is to develop a hydrometallurgical process to leach honeycomb substrate of spent catalytic converters with aqua regia which is composed of (3HCl:1 HNO$_3$) followed by precipitation process for recovery of platinum and palladium. The optimum conditions for maximum leaching of platinum and palladium is investigate by using Taguchi Design. The various experiments are design by computer simulating software (Minitab) with leaching parameters, such as time, temperature, and S/L ratio. The effect of time, temperature, and S/L ratio on leaching platinum and palladium have been investigated. The possibility of recovery of platinum and palladium from leaching solution is investigate by the chemical precipitation using ammonium chloride (NH$_4$Cl) and sodium chlorate (NaClO$_3$) as precipitant agents for recovery of platinum and palladium, respectively.

2. Experimental Work

In this study, spent catalytic converters of BMW car were used for platinum and palladium recovery because the BMW car manufacturing company has a high quality and mandatory agreements in the use of PGM metals as a catalyst .Catalytic converter scrap was assembled from maintenance shops of catalytic converters. Opening way of the catalyst converter is carried out by flame cutting (torch), taking into consideration the clean cut so as not to monolith damage. After taking out monolith structure from the body, then cleaning was done by blowing air to get rid of dust and the remains of the combustion products .Fig.(3) represents flow chart for recovery of Pt, Pd from spent catalytic converter. Comminution is to be a two-stages, crushing, and grinding to get to the required particle size .The first stage was crushing by using cast iron mortar and pestle which it is similar to coffee grinder to reduce the honeycomb structure to a suitable size .The second stage was grinded by using a ball mill machine (Retsch). A ball mill is a fairly simple machinery consisting of a drum inside it three steel balls which rotates horizontally. (1250) grams of the sample powder was fed into the drum, the charge was divided into seven small weights, each run took up 10 minutes, magnet is used to get rid of ferrous contamination. The grinded powder was reduced in size to less than +250 µm at end of the operation.. Catalyst powder is washed several times with distilled water to get rid of dust and other pollutants. Then, the powder was placed in refractory dish for adding in an electric oven at 100°C for one hour to remove moisture, and also combustion products, coal, and gas. The powder sample was then sieved to (-250)µm,(+60 mesh) using mechanical auto sieve shaker (AD60-01) according to ASTM standard sieves . The laboratory technique used

dependent for sampling was by using the Chute Riffler. The chemical analyses of the catalytic powder is carried out by flame atomic absorption spectrometry (NOVAA-350).The analysis results are shown in table (1)as weight percentage.

Table 1,
Chemical composition of the catalytic powder as determined by the flame atomic absorption spectrometry.

Components	Al_2O_3	SiO_2	CaO	Fe_2O_3	Pd	Pt	Zn	Ag	Ni
Wt.%	43.608	10.502	0.0316	0.054	0.370	0.075	0.046	0.0007	0.0106
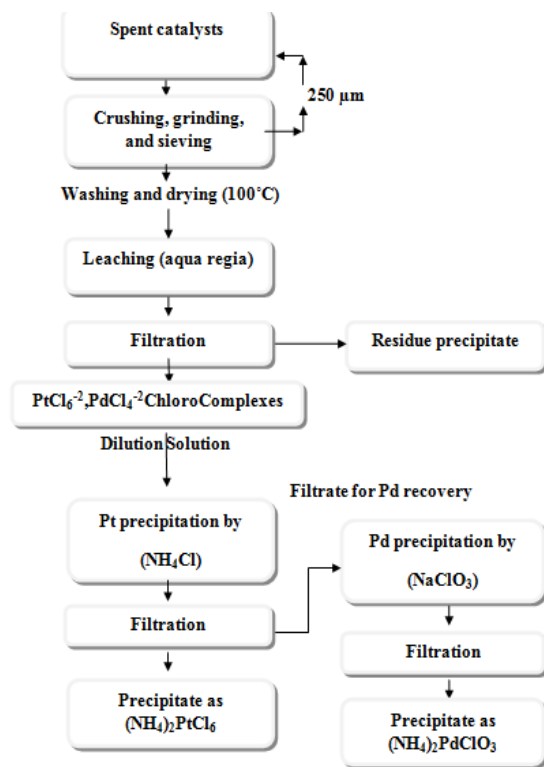
Fig. 3. Flow chart for recovery of Pt, Pd from spent catalytic converter.

The catalytic converter powder was dissolved in strong acidic solutions. Aqua regia solution ($3HCl:1HNO_3$) consisting of high purity hydrochloric acid (37%) and concentrated nitric acid (65%) were employed in all dissolution experiments.

3. Taguchi Method

In this study, experiments are designed by using Taguchi method to create three parameters, time, temperature, and solid/liquid ratio for the purpose of determining optimal conditions of leaching processes, unlike previous research that used the old traditional ways (classic ways). Three controlled factors and their levels are designed as given in table (2).

Table 2,
Three controlled factors and their levels are used in leaching process of the catalytic powder by aqua regia solution

Controlled factors	Levels			
	L1	L2	L3	L4
Time (min)	30	60	90	120
Temp(°C)	25	50	75	100
Solid/Liquid Ratio (wt/wt)	1/5	1/10	1/15	1/20

4. Selection of Orthogonal Array

Taguchi orthogonal array (OA) designs are usually used in design experiments on many factor levels. These standard arrays give the method to reduces the minimal number of experiments which could give the complete information of all the factors that have effects on the performance parameters .Orthogonal array notation is *La(b^c)* where *L* represents the orthogonal array notation in the table ,*a* is mean number of experiments ,*b* is mean number of levels ,and *c* is mean number of parameters[18].The design used in the experiments of leaching processes is Taguchi orthogonal arrays. By using the array selector, see table (3) three parameters and four levels were selected the proper array was *L16(4^3)*.

Table 3,
Array selector [17].

Orthogonal Array Selector

Number of Levels		2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31
	2	L4	L4	L8	L8	L8	L8	L12	L12	L12	L12	L16	L16	L16	L16	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32	L32
	3	L9	L9	L9	L18	L18	L18	L18	L27	L27	L27	L27	L27	L36	L36	L36	L36	L36	L36	L36	L36	L36	L36								
	4	L'16	L'16	L'16	L'16	L'32	L'32	L'32	L'32	L'32																					
	5	L25	L25	L25	L25	L25	L50	L50	L50	L50	L50	L50																			

Table 4,
Design of experiments used in the leaching process for the catalytic converter.

Exp.	Time (min)	Temp (°C)	Solid/Liquid Ratio (wt/wt)
1	30	25	1/5
2	30	50	1/10
3	30	75	1/15
4	30	100	1/20
5	60	25	1/10
6	60	50	1/5
7	60	75	1/20
8	60	100	1/15
9	90	25	1/15
10	90	50	1/20
11	90	75	1/5
12	90	100	1/10
13	120	25	1/20
14	120	50	1/15
15	120	75	1/10
16	120	100	1/5

5. Signal–Noise(S/N) Ratio

At parameters design, the factors were divided into two kinds: control factors which can be easily controlled by the experimenters and noise factors which can be difficult or expensive to control during operation. To determine (S/N) ratio, there are three types of formulations as shown in table (5).In this study, the second formula is used which represents the larger signal to noise ratio giving the best leaching efficiency [18].

Table 5,
S/N Ratios Formulations [19].

1-Lower is the better	$\frac{S}{N}$ = -10 log($\sum_{i=1}^{n} \frac{Yi^2}{n}$)
2-Higher is the better	$\frac{S}{N}$ = -10 log($\sum_{i=1}^{n} \frac{1}{nYi^2}$)
3-Nominal is the better	$\frac{S}{N}$ = 10 log($\sum_{i=1}^{n} \frac{Yi^2}{S^2}$)

6. Analysis of Variance (ANOVA)

Analysis of variance was used to evaluate the response value in percent (%) for each variable in the orthogonal experiments. In this study, it was used to identify two tries result for three parameters of leaching. The main calculation of ANOVA was that the total sums of the squares are equal to the sum of the SS for all parameters(time ,temperature, and solid/liquid ratio) and error as shown in equation (15) [20].

$$SS_T = SS_{time} + SS_{temp.} + SS_{s/l} + SS_{error}$$
… (15)

$$SS_T = \sum_{1}^{n} x_i^2 - \frac{G^2}{N}$$
… (16)

$$SS_C = \frac{\sum_{1}^{n} x_i^2}{n} - \frac{G^2}{N}$$
… (17)

Where :SS_T is total sums of squares parameters and error; G is sum of the results data for all the experiments; N is total number for all the experiments; i is level number; C is one of the tested parameters;$\sum x_i$ is sum of results for all experiments; i.e, at parameter c, and at level i; n is number of parameters. Degree of freedom was calculated by the following equation:

$$DOF_c = C - 1 \qquad \qquad \dots (18)$$
$$DOF_T = T - 1 \qquad \qquad \dots (19)$$

Where; C represents the levels for each parameter (controlled factor),T represents the number of experiments. Also, contribution percentage ($\rho\%$) was calculated for each controlled factor and for the error from the following equation[21]:

$$\rho\% = \frac{SS_C}{SS_T} \qquad \qquad \dots (20)$$

When:

$\rho_{error}\leq 15\%$,so the chosen controlled factors are not active.

$\rho_{error}\geq 50\%$, so chosen controlled factors are active.

Table 6,
ANOVA for the Platinum leaching by aqua regia solution for each controlled factor.

Factors	DOFc	SSc	%ρ
Time (min)	3	11.638	3.661
Temp(˚C)	3	42.265	13.299
Solid/Liquid Ratio(wt/wt)	3	204.817	64.447
Error	6	59.085	18.591
Total	15	317.805	100

Table 7,
ANOVA for the Palladium leaching by aqua regia solution for each controlled factor.

Factors	DOFc	SSc	%ρ
Time (min)	3	0.977	1.538
Temp(˚C)	3	4.610	7.259
Solid/Liquid Ratio(wt/wt)	3	44.67	70.34
Error	6	13.25	20.866
Total	15	63.50	100

7. Leaching Process

All experiments of leaching processes for catalytic powder containing platinum and palladium in aqua regia shown in the table (4) are subject to three variables (time, temperature, and solid/liquid ratio). The weight of the sample in each experiment was 10 grams with particle size (-250)µm. The dissolution process involved the following steps:

1-The HCl-HNO$_3$ mixture was prepared in a volume ratio 3HCl:1HNO$_3$ to form an acidic solution called aqua regia of a reddish brown color.

2-Aqua regia is heated by hot plate magnetic stirrer to the specified point according to each

experiment in a glass container containing three vents from up for (condenser ,thermometer, and for adding materials) with (400) rpm agitating for all experiments.

3-The catalytic converter powder is added to the glass container after heated aqua regia to a specific temperature at a specific time according to each experiment.

4-At the end of leaching process, filtration step is done using glass funnel and ashless filter paper (Whatman England /Circle=12.5cm) to separate the slurry from the solution.

5-After filtration stage, platinum and palladium concentration in the solution were determined by using atomic absorption spectrometry analysis (NOVAA-350).

6-In order to obtain accurate leaching rate, sixteen experiments of Taguchi method is performed twice.

7-The percentage of leaching for each experiment is calculated by the following formula:

$$\% \boldsymbol{Leaching} = \frac{C.V}{P.W}\boldsymbol{100}$$

Where: C= Concentration platinum or palladium after leaching (g/l); V=Volume of solution after leaching (l); W=Weight of catalytic powder in leaching solution (g); P=Percentage of platinum or palladium in catalytic converter powder (%).

8. Precipitation Process

The removal of metals (Pt and Pd) from chloride solutions at optimum condition of leaching using (NH$_4$Cl) and (NaClO$_3$) as the precipitant agents are investigated, unlike previous research that used precipitators Other. The percentage of recovery platinum and palladium are calculated by the following formula:-

$$\% \boldsymbol{Recovery} = \frac{C_1 L_1 - C_2 L_2}{C_1 L_1}$$

Where: C_1= The concentration of element before precipitation (g/l); L_1=Volume of solution before precipitation (l); C_2=The concentration of element after precipitation (g/l); L_2=Volume of solution after precipitation(l).The leaching and precipitation process involved the following steps:-

1-Leaching of (20) grams sample of catalyst powder with particle size (-250)µm in (320) ml of aqua regia at optimum condition (Time=120 min;Temp=100˚C;S/L=1/20).The concentration of platinum and palladium is determined by atomic absorption spectrometry analysis before

precipitation is carried out. 2-The diluted solution was prepared by boiling aqua regia solution to 100˚C, which containing platinum and palladium until the solution was condensed and mixed properly. Then cooled to 50˚C for adding HCl to get rid of the nitroso-compounds in the solution. (5) ml of hydrochloric acid were add to the solution every 10 minutes for ten times This process was conducted in moderate temperatures without allowing the solution to evaporate completely .The hot solution was diluted with four volumes of tap water in a large bowl and then filtered.

3-By dissolving (214) grams of ammonium chloride (NH_4Cl) in 1 litter of distilled water .The solution is used to precipitate platinum. 4-The diluted solution which contains platinum and palladium was heated at medium temperature (50˚C). Platinum was firstly precipitated by adding (5ml/l) of ammonium chloride solution, slowly with stirring continuously until the formation of yellowish orange solution ,after 1 hour the solution was filtrated , platinum was precipitated as a bright orange powder which is ammonium chloroplatinate $(NH_4)_2PtCl_6$ then concentration of platinum in the solution was determined by flame atomic absorption spectrometry analysis.

5- After platinum removal, palladium was precipitated from remaining filtrated solution by adding sodium chlorate ($NaClO_3$) ,the solution was placed in a beaker and boiled it at 100˚C,then it is cooled to just less boiling and (5g/l) of sodium chlorate crystals was added into beaker with shaking carefully until the formation of bright red solution , after 1 hour the solution was filtrated , palladium was precipitated as a bright red powder which is palladium ammonium chlorate $(NH_4)_2PdClO_3$ then the concentration of palladium in the solution was determined by flame atomic absorption spectrometry analysis.

9. Results& Discussion
9.1. Effect of Time

The time of platinum and palladium leaching process was studied. Two experiments whose durations varied (30, 60, 90, and 120) minutes, were carried out to discover the time advance of leaching in aqua regia solution. Figure (4) show platinum and palladium leaching by aqua regia solution as a function of time, respectively. Leaching efficiency of platinum and palladium started rising gradually at different times .The percentage of leaching of platinum was52.8% at 120 minutes, while the percentage of leaching palladium was 62.8% at 120 minutes.
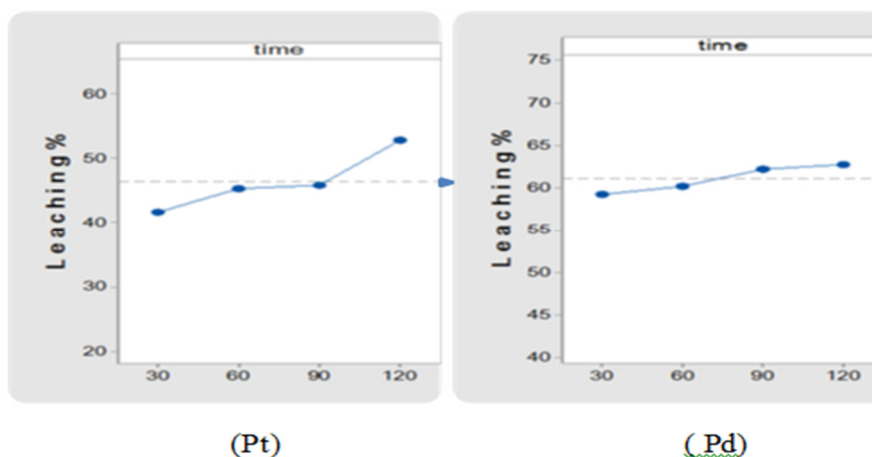

Fig. 4. Efficiency of platinum and palladium leaching by aqua regia solution for each level as a function of time.

9.2. Effect of Temperature

Platinum and palladium leaching kinetics is studied at four temperature levels (25, 50, 75, and 100) ˚C. Each leaching experiment was conducted at different times and solid to liquid ratios, but an agitation rate was fixed at (400 rpm).Results data of the two experiments of platinum and palladium leaching efficiency as a temperature function are presented in figure (5).They, indicate that the increase in temperature greatly improves the leaching kinetics of platinum and palladium .This

shows that the activation energy of the leaching reactions increases with increased temperatures. Platinum leaching efficiency was 52.9% achieved at 100°C, while Palladium leaching efficiency was 67% achieved at 100°C.
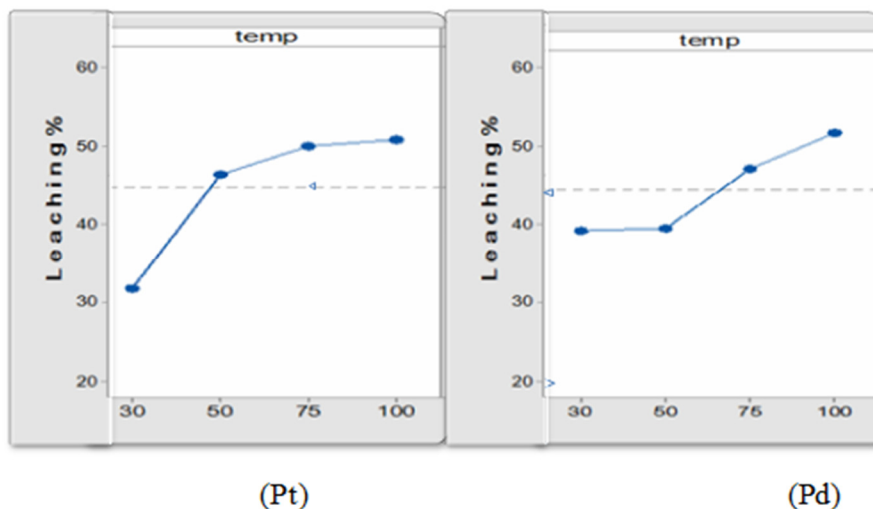

Fig. 5. Efficiency of platinum and palladium leaching by aqua regia solution for each level as a function of temperature.

9.3. Effect Solid/Liquid Ratio

The weight ratio of solid/liquid (wt/wt) was another investigated variable. Four levels 1/5, 1/10, 1/15, and 1/20 of S/L ratios were tested. Platinum and palladium leaching results for the two experiments are obtained as shown in figure (6) as S/L ratios function. The leaching kinetics in aqua regia solution becomes faster with increased S/L ratio. Platinum and palladium leaching efficiency reached to 62.9%, and 73.8%, respectively at1/20 as solid to liquid ratio.
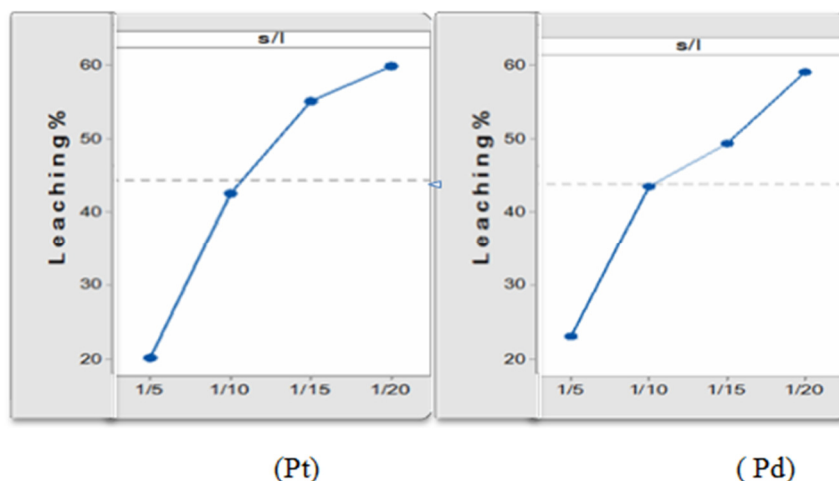

Fig. 6. Efficiency of platinum and palladium leaching by aqua regia solution for each level as a function of solid/liquid ratio.

10. Leaching Platinum and Palladium

The sample of catalyst powder was dissolved by using aqua regia solution (3HCl:HNO₃).HNO₃ was added to increase the oxidation of platinum and palladium during leaching .PtCl$_6^{2-}$ and PdCl$_4^{2-}$ were formed during the leaching process that undergone according to the reactions (8), and (9) .The color of chloro complex Pd is darker than of Pt. Color of filtrated leachate is not to be an indicator of platinum and palladium dissolution efficiency. Platinum and palladium were

completely leaching in aqua regia solution; the separation is more complicated between platinum group metals because they have similar chemical and physical properties. Increasing the concentration of hydrochloric acid as a volume unit with three times of nitric acid led to obtain high leaching of platinum and palladium were (97.58% ,93.23%) respectively.

11. Precipitation of platinum and palladium

Precipitation is considered one of the separation methods, precipitation step have a major influence on the recovery of platinum and palladium .Firstly ,through adding ammonium chloride (NH$_4$Cl)to the diluted solution, platinum complex (PtCl$_6^{2-}$)was precipitated because its less soluble according to precipitation reaction (see equ(11)).While, palladium complex(PdCl$_4^{2-}$) remained dissolved in the diluted solution. Secondly, palladium complex (PdCl$_4^{2-}$) was precipitated by adding sodium chlorate (NaClO$_3$) to the diluted solution. Temperatures had a clear effect on the precipitation process, all the platinum group metals complexes are fast influenced to hydrolysis, especially at high temperatures. After completion of the precipitation, the results of the analysis show that the platinum and palladium content could completely precipitate from the catalytic converter, and this has been proved possibility of extracting efficiently. The percentage of efficient recovery of platinum and palladium were (91.94%, and 98.82%) respectively.

12. Conclusion

Based on this study of the recovery of platinum and palladium from spent catalytic converter, using aqua regia solution, the following conclusions can be drawn:
1. The percentages of platinum and palladium in the powder sample of spent catalytic converter were 0.075% and 0.37%, respectively based on chemical analysis by flame atomic absorption.
2. Leaching experiments have been done and chosen by Taguchi Design to find optimum conditions to leach platinum and palladium by aqua regia. Effect of time, temperature, and S/L ratio on leaching efficiency are investigated .The relationship was direct between effect of time, temperature, and S/L ratio on leaching efficiency. The increase of

the level of each effective tends to increase the leaching efficiency of platinum and palladium.
3. The optimum dissolution of platinum and palladium by aqua regia were (97.58%) ,and (93.23%) ,respectively at a time of 120 minutes, temperature 100°C,and solid/liquid ratio 1/20.   .
4. Recovery of platinum and palladium was possible by the precipitation using ammonium chloride (NH4Cl) and sodium chlorate (NaClO3) as precipitation agents at 50°C for 1 hour. Recovery efficiency of platinum was 91.94%, while recovery efficiency of palladium was 98.82%.

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استرجاع البلاتين والبلاديوم من مخلفات المحول الحفاز في السيارات

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الخلاصة

الطريقة المائية كانت قد استخدمت في اذابة عنصري البلاتين والبلاديوم في محلول الماء الملكي(3HCl: HNO$_3$) .وكانت صممت تجارب الاذابة لمعرفة الظروف المثالية للاذابة بواسطة طريقة تاكوشي بواقع ستة عشر تجربة عند ثلاث عوامل مؤثرة هي درجة الحرارة والوقت ونسبة الصلب الى السائل.اذابة مسحوق المحول الحفاز الذي يحتوي على البلاتين والبلاديوم التي كانت قد اجريت على اساس تشكيل مركبات كلوريدية من البلاتين والبلاديوم (PtCl$_6^{2-}$, PdCl$_4^{2-}$) بتراكيز مختلفة في المحلول الحامضي.الظروف المثالية لعملية الاذابة كانت (درجة الحرارة = ,100°C والوقت=١٢٠ دقيقة ونسبة الصلب الى السائل=٢٠/١)وكانت افضل كفاءة اذابة عند هذه الظروف للبلاتين(٩٧,٥٨%) والبلاديوم(٩٣,٢٣%).بعد عملية الاذابة عند الظروف المثالية ,كان قد ترسب البلاتين اولا على شكل (NH$_4$)$_2$PtCl$_6$ (ammonium chloroplatinate) بواسطة اضافة (٥ مل لكل لتر) من كلوريد الامونيوم ammonium chloride (NH$_4$Cl) كعامل مرسب,بعد ذلك كان ترسب البلاديوم على شكل (NH$_4$)$_2$PdClO$_3$ (palladium ammonium chlorate) بواسطة اضافة (٥ غم لكل لتر) من كلورات الصوديوم (NaClO$_3$) sodium chlorate) كعامل مرسب الى المحلول المتبقي بعد ازالة البلاتين.وكانت افضل كفاءة استرجاع عند ظروف الاذابة المثالية للبلاتين والبلاديوم هي (91.94% و 98.82%) على التوالي) .

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