IOP Conference Series: Materials Science and Engineering

Time-Dependent Ultrasonic Assisted Recovery of Platinum from Spent Removing Catalyst of Pt/Al_2O_3 by Acid Leaching

IOP Conf. Series: Materials Science and Engineering **515** (2019) 012068 doi:10.1088/1757-899X/515/1/012068

Time-Dependent Ultrasonic Assisted Recovery of Platinum from Spent Removing Catalyst of Pt/Al_2O_3 by Acid Leaching

R Kurniawan[1], M S Nuroni[1], A A Fibriyanti[1], J Utomo[1], A Fuad[1,2], N Mufti[1,2], E Latifah[1,*]

[1] Department of Physics, Faculty of Mathematics and Natural Sciences, Universitas Negeri Malang, Jl. Semarang 5, Malang 65145, East Java, Indonesia
[2] Central Laboratory of Mineral and Advanced Materials, Faculty of Mathematics and Natural Sciences, Universitas Negeri Malang, Jl. Semarang 5, Malang 65145, East Java, Indonesia

*Corresponding author's email: eny.latifah.fmipa@um.ac.id

Abstract. A standard recovery of platinum (Pt) in a spent removing catalyst of Pt/Al_2O_3 was performed by acid leaching of aqua regia. The time-dependent extraction of Pt from Pt/Al_2O_3 showed that the percentage of the Pt in Pt/Al_2O_3 decreased by increasing the extraction time, which indicated an increase of the Pt in the leachate. Furthermore, a time-dependent ultrasonic-assisted extraction of Pt from Pt/Al_2O_3 was also performed. A small number of Pt content in the Pt/Al_2O_3 obtaining ultrasonic method revealed that the ultrasonic effectively enhanced the Pt extraction compared with standard extraction due to the agitation of high-frequency pressure (sound) waves. The further morphological analysis was also performed to study the morphological modification of Pt/Al_2O_3. Our results provide a good understanding of the recovery of Pt from spent removing catalyst of Pt/Al_2O_3 and give an alternative method to increase the Pt extraction.

Keywords: Pt/Al_2O_3 catalyst, acid leaching, ultrasonic treatment

1. Introduction

Platinum (Pt) is one of the precious metals that have a high economic value, besides gold, silver, and palladium [1]. Platinum possesses unique characteristics, such as high electrical conductivity, resistance to corrosion and oxidation, catalytic activity and high melting point [2]. Unlike gold, platinum is widely used in industrial sectors, such as automobiles [3], medical devices [4], jewelry [5], and catalysts [6]. Lee *et al*. reported that the presence of platinum in the earth's crust as only 0.003 ppb, 30 times smaller than gold [7]. The latest statistical data shows that the problem comes from the global economic sector. Here, the demand for Pt in the market is increasing, while the supply is constrained due to its limited availability in nature [8].

Platinum replacement with other metals is difficult to do due to its special characteristics [9]. Because of the limited natural resources of platinum, the recovery of Pt from industrial waste is very crucial to overcoming the demand for Pt in the global economic sector, such as recovery of Pt from industrial waste [10-12]. A new technique for Pt extraction using cyanide solutions was reported, but this

technique was high cost and cause an environmental problem [13]. Other methods were also reported, where iodine solution [14], $KHSO_4$ solvent [15], and oxalic acid [16] were used as leachate. However, these methods require a difficult work condition, such as high-speed stirring, and yield a small number of Pt.

In this study, a method of recovery of Pt has been proposed. The recovery of Pt from Pt/Al_2O_3 catalyst is carried out using aqua regia supported by ultrasonic treatment. Generally, ultrasonic treatment has been used for sonication-based material cleaning [17-19]. The role of ultrasonic treatment on surface change of Pt/Al_2O_3 and Pt content on Al_2O_3 balls are investigated by comprehensive studies. Our result could be used as an alternative to optimize the series of Pt extraction procedure to overcome the demand for Pt in the global economic sector.

2. Methods

Platinum (Pt) metal was separated from Pt/Al_2O_3 catalyst using the acid leaching method. Pre-treatment procedure was performed by annealing of the Pt/Al_2O_3 at temperature 500°C for 5 hr. Here, aqua regia was used as the leachate. Aqua regia was prepared by mixing HNO_3 and HCl with a ratio of 3:1. In this study, two types recovery treatment was performed; Pt/Al_2O_3 catalyst was leached in aqua regia with the duration of 10 and 20 min (type 1) and the Pt/Al_2O_3 catalyst was leached in aqua regia supported by ultrasonic treatment with the duration of 10 and 20 min (type 2). Furthermore, Pt/Al_2O_3 balls were separated from the solution, so that the solution contains aqua regia and leaching product. The detail of the leaching procedure is presented in Figure 1. In this study, leached Al_2O_3 balls were investigated to determine the reduction of Pt content on Al_2O_3 balls. The morphology of the samples was analyzed using surface mapping from scanning electron microscopy (SEM) measurement; FEI-INSPECT S50. Furthermore, the material composition of the samples was confirmed using the X-ray fluorescence (XRF); PANalytical, MiniPal 4.

3. Results and Discussion

Figure 2 presents a photograph of Pt/Al_2O_3 catalyst before and after the leaching process. Here, two type treatments were performed; type 1 (leaching) and type 2 (leaching supported by ultrasonic treatment). Here, the annealed Pt/Al_2O_3 sample has a grayish black color. We found that the color of the sample changed from grayish black to become white after leaching treatment. Here, white color is ascribed to the disintegration of materials from the surface of the Al_2O_3 balls. Moreover, the increase of leaching duration promoted a significant color change. To confirm the disintegration of materials the surface of the Al_2O_3 balls, the morphological investigation was performed. Here, surface changes of Pt/Al_2O_3 were investigated using surface mapping from SEM.

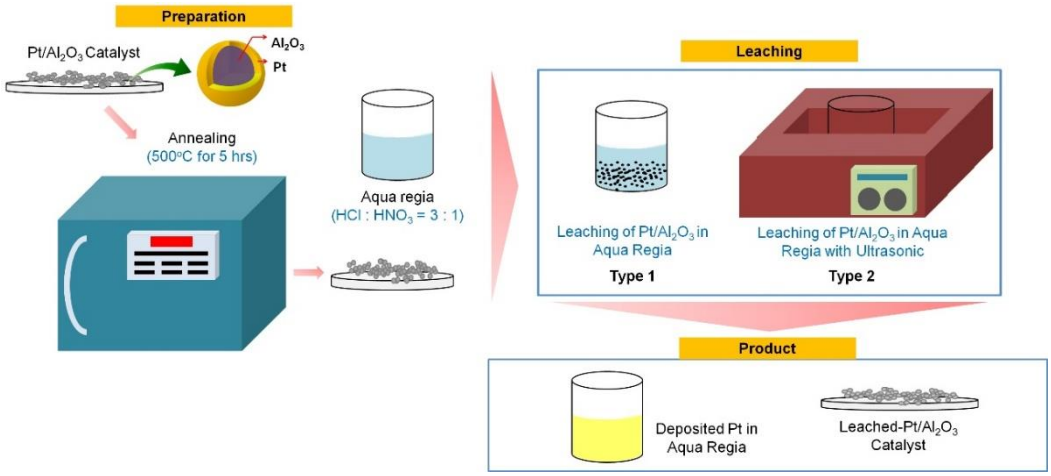


Figure 1. Schematic illustration of recovery of Pt from Pt/Al_2O_3 catalyst by using aqua regia leaching. The leaching was performed without and with ultrasonic treatment.

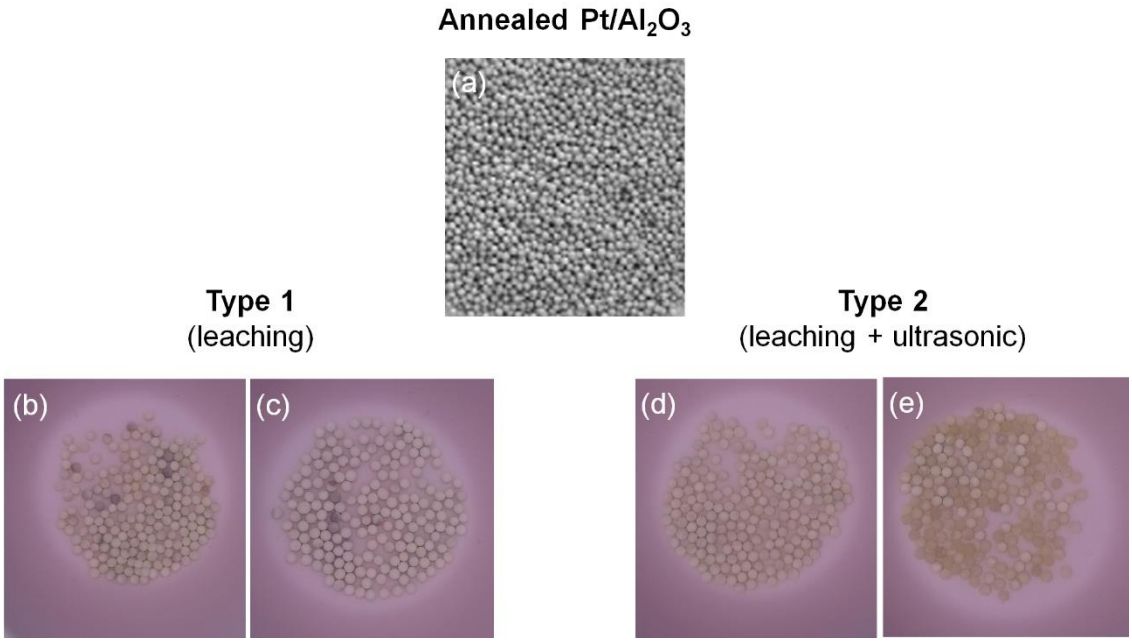


Figure 2. The color change of Pt/Al_2O_3 samples; annealed Pt/Al_2O_3 at 500 °C (a), leached Pt/Al_2O_3 for 10 min (b) and 20 min (c), leached Pt/Al_2O_3 with ultrasonic for 10 min (d) and 20 min (e).

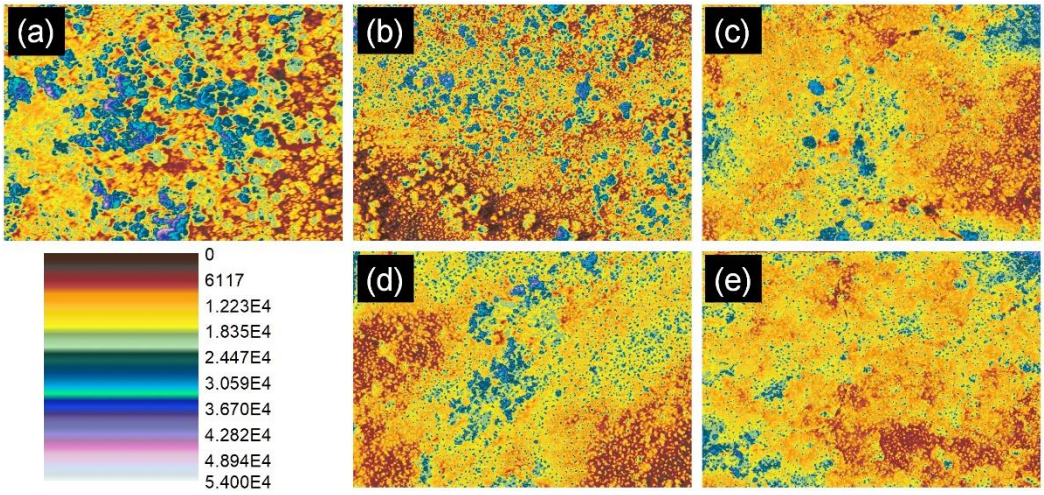
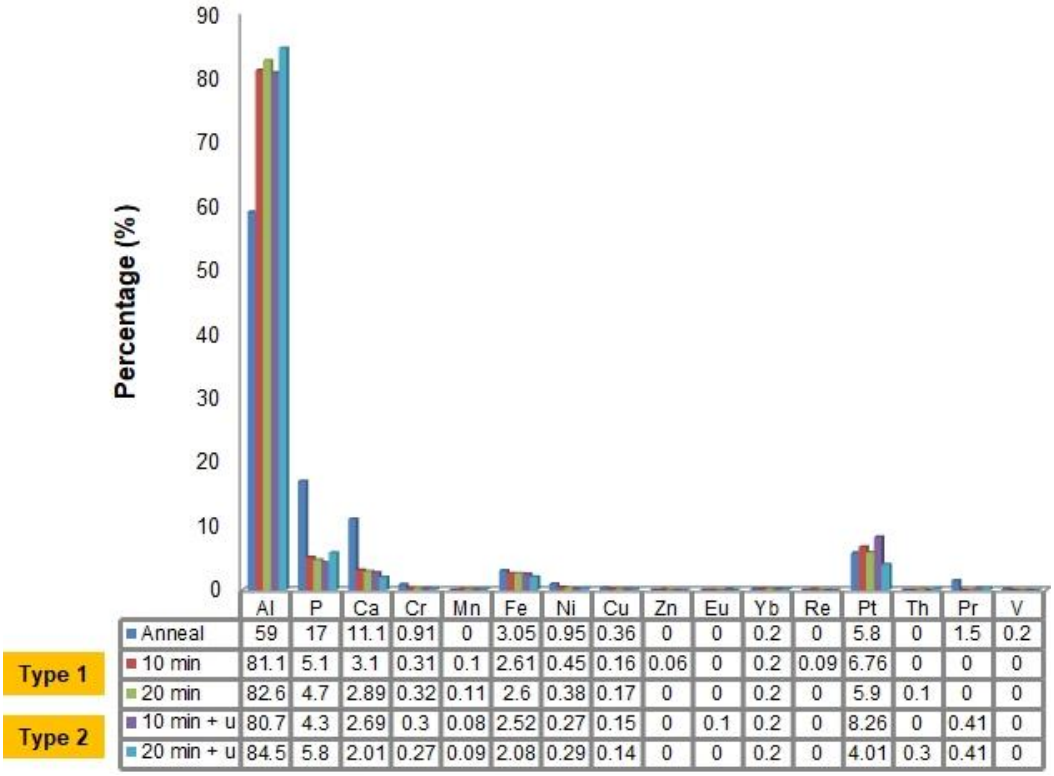


Figure 3. Surface mapping of Pt/Al_2O_3 samples; (a) annealed Pt/Al_2O_3 at 500 °C, (b) leached Pt/Al_2O_3 for 10 min, (c) 20 min, (d) leached Pt/Al_2O_3 with ultrasonic for 10 min, and (e) 20 min.

Table 1. The Population of Pt on the surface of the Al_2O_3.

Samples		Duration (min.)	Population of Pt (in %)
Annealing			71
Leaching	Type-1 (leaching)	10	59.5
		20	25
	Type-2 (leaching + ultrasonic)	10	50.2
		20	11.7



		Al	P	Ca	Cr	Mn	Fe	Ni	Cu	Zn	Eu	Yb	Re	Pt	Th	Pr	V
	Anneal	59	17	11.1	0.91	0	3.05	0.95	0.36	0	0	0.2	0	5.8	0	1.5	0.2
Type 1	10 min	81.1	5.1	3.1	0.31	0.1	2.61	0.45	0.16	0.06	0	0.2	0.09	6.76	0	0	0
	20 min	82.6	4.7	2.89	0.32	0.11	2.6	0.38	0.17	0	0	0.2	0	5.9	0.1	0	0
Type 2	10 min + u	80.7	4.3	2.69	0.3	0.08	2.52	0.27	0.15	0	0.1	0.2	0	8.26	0	0.41	0
	20 min + u	84.5	5.8	2.01	0.27	0.09	2.08	0.29	0.14	0	0	0.2	0	4.01	0.3	0.41	0

Figure 4. The Composition of Pt/Al_2O_3 samples.

Figure 3 shows the evolution of the sample surface after the leaching process. Surface mapping shows that Pt particles were observed on the surface of the Al_2O_3 balls, where Pt is agglomerated and forming clusters. Here, Pt population is indicated by the blue color, where the color scale represents the material thickness. Surface mapping confirms that Pt population decreases when a long leaching duration is performed. It can be shown that leaching treatment (type 1) for 10 min promotes the disintegration of Pt the surface of the Al_2O_3 balls. The increase of leaching duration for 20 min showed a higher disintegration than that of leaching for 10 min. The disintegration of Pt was also observed in leached Pt/ Al_2O_3 supported by ultrasonic treatment (type 2), where samples with leaching duration for 20 min had higher disintegration than the sample with leaching duration for 10 min. We found that type 2 treatment had a higher disintegration than type 1 treatment. Furthermore, Pt population is estimated by comparing the distribution of Pt with the total sample distribution. List of Pt population is presented in Table 1.

Furthermore, changes in Pt content in the sample have been investigated using XRF. Figure 4 presents the material content/composition of the Pt/ Al_2O_3 samples. This result indicates that the percentage of Pt decreases with the increase of leaching duration, which has good agreement with the results above. In addition, XRF result provides the explanation of the origin of grayish black appearance in the annealed Pt/ Al_2O_3 sample. The annealed Pt/ Al_2O_3 sample contained a high P and Fe materials compared to other samples. Previous research has confirmed that the Al_2O_3 -based materials with P and Fe in the form of oxide have dark (black) appearance [20].

The results above show that our procedure can increase the recovery of Pt from Pt/ Al_2O_3. The ultrasonic treatment can support effectively the disintegration of Pt from Al_2O_3 balls. The high-frequency wave can create agitation in liquids, which is produced from high-pressure sound waves and subsequently provide a sufficient force to disintegrate Pt from Al_2O_3 balls. This combination of force (from ultrasonic) and chemical reactions of the leaching process can optimize the disintegration of Pt from Al_2O_3 balls.

4. Conclusion

Recovery Pt from Pt/Al_2O_3 has been conducted by leaching treatment with varying the leaching duration. The role of the ultrasonic treatment on the leaching process has been studied. We found that the ultrasonic treatment successfully increased the disintegration of Pt from Pt/Al_2O_3, which was confirmed from the surface mapping. We confirmed that the increase of leaching duration promoted the increase of disintegration of Pt from Pt/Al_2O_3 in both (leaching with and without ultrasonic) treatments. Moreover, the disintegration of the materials from the surface of the Al_2O_3 balls can be observed from the color change of the sample from grayish black to white. This condition is suggested from the disintegration of P and Fe materials. The disintegration of Pt and other materials from the surface of the Al_2O_3 balls has been confirmed from the results of XRF. However, the extraction process of Pt from other materials still needs to be done, considering that our treatment also disintegrates other elements, besides Pt. Our results can be used to optimize the next stage in the series of recovery processes of Pt/Al_2O_3.

References

[1] Figuerola-Ferretti I and McCrorie J R 2016 *J. Empir. Financ.* **38** 717-38
[2] Reardon A C 2011 *Metallurgy for the Non-Metallurgist, Second Edition* (Ohio, USA: ASM International) p 327-328
[3] Alonso E, Field F R and Kirchain R E 2012 *Environ. Sci. Technol.* **46** 12986-93
[4] Cowley A and Woodward B 2011 *Platin. Met. Rev.* **55** 98-107
[5] Klotz U E and Drago T 2011 *Platin. Met. Rev.* **55** 20-7
[6] Seo Y and Morimoto S 2017 *Resources* **6** 61
[7] Lee J-Y, Kumar J R, Kim J-S, Kim D-J and Yoon H-S 2009 *J. Ind. Eng. Chem.* **15** 359-64
[8] Wilson P 2018 *Platinum Quarterly Q2 2018* (London: World Platinum Investment Council Ltd) p 1-20
[9] Rao C R M and Reddi G S 2000 *Trends Analyt. Chem.* **19** 565-86
[10] Marinho R S, Afonso J C and da Cunha J W S D 2010 *J. Hazard. Mater.* **179** 488-94
[11] Wongkaew K, Wannachod T, Mohdee V, Pancharoen U, Arpornwichanop A and Lothongkum A W 2016 *J. Ind. Eng. Chem.* **42** 23-35
[12] Taninouchi Y-k and Okabe T H 2018 *Metall. Mater. Trans. B* **49** 1781-93
[13] Chen J and Huang K 2006 *Hydrometallurgy* **82** 164-71
[14] Zanjani A and Baghalha M 2009 *Hydrometallurgy* **97** 119-25
[15] Batista S G and Afonso J C 2010 *J. Hazard. Mater.* **184** 717-23
[16] Masuda C, Yonezu K, Watanabe K and Yokoyama T 2013 *Procedia Earth Planet. Sci.* **6** 435-40
[17] Tuziuti T 2016 *Ultrason. Sonochem.* **29** 604-11
[18] Tangsopa W, Keawklan T, Kesngam K, Ngaochai S and Thongsri J 2018 *IOP Conf. Ser. Earth Environ. Sci.* **159** 012042
[19] Yamashita T, Yamauchi R and Ando K 2018 *Jpn. J. Multiph. Flow* **32** 210-7
[20] Radivojević M 2015 *Cambridge Arch. J.* **25** 321-38

Acknowledgments

This research was supported by the Ministry of Research, Technology, and Higher Education of the Republic of Indonesia through Penelitian Terapan Unggulan Perguruan Tinggi (PTUPT) 2018. The authors acknowledge Central Laboratory of Mineral and Advanced Materials, Faculty of Mathematics and Natural Sciences, Universitas Negeri Malang for the characterization facilities.