IOP Conference Series: Materials Science and Engineering

PAPER • **OPEN ACCESS**

Study: leaching of zinc dust from electric arc furnace waste using oxalic acid

To cite this article: R Kusumaningrum *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **478** 012015

View the article online for updates and enhancements.

This content was downloaded from IP address 123.113.27.253 on 15/10/2019 at 17:13

Study: leaching of zinc dust from electric arc furnace waste using oxalic acid

R Kusumaningrum[1], A Fitroturokhmah[1], G S T Sinaga[1], A S Wismogroho[1,2], W B Widayatno[1,2], L J Diguna[3] and M I Amal[4]

[1]Nano Center Indonesia, PUSPIPTEK South Tangerang, Banten 15314 Indonesia
[2]Research Center for Physics, Indonesian Institute of Sciences, Bld. 440/442, PUSPIPTEK Serpong, Tangerang Selatan 15314, Indonesia
[3]Department of Renewable Energy Engineering, Prasetiya Mulya University, Kavling Edutown I.1, Jl. BSD Raya Utama, BSD City, Tangerang 15339, Indonesia
[4]Research Center for Metallurgy and Materials, Indonesian Institute of Sciences, PUSPIPTEK Tangerang Selatan, Banten 15314, Indonesia

Email: muha137@lipi.go.id

Abstract. A study has been conducted on the process of leaching of zinc dust using oxalic acid by hydrometallurgical method. The sample preparation process starts from the dust zinc washing. The leaching process was started by mixing the sample with solution of 5 M oxalic acid. There were three variations; the ratio of mole, temperature and leaching time to know the optimum condition. From the variation of leaching process that has been done, an optimum condition was obtained where the ratio of zinc mole dust to 5 M oxalic acid solution is 1:10 with temperature 90°C and leaching time for 150 minutes. The leaching filtrate was precipitated to obtain zinc oxalate with a solution of sodium hydroxide up to pH 7. The FTIR and XRD analysis showed that the leaching products confirmed as hydrozincite ($Zn_5(CO_3)_2(OH)_6$) and sodium bicarbonate ($NaHCO_3$). The carbonate compound may be caused by decarboxylation during the leaching process. The calcination of leaching products to convert hydrozincite turn out zinc oxide was done at 600 °C. The results of the XRD analysis show that there were zinc oxide and still found zinc carbonate and sodium carbonate.

1. Introduction

Zinc is very useful in many aspect of human life. For instance, in human body zinc have a role in growth and as an integral part of more than 200 enzymes [1,2]. Besides from natural resources, the utilization of zinc dust from electric arc furnace (EAF) can be used as an alternative source to obtain zinc. This dust contains a huge amount of zinc that proportional to 20-30 wt% zinc oxides [3].

Some studies to leach zinc from its dust have been reported. Two common methods for zinc dust recovery are pyrometallurgy and hydrometallurgy. The pyrometallurgy method requires expensive cost and high energy. Poorly, this method generated heavy and toxic air pollution that difficult to handling. The

Content from this work may be used under the terms of the Creative Commons Attribution 3.0 licence. Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.
Published under licence by IOP Publishing Ltd

hydrometallurgy method has been known as an easy and simple technique. This method is feasible for scaling up to build a pilot plan. The important process in hydrometallurgy is leaching which means an extraction of value metal from its source using acid, base or water [4]. Mostly, leaching process is brought out in acidic solution such as sulfuric acid, nitrate acid and chloric acid. Yet, this waste treatment is harmful and needs high treatment cost.

Oxalic acid is a strong organic acid, environmental friendly and usually used as a leaching agent. Oxalic acid is suitable as selective precipitating agent for zinc and iron separation based on the solubility product constant (Ksp) values which 1.4 x 10^{-9} for zinc oxalate and 2 x $10^{-7}$for iron oxalate [5]. The previous study gives information that zinc oxalate can be precipitated at pH 6.2-6.4 [6]. The leaching study of zinc dust using oxalic acid has not been reported intensively. In this study, oxalic acid was used as a leaching agent and sodium hydroxide as a precipitating agent of zinc oxalate.

2. Experimental Procedures

Zinc dust that used in this study was a waste of steel processing using electric arc furnace. Zinc dust was analyzed by X-Ray Diffraction (XRD) to confirm the containing phase in this dust. To remove the polar impurity, zinc dust was washed by demineralization water at 60 °C for one hour. The residues were dried at 100 °C.

Leaching process of zinc dust was driven by oxalic acid 5 M with mole ratio solid/liquid (s/l) 1/5 and 1/10. The leaching time and temperature was studied by calculation of yield product percentage. Zinc oxalate was precipitated by NaOH addition till reach pH 6-12. The white powder then was calcined at 600 °C to form zinc oxide. The leaching product and as-calcined product was analysis by X-Ray Diffraction and FTIR.

3. Result and Discussion

Zinc dust of EAF process was analyzed using XRD for analyzing the phase elements content. The percentages of elements content were analysis using X-Ray Fluorescence (XRF). The XRD pattern of zinc dust (Figure 1) showed that all phase was detected refers to ZnO. The content of elements in its dust was confirmed 96.3% Zn and 3.6 % Fe. Hence, the purification of zinc dust has to be examined.

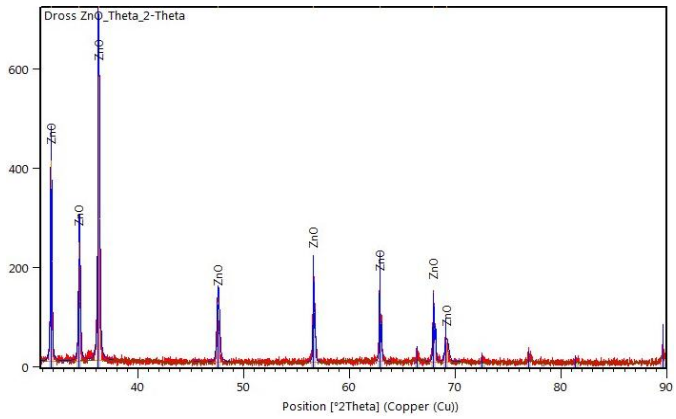


Figure 1. Diffractogram of zinc dust waste

Ratio mole s/l of zinc dust and oxalic acid solution were studies at 1:5 and 1:10 to investigate the optimum leaching condition. The reaction between oxalic acid and zinc dust (ZnO) were controlled at 60

°C for 120 minutes to form zinc oxalate. To precipitate the zinc oxalate, the sodium hydroxide was added until the pH of solution reach into 7 at room temperature. The calculation of yield percentage gave that mole ratio of s/l for 1:5 and 1:10 was 46.03% and 69.48%, respectively. The most effective leaching was given by the mole ratio of 1:10. It may be due to a great amount of oxalate ion that sufficient to extract more zinc from its dust. The possible reaction can be written as follows:

1. Oxalic acid running through dissociation in water:

$H_2C_2O_{4\,(aq)} \leftrightarrow H^+ + HC_2O_4^-$ (1)

$HC_2O_4^-{}_{(aq)} \leftrightarrow H^+ + C_2O_4^-$ (2)

Over all reaction could be written as

$H_2C_2O_{4\,(aq)} \leftrightarrow 2H^+ + C_2O_4^{2-}$ (3)

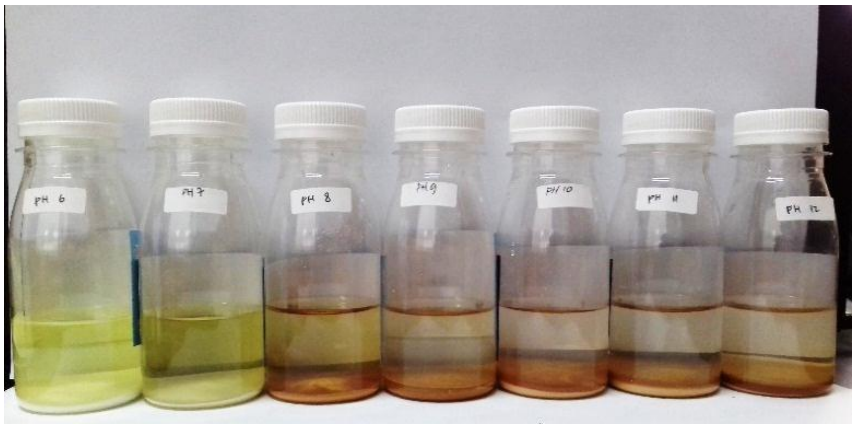
2. Extraction process to form zinc oxalate:

$ZnO_{(s)} + 2H^+ + C_2O_4^{2-} \rightarrow ZnC_2O_{4\,(aq)} + H_2O_{(l)}$ (4)

$FeO_{(aq)} + 2H^+ + C_2O_4^{2-}{}_{(aq)} \rightarrow FeC_2O_{4\,(aq)} + H_2O_{(l)}$ (5)

$Fe_2O_{3\,(aq)} + 2H^+ + C_2O_4^{2-}{}_{(aq)} \rightarrow Fe_2(C_2O_4)_{3\,(aq)} + H_2O_{(l)}$ (6)

The effect of pH in zinc oxalate precipitation was also studied. In pH 6 and 7, it was shown that the white precipitate was formed. Meanwhile, the addition of sodium hydroxide to obtain a base condition (pH 8-12) generated a brown precipitate that probably the iron elements. The addition of sodium hydroxide till pH 7 only resulted a zinc oxalate precipitation. The difference of Ksp value between zinc oxalate and iron oxalate which are 1,4 x 10^{-9} and 2 x 10^{-7}, respectively, may have caused the zinc oxalate to precipitate earlier. The precipitation was occurred at room temperature spontaneously.



pH 6 7 8 9 10 11 12

Figure 2. Zinc oxalate precipitation by pH 6-11

The effect of temperature and time in leaching process were studied twice for every procedure. The yield percentage is shown in Figure 3. The increasing temperature in leaching process caused an increasing of zinc oxalate precipitation. It may be caused by the increasing of collision among molecules when more zinc was extracted. The optimum temperature was 90 °C with a yield percentage of 57.32%. For reaction time effect study, the leaching was done at 90 °C. Such as reaction time effect, the yield percentage increased significantly till reaction time of 150 minutes (65.78%). Characterization of zinc

oxalate and zinc oxide synthesis products were taken at optimum condition based on yield percentage calculation which was temperature of 90 °C and reaction time of 150 minutes.

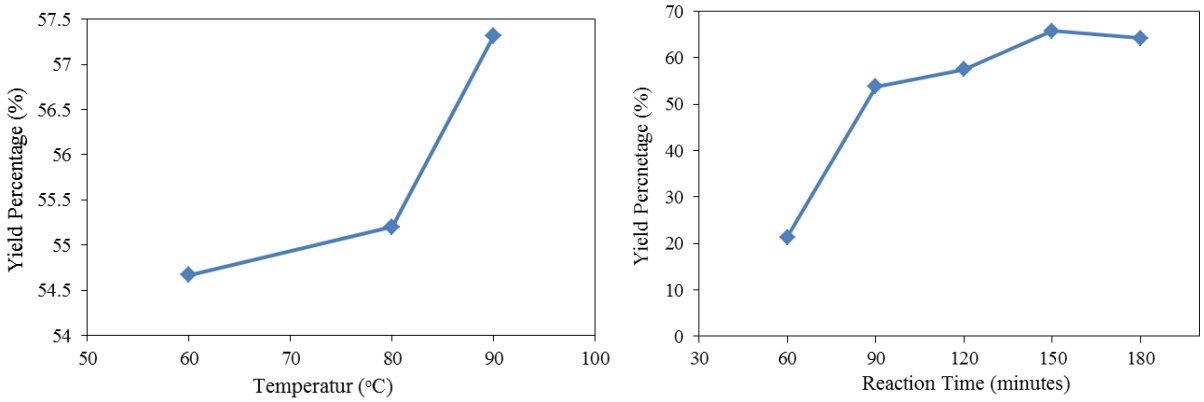


Figure 3. Yield percentage of leaching products by temperature and reaction times

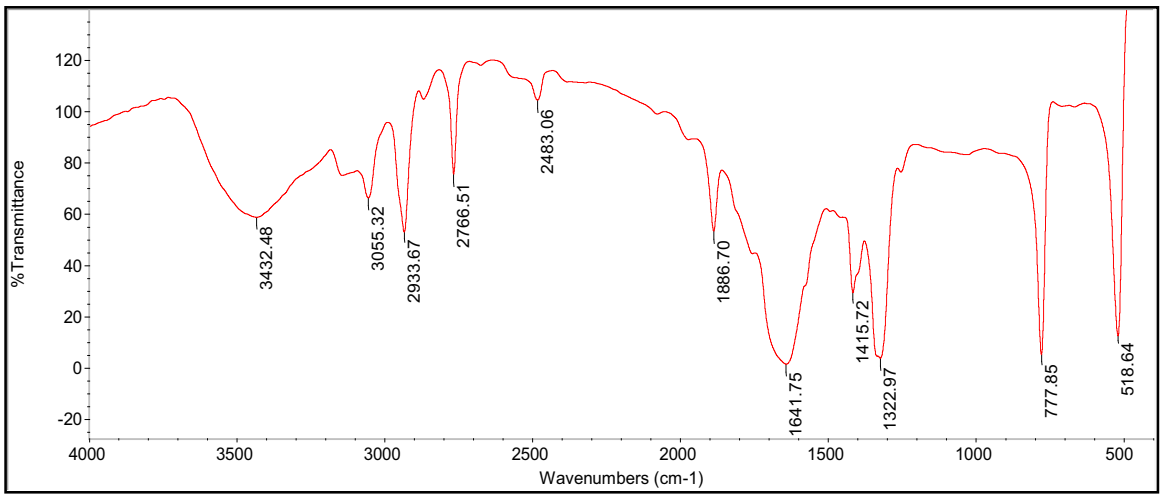


Figure 4. Spectra of leaching products

Zinc oxalate was characterized by FTIR is shown in Figure 4. Wavelengths number at 518 cm^{-1} referred to vibration of metal oxygen bonding. Wavelengths number give strong peak on 759 cm^{-1} ($\nu_{O=C=O}$ symmetric), 1349 cm^{-1} (ν_{C-O}), 1415 cm^{-1} (ν_{C-C}), 1655 cm^{-1} (ν_{C-O} asymmetric), and 2933 cm^{-1} (ν_{C-H}). The weak peak supported $\nu_{O=C=O}$ at 1886 cm^{-1} and 2483 cm^{-1}. The wavelengths number at 3432, 3055 and 2766 cm^{-1} corresponding to water molecule that trapped crystal lattice. Confirmation of phase leaching product was analyzed by XRD. The pattern of XRD (Figure 6) showed that leaching product were in dominant by sodium bicarbonate, partly peaks as zinc oxide and the rest as hydrozincite ($Zn_5(CO_3)_2(OH)_6$). Carbonate could be emerged by decarboxylation during the leaching process. Probably after dissociation process of

oxalic acid, the oxalate ions went through carbonate by carboxyl group release. This phenomenon caused the leaching products were in carbonate compound. Combine with FTIR analysis, sodium bicarbonate supported by appearance of C-H vibration at 2933 cm^{-1}. In line with the calcination product at 600 °C, XRD analysis confirmed into sodium carbonate, zinc carbonate and zinc oxide. Zinc carbonate occurred from dehydroxylation of hydrozincite. The other assume is zinc carbonate forming occur due to the existence of CO_2 in a large amount in the system and enable to drag the equilibrium of reaction into zinc carbonate formation. The other studies reported that carbon balance at atmosphere take a rule in the hydration-carbonation or hydration and carbonation reaction path in the $ZnO\text{-}CO_2\text{-}H_2O$ system [7].

1. Decarboxylation turn into carbonate acid

$$H_2C_2O_{4\,(aq)} \leftrightarrow CO_{\,(g)} + H_2CO_{3\,(aq)} \quad (7)$$

2. Hydrozincite Forming Reaction

$$5ZnO_{\,(s)} + 3\,H_2CO_{3\,(aq)} + NaOH_{\,(aq)} \rightarrow Zn_5(CO_3)_2(OH)_{6\,(s)} + NaHCO_{3\,(s)} \quad (8)$$

3. Zinc oxide Forming at 600 °C

$$Zn_5(CO_3)_2(OH)_{6\,(s)} \rightarrow 2\,ZnCO_{3\,(s)} + 3\,Zn(OH)_{2\,(s)} \quad (9)$$

$$Zn(OH)_{2\,(s)} \leftrightarrow ZnO_{\,(s)} + H_2O_{\,(g)} \quad (10)$$

$$ZnCO_{3(s)} \leftrightarrow 2\,ZnO_{\,(s)} + CO_{2\,(g)} \quad (11)$$

$$2NaHCO_{3\,(aq)} \rightarrow Na_2CO_{3\,(s)} + H_2O_{\,(g)} + CO_{2\,(g)} \quad (12)$$

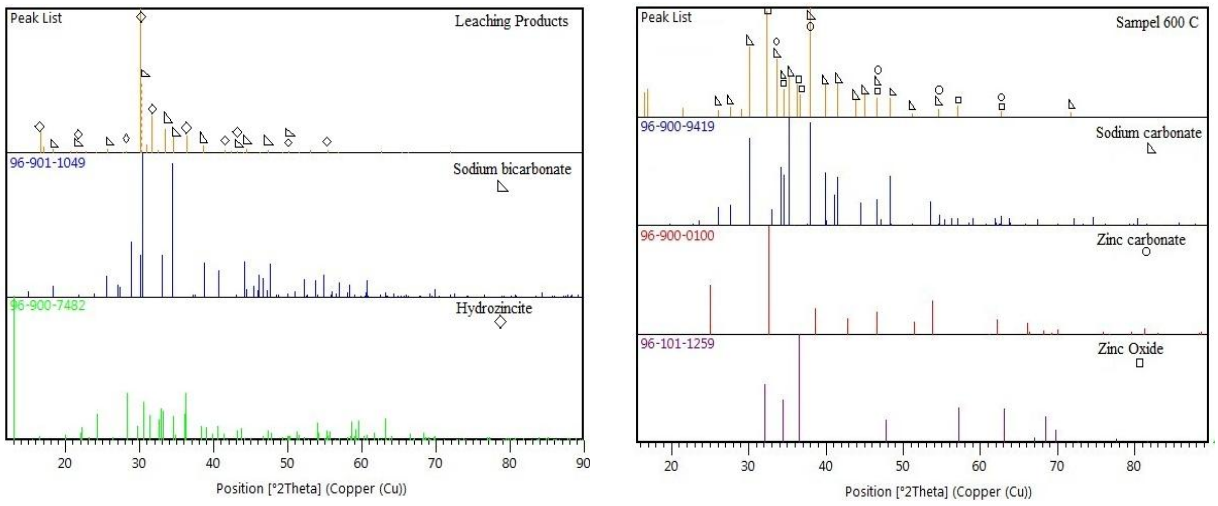


Figure 5. Difractogram XRD leaching product (left) and calcinated product (right)

4. Conclusion

Leaching zinc dust from the electric arc furnace waste by using oxalic acid at 90 °C for 150 minutes resulted to the hydrozincite and sodium bicarbonate formation. This perhaps caused by decarboxylation phenomenon during leaching process which encouraged the transformation of oxalate into carbonate. The existence of CO_2 in atmosphere caused the calcination process at 600 °C that resulted zinc carbonate, zinc oxide and sodium carbonate.

References

[1] Roohani N, Hurrell R, Kelishadi R, and Schulin R 2013 *J Res Med Sci*. **18(2)** 144-157
[2] Hsu C 2017 *Nanomed Nanotech*. **2(1)** 000114
[3] Shawabkeh RA 2010, *Hydrometallurgy* **104** 61-65
[4] Rahmani S A, Meidianto A, Amal M I, Wismogroho A S, Widayatno W B 2018 *J Phys Conf Ser* **985 (1)** 012033
[5] Pathak A, Roy A, and Manna M 2016 *Hydrometallurgy* **163** 161-166
[6] Marta L and Horouhz M 2003 *Leonardo Journal of Science* 72-78
[7] Vagvolgyi V, Hales M, Martens W, Kristof J, Horvath E, and Frost R L 2008 *J. Therm. Anal. Calorim.* **92** 911-916