Journal of Metallic Material Research
https://ojs.bilpublishing.com/index.php/jmmr

ARTICLE

Producing High Purity Nickel Metal Powder from Nickel Wastes through Acidic Leaching by Sulfuric Acid

Mohammad Bagher Oshrieh **Khanali Nekouee*** **Hasan Abbaszadeh**

Faculty of Materials and Manufacturing Technologies, Malek Ashtar University of Technology, Iran

ARTICLE INFO

Article history
Received: 8 July 2022
Revised: 27 July 2022
Accepted: 29 July 2022
Published Online: 22 August 2022

Keywords:
Nickel
Metal powder
Recovery
Acidic leaching
Sulfuric acid

ABSTRACT

Nickel has found increasing application in electronic, automobile manufacturing, plating, and metal industries and so on. Producing high quality metal powders to satisfy increasing demand for advanced materials is of very high importance. There are a few numbers of standard powder production techniques. An acidic leaching has been applied in present research. Sulfuric acid has been used to leach nickel wastes of plating industry. To produce nickel oxide powder furnaces with no protecting atmosphere and to produce pure nickel powder, tube furnace with hydrogen atmosphere has been applied. Variables performed in the research are time, density of sulfuric acid, and amount of hydrogen peroxide. To analyze powders produced, EDS element analysis and to determine size of powder particles, SEM has been applied. It was shown by the results that the highest amount of nickel dissolution in sulfuric acid (98%) has taken place during one hour and there is a direct relationship between hydrogen peroxide amount and nickel dissolution in sulfuric acid.

1. Introduction

Nickel as 24th metal element is one of the most frequent element available in earth's crust. Wide range usage made of nickel in different industries has led it to be considered as a strategic metal. In fact, this variety of application is resulted from various physical and chemical properties of this metal [1,2]. This is one of the important metals in industry and a basic one for modern technologies and industry. It is also important in energy, telecommunication, shape memory alloy, and electronics industries that are necessary for economy, medical industry and or substructures [3,4]. Some studies have been performed in this respect which will be referred to.

Mansi et al. [5] with leaching of catalysts in a solution of 50% sulfuric acid, solid to liquid ratio 1:12 and in particle size less than 500 microns for 5 hours, at 100 °C with a stirring of 800 rpm, about 99% Recover of nickel. Oza et al. [6] used ultrasound to recover nickel from the catalysts along with the leaching method. In this work, they were able to extract about 95% of nickel from the text of nickel-alumina catalysts by nitric acid at a concentration

---

*Corresponding Author:
Khanali Nekouee,
Faculty of Materials and Manufacturing Technologies, Malek Ashtar University of Technology, Iran;
Email: khnekuee@gmail.com

DOI: https://doi.org/10.30564/jmmr.v5i2.4877

of 40% at 90 °C and a solid to liquid ratio of 1:10 (g/mL) for 50 minutes. Using ultrasound, the recovery time was reduced from 7 hours to 50 minutes.

Randhawa et al. [7] conducted a study in which the leaching kinetics of nickel-cadmium batteries by sulfuric acid were investigated. Their results showed that the concentration of sulfuric acid up to 10% had a significant effect on the recovery of cadmium and nickel. However, nickel recovery was much lower than cadmium. Addition of hydrogen peroxide improved the nickel leaching kinetics, and nickel recovery increased to 96% at 5 hours of leaching, and nickel leaching also increased with increasing temperature. Driss et al. [8] used sulfuric acid and hydrochloric acid to recover nickel from palm oil catalysts. In this work, they investigated the effect of acid concentration parameters, solid to liquid ratio, temperature and leach time and found that sulfuric acid is more effective for nickel recovery. If the concentration of sulfuric acid is 67%, the leaching time is 140 minutes, the solid to liquid ratio is 1:14, about 60% of nickel will be recovered at 60 °C. If the temperature is above 80 degrees Celsius, this value will decrease.

In another study by Abrar et al. [9], the recovery of nickel from a catalyst was investigated using the pressure dissolution method in nitric acid. The catalyst for this study is NiO/Al_2O_3, which is used for direct reduction and production of sponge iron. Results showed that at 140 °C maximum dissolution can be obtained but over this optimum temperature a reverse effect is noticed. Another important factor is the Rpm change effect, as a change from 600 to 300 caused a decrease of 10% of dissolution efficiency. Finally, it is shown that efficiency was improved when dissolution time increased.

Gharabaghi et al. [10] conducted a study entitled leaching kinetics of nickel extraction from hazardous scrap by sulfuric acid and optimization of dissolution conditions. It was found that nickel extraction increases with increasing sulfuric acid concentration, temperature and leaching time. Reducing particle size and solid to liquid ratio increased the dissolution of nickel. The extraction speed increased with increasing stirring speed and reached the highest level at 500 rpm. The maximum leaching rate was 97% at 25 °C after 30 minutes.

Liu and et al. [11] conducted research to produce high-purity basic nickel carbonate through chemical precipitation. They used electric double layer model to illustrate the adsorption mechanism of ions of sodium and chlorine and other impurities on solid surface. Experimental results indicate that the new process is very effective to remove Na^+ and Cl^- to a level of both less than 0.01 wt%. Li and et al. [12] also conducted research for the hydrothermal synthesis of nickel or cobalt-based carbonate hydroxides used in the production of supercapacitor electrodes. The results showed that the nickel-cobalt carbonate hydroxide electrode shows excellent cycling stability and is suitable for the production of supercapacitors.

In a study, Wu and et al. [13] produced nickel nanoparticles used in supercapacitors using hydrazine hydrate reduction. After conducting the experiment, nickel nanoparticles with a particle size of about 12 nm were obtained and they concluded that due to the good conductivity of nickel particles, it is a suitable option for producing supercapacitors. Huang and et al. [14] investigated the method by which nickel nanoparticles were produced. In this method, the production cost was reduced and nickel nanoparticles in the range of 180 nm to 260 nm were produced. They also controlled the size range of nickel particles by adjusting the reaction molar ratio and concentration. In one study, Li and et al. [15] produced ultrafine nickel powder and crystalline film using chemically controlled reduction. Their results showed that pure black nickel powder can be obtained when the pH > 10.0 and the temperature is in the range of 85 °C ~ 958 °C. RF plasma synthesis of nickel nanopowders via hydrogen hydroxide/nickel carbonate reduction was performed by Bai et al. [16]. They were able to produce nickel nanoparticles in the size range of 60 nm to 100 nm, which are used in the production of electrode materials.

Ultra fine nickel powder was produced by polyol method and its oxidation product by Zhou and et al. [17]. The results of this research showed that the high content of water in $Ni(OH)_2$ increases the reaction rate in the solution and decreases the average particle size of nickel powders. Paserin and et al. [18] investigated different methods for the mass production of nickel-based nanomaterials by the carbonyl process in a review article. In the research conducted by Alena and et al. [19], the structure and properties of nickel nanoparticles prepared by selective leaching were investigated. They came to the conclusion that with this method it is possible to achieve a particle size of less than 5 nm and by increasing the temperature from 20 °C to 80 °C, the particle size will be in the range of 10 nm to 15 nm. Archana and et al. [20] conducted a research titled recovery of nickel powder from copper electrolyte by electrolysis. They were able to recover 87% of the nickel, which increased to 99% under certain conditions. The purity of the produced powder was 89.99%.

Since nickel powder is one of the basic and important metals in the industry, the production of this powder with high purity will be of special importance. After a lot of research, we came to the conclusion that one of the important sources for the production of nickel powder is the use of nickel wastes, especially nickel wastes produced by electroplating. As shown in the research, one of the best methods for producing metal powder is the acid leaching method. For this reason, we decided to produce high-purity nickel powder, which is used in various industries, from nickel waste by acid leaching method, which will be fully explained in the following process.

2. Materials and Method

Main materials used in the research have been prepared from plating wastes. EDS analysis of the material is shown in Figure 1. For acidic leaching of the material also, sulfuric acid Merck (98 wt%) has been used.

Reaction times have been selected 5, 10, 20, 30, 40, 50, and 60 minutes; while, sulfuric acid concentrations have been 20%, 40%, 60%, 80%, and 98%. One of the methods to increase dissolution speed of nickel wastes in sulfuric acid is adding H_2O_2 which effect of it has been studied through adding 50 cc, 100 cc, 150 cc, 200 cc, and 250 cc of the material. To stir and heat the solution in different stages of producing nickel powder, IKA/RCT basic hot plate stirrer (made in Germany) has been used. Producing nickel carbonate from nickel sulfate would be performed by sodium carbonate. Industrial type of sodium carbonate has been used in present research. To better perform the reaction between nickel sulfate and sodium carbonate, the temperature of the hot plate was set to 60 degrees Celsius. After finishing the reaction (Chemical reaction 2) and producing nickel carbonate, filter paper (Whatman) was used to separate the solution from it. There is a possibility that there is sodium in the nickel carbonate, which requires a lot of washing to remove it, because sodium is soluble in water. To do so, distilled water has been used to produce nickel oxide and finally pure nickel, two different furnaces with controlling atmosphere have been required. In the research and to produce nickel oxide from nickel carbonate; ordinary furnace (AZAR-FURNACE M35L) with no controlling atmosphere has been used for one hour at 700 °C (Chemical reaction 3). Finally, a hydrogen Fulled tube furnace (AZAR-FURNACE TF5/25) was used to convert nickel oxide into high purity nickel powder (Chemical reaction 4). The reduction process took place in two different cycles. The first cycle was performed at 700 °C for 2 hours and the second cycle at 600 °C for 4 hours. In general, production stages of nickel powder from nickel wastes include the four following stages:

Producing nickel sulfate from nickel wastes:

$$(H_2SO_4 + Ni = NiSO_4 + H_2) \tag{1}$$

Producing nickel carbonate from nickel sulfate:

$$(NiSO_4 + Na_2CO_3 = NiCO_3 + Na_2SO_4) \tag{2}$$

Producing nickel oxide from nickel carbonate:

$$(NiCO_3 = NiO + CO_2) \tag{3}$$

Producing high purity nickel powders from nickel oxide:

$$(NiO + H_2 = Ni + H_2O) \tag{4}$$

General production procedure of nickel powder is shown in Figure 2.

To determine EDS point analysis and to image produced powders, Vega/TScan SEM (made in China) has been used. Minimum diameter of the device where the sample is contacted is 2 nanometers.

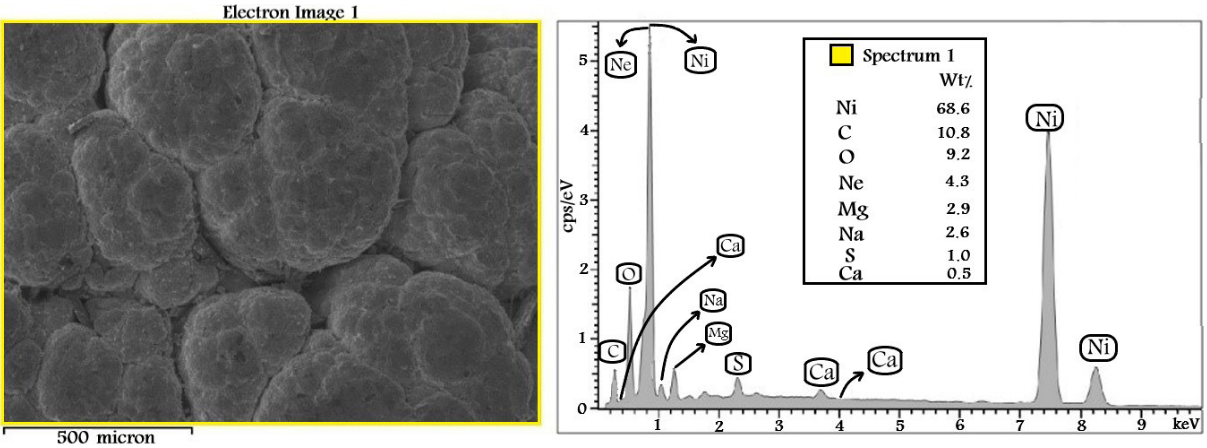

Figure 1. EDS analysis of wastes used in the research.

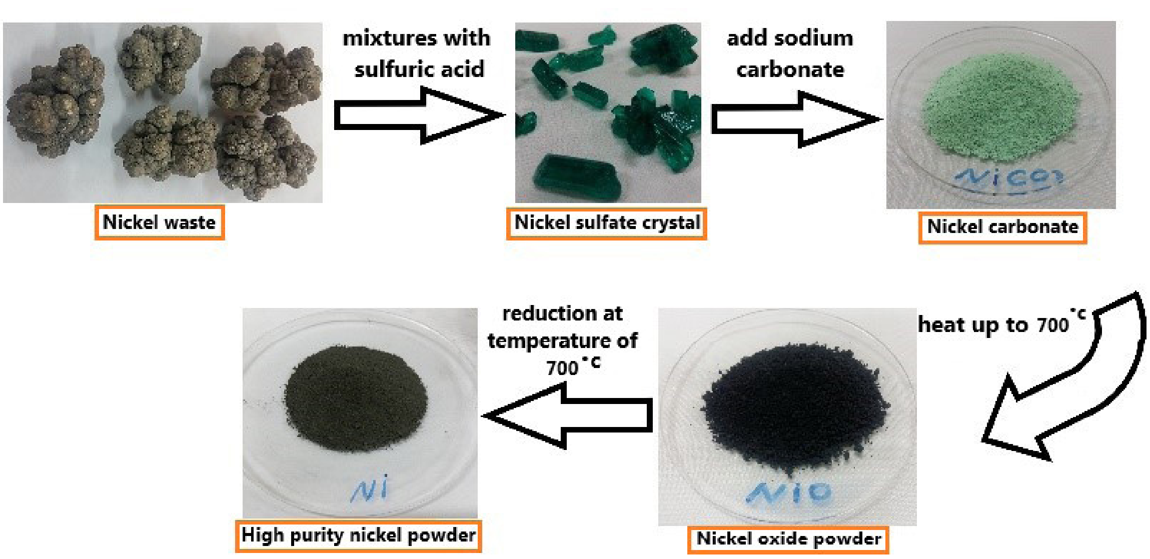

Figure 2. Flowchart of the research to produce high purity nickel powder.

3. Results and Discussion

After all four stages of producing nickel powder through acidic leaching, amounts of the product obtained in tests with final product have been computed and compared in reactions. In Table 1, the obtained have been compared with reaction stoichiometry reactions. At first stage, final product is nickel sulfate.

According to stoichiometry of the reaction, about 59.84 gr nickel would be dissolved in 100 gr sulfuric acid. The test and solving nickel waste in sulfuric acid has been performed for several times; and, nickel amount dissolved has been about 15 gr ~ 20 gr. End of the test has been considered to be at the point that no additional nickel would be dissolved in the solution and it would be considered as saturated. Final product of the second stage is nickel carbonate. According to the reaction stoichiometry, 68.48 gr sodium carbonate has to be consumed in each 100 gr of nickel sulfate so that the reaction would be complete; and, finally, there would be 76.7 gr nickel carbonate. This amount of sodium carbonate used for produced nickel sulfate in the research is very high; because, less nickel has been dissolved in sulfuric acid, compared to stoichiometry. Therefore, different amounts of sodium carbonate have been tested. Finally, it became clear that 30 gr sodium carbonate is appropriate for each 100 gr of nickel sulfate solution and the reaction would be complete. Third and fourth stages are well matched with stoichiometry. High difference in stages one and two can be due to type of nickel wastes used as well as purity of acid. Numbers obtained in stoichiometries reactions' will consider all conditions as ideal which their performance would be very hard and sometimes impossible in practical and lab conditions.

As referred to in the Research Materials and Method, one of the most important parameters in increasing dissolution of nickel wastes is sulfuric acid concentration, time and amount of hydrogen peroxide. The results obtained

Table 1. Comparing reaction products obtained in the research through stoichiometry.

Stage number	Final product	Final weight of the product in the laboratory	Final product weight according to stoichiometry	Difference (%)
1	$NiSO_4$	52.73	157.78	66.5
2	$NiCO_3$	33.6	76.7	56.2
3	NiO	61.36	62.92	2.46
4	Ni	77.17	78.57	1.27

from effect of these three parameters in dissolution of nickel wastes in sulfuric acid would be presented in this paper. Figure 3 shows effect of sulfuric acid concentration on amount of nickel wastes' dissolution. The test for all concentrations has been performed during one hour. As shown in the above figure, through increase of sulfuric acid concentration, dissolution amount of nickel wastes would be increased. At 20% and 40% concentrations, dissolution amount is very low and there is no main difference between the two concentrations; however, at 60% to 98% concentrations, a steep slope would be created in dissolution of nickel wastes. This shows that sulfuric acid with concentrations more than 60% can end to acceptable results.

In continuation of studying parameters, effect of reaction time duration has been dealt with. The test has been performed in sulfuric acid (98%), showing the highest efficiency in terms of dissolution in previous section. The results are provided in Figure 4. Figure 4 showed that, through increase of dissolution process time, more nickel wastes have been dissolved in sulfuric acid. This dissolution would be continued till saturation of the solution.

Another important parameter studied was the effect of amount of hydrogen peroxide on amount of dissolution (Figure 5). The test has been performed during one hour dissolution time in sulfuric acid (98%).

It became clear that adding hydrogen peroxide has a positive effect in increasing dissolution of nickel wastes in sulfuric acid. The reason for increased dissolution is increased oxidation and activation of surface of nickel wastes. As shown by the results, in previous test and during one hour 15.3 gr nickel wastes have been dissolved in sulfuric acid (98%). This amount has been increased to 16.91 gr after adding 50 cc hydrogen peroxide to the solution, through increasing of hydrogen peroxide to the solution, dissolution amount of nickel wastes also has been increased. To study purity of nickel powder produced in the research, EDS element analysis has been performed on powders (Figure 6).

Nickel powder produced is of 99.6% purity which is high; however, existence of 0.4% of sulfur in analyses of powders can be due to following reasons: firstly, in initial analysis of nickel wastes there is about 1% sulfur which has been remained till the final stage of powder production. Secondly, the reason justifying this amount of sulfur goes back to the first stage of nickel powder production and preparation of nickel sulfate through sulfuric acid. To study shape of nickel powder particles produced, images from SEM are shown in Figure 7.

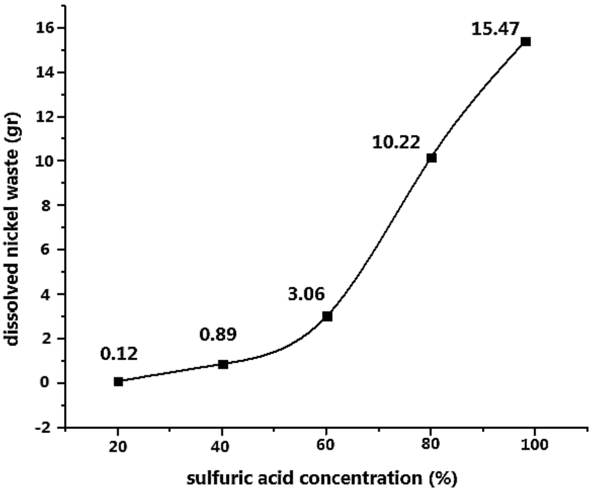

Figure 3. Effect of sulfuric acid concentration in dissolution of nickel wastes.

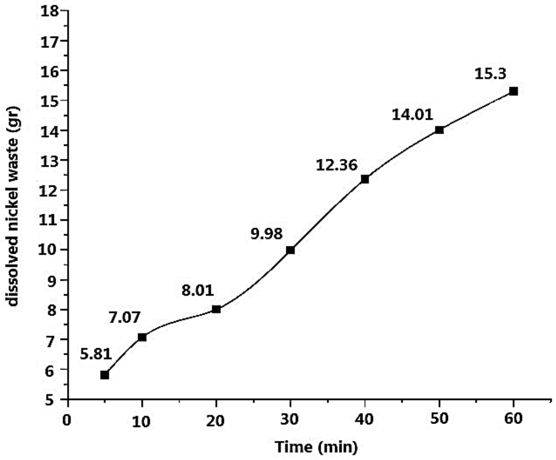

Figure 4. Effect of reaction time duration on dissolution of nickel wastes.

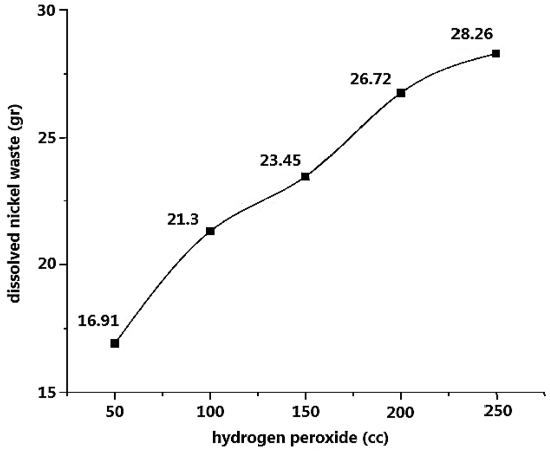

Figure 5. Effect of amount of hydrogen peroxide on dissolution of nickel wastes.

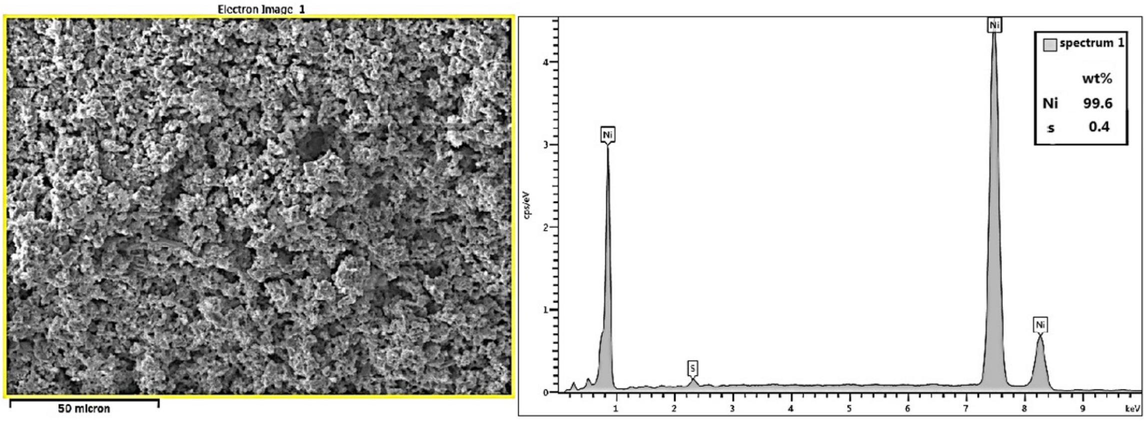

Figure 6. EDS element analysis performed on produced nickel powder.

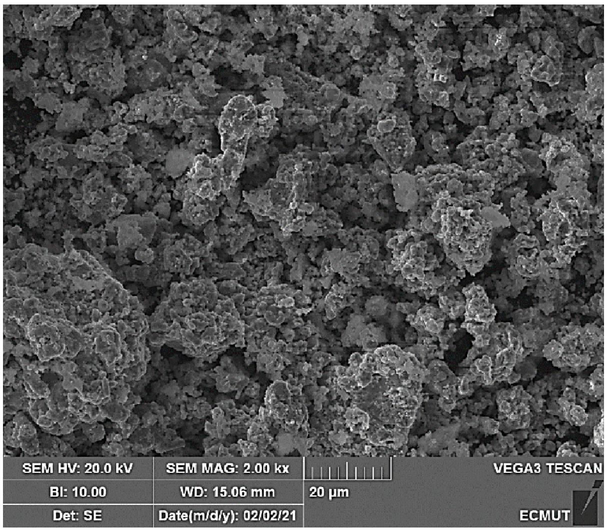

Figure 7. SEM images from nickel powder produced.

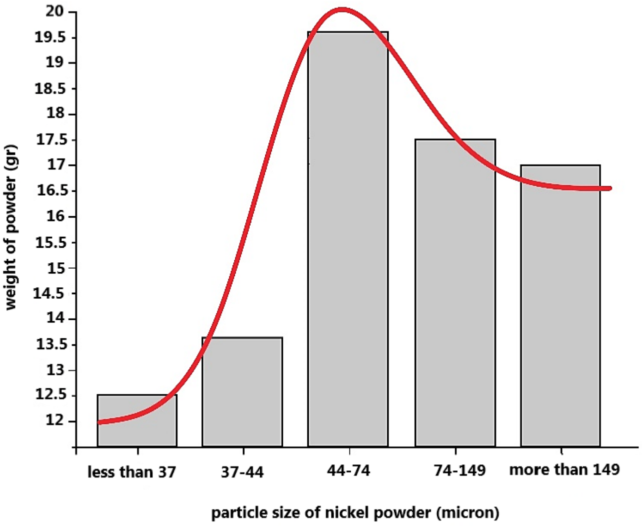

Figure 8. Size distribution of nickel powder produced in the research.

It can be suggested that powder particles produced had porous and irregular shape. To determine size distribution of produced powder particles, sifting is one of the common methods. To do so, 80 gr of nickel powder has been separated and passed through screen to determine size of particles. Size distribution of powder particles is shown in Figure 8.

The highest distribution is related to the range of 44 microns ~ 74 microns. Size of powder particles during third and fourth stages related to production of nickel oxide and pure nickel can be controlled. If at reduction stage of powders, temperature would be reduced and time would be increased; lower size of particles would be resulted. Therefore, the reduction temperature was 600 °C and the time was 4 hours. Elemental analysis (EDS) of nickel powder produced was performed after the completion of this cycle (Figure 9). As shown in the Figure 9, the nickel analysis shows a value of 100%.

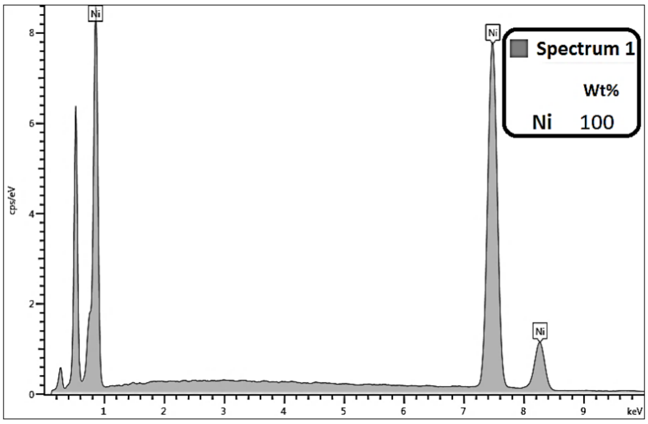

Figure 9. Elemental analysis (EDS) of nickel powder after a cycle of 600 °C and a time of 4 hours.

SEM images were also obtained from samples of powders produced in this cycle (Figure 10). Due to this shape and compared to the sample of powders produced from the previous cycle and Figure 7, the produced powders were much finer. The dispersion of the powder particle size produced in the second cycle was measured (Figure 11). According to this figure, 80.22% of the produced powders were less than 45 microns.

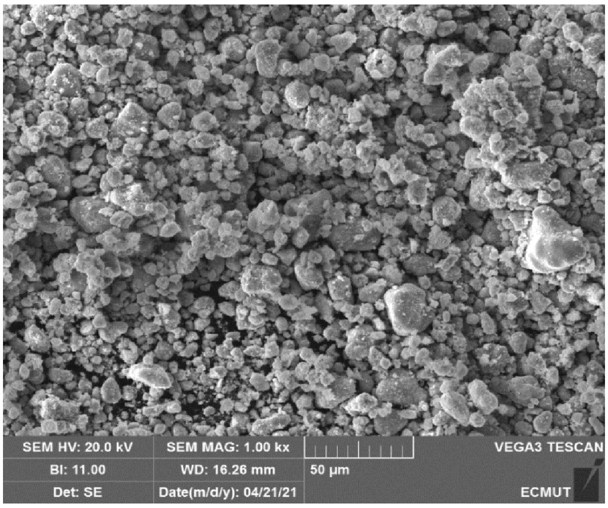

Figure 10. SEM image of nickel powder produced after the second reduction cycle.

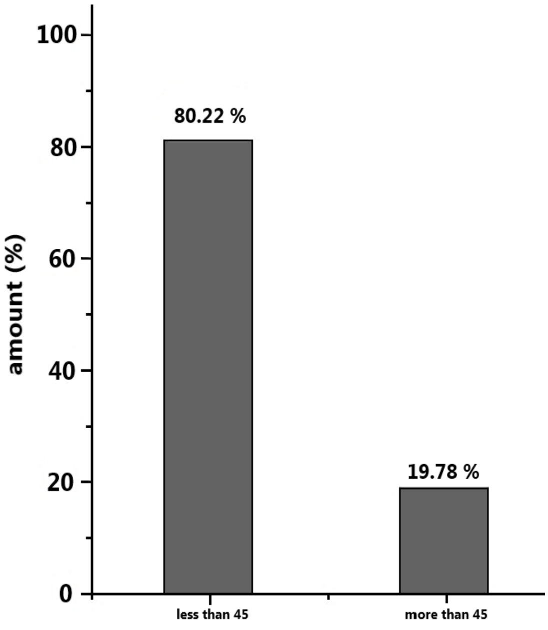

Figure 11. Nickel powder particle size dispersion after the second reduction cycle.

4. Conclusions

Acidic leaching to produce high purity nickel powder from nickel wastes through sulfuric acid has been performed in the research and the results are as follows:

1) The two parameters of sulfuric acid concentration and duration of process have had a direct effect on dissolution of nickel wastes in sulfuric acid. That is, upon increase of acid concentration from 20% to 98%, dissolution amount of nickel wastes has been increased from 0.12 gr to 15.47 gr. Also, upon increasing of time from five to 60 minutes, dissolution amount of nickel wastes has been increased from 0.81 gr to 15.3 gr.

2) Hydrogen peroxide (H_2O_2) has been considered as one of the main and important factors in increasing dissolution of nickel wastes in sulfuric acid. That is during one hour fixed, 15.3 gr ~ 15.47 gr of nickel wastes have been dissolved in the solution with no hydrogen peroxide; however, adding 50cc of hydrogen peroxide during the same time has led to increase of nickel wastes' dissolution to about 16.91 gr. This is about 10% higher than the time no hydrogen peroxide has been used. In industrial scale, it is considered as a considerable amount. In general and upon increase of hydrogen peroxide, dissolution amount of nickel wastes also has been increased due to surface of nickel wastes becoming activated.

3) Nickel powder produced through acidic leaching has been highly pure. Elemental analysis (EDS) of this powder in the first reduction cycle (temperature 700 °C and time 2 hours), showed 99.6%, which is considered as an acceptable purity. Also, particles of nickel powder have been porous and irregular. Size range of nickel powder has been computed and maximum value has been related to size of particles within the range of 44 microns ~ 74 microns.

4) After the second reduction cycle (temperature of 600 °C and time of 4 hours), elemental analysis of the produced nickel powder showed 100%. Also, after this cycle, the particle size of nickel powder became smaller than the first cycle. Most of the particle size dispersions were obtained in the range below 45 microns.

Conflict of Interest

There is no conflict of interest.

References

[1] Rhamdhani, M.A., Jak, E., Hayes, P.C., 2008. Basic nickel carbonate: Part I. Microstructure and phase changes during oxidation and reduction processes. Metallurgical and Materials Transactions B. 39(2), 218-233.
DOI: https://doi.org/10.1007/s11663-007-9124-4

[2] Rhamdhani, M.A., Jak, E., Hayes, P.C., 2008. Basic

nickel carbonate: Part II. Microstructure evolution during industrial nickel production from basic nickel carbonate. Metallurgical and Materials Transactions B. 39(2), 234-245.
DOI: https://doi.org/10.1007/s11663-008-9139-5

[3] Mackenzie, M., Virnig, M., Feather, A., 2006. The recovery of nickel from high-pressure acid leach solutions using mixed hydroxide product–LIX® 84-INS technology. Minerals Engineering. 19(1), 1220-1233.
DOI: https://doi.org/10.1016/j.mineng.2006.01.003

[4] Shaheen, W.M., 2002. Thermal behaviour of pure and binary basic nickel carbonate and ammonium molybdate systems. Materials Letters. 52(4-5), 272-282.
DOI: https://doi.org/10.1016/S0167-577X(01)00406-2

[5] Al-Mansi, N.M., Abdel Monem, N.M., 2002. Recovery of nickel oxide from spent catalyst. Waste Management. 22(1), 85-90.
DOI: https://doi.org/10.1016/S0956-053X(01)00024-1

[6] Oza, R., Nikhil, S., Sanjay, P., 2011. Recovery of nickel from spent catalysts using ultrasonication-assisted leaching. Journal of Chemical Technology & Biotechnology. 86(10), 1276-1281.
DOI: https://doi.org/10.1002/jctb.2649

[7] Randhawa, N.S., Kalpataru, G., Manoj, K., 2016. Leaching kinetics of spent nickel–cadmium battery in sulphuric acid. Hydrometallurgy. 165(1), 191-198.
DOI: https://doi.org/10.1016/j.hydromet.2015.09.011

[8] Idris, J., Musa, M., Yin, C.Y., et al., 2010. Recovery of nickel from spent catalyst from palm oil hydrogenation process using acidic solutions. Journal of Industrial and Engineering Chemistry. 16(2), 251-255.
DOI: https://doi.org/10.1016/j.jiec.2010.01.044

[9] Abrar, B., Mohammad, H., Ali, P., 2016. Recovery of nickel from reformer catalysts of direct reduction, using the pressurized dissolving method in nitric acid. Engineering, Technology & Applied Science Research. 6(5), 1158-1161.
DOI: https://doi.org/10.48084/etasr.731

[10] Gharabaghi, M., Mehdi, I., Amir, R.A., 2013. Leaching kinetics of nickel extraction from hazardous waste by sulphuric acid and optimization dissolution conditions. Chemical Engineering Research and Design. 91(2), 325-331.
DOI: https://doi.org/10.1016/j.cherd.2012.11.016

[11] Liu, F., Li, N., Zhang, Z., et al., 2008. An improved purification method for preparation of basic nickel carbonate of high purity via chemical precipitation. Journal of Wuhan University of Technology-Mater. 23(3), 331-333.
DOI: https://doi.org/10.1007/s11595-007-3331-3

[12] Li, Y., Li, P., Xin, Z., 2017. Hydrothermal synthesis of hierarchical nickel-or cobalt-based carbonate hydroxides for supercapacitor electrodes. International Journal of Electrochemical Science. 12, 4016-4024.
DOI: https://doi.org/10.20964/2017.05.50

[13] Wu, X., Xing, W., Zhang, L., 2012. Nickel nanoparticles prepared by hydrazine hydrate reduction and their application in supercapacitor. Powder Technology. 224, 162-167.
DOI: https://doi.org/10.1016/j.powtec.2012.02.048

[14] Huang, G.Y., Xu, S.M., Gang, X.U., et al., 2009. Preparation of fine nickel powders via reduction of nickel hydrazine complex precursors. Transactions of Nonferrous Metals Society of China. 19(2), 389-393.
DOI: https://doi.org/10.1016/S1003-6326(08)60283-6

[15] Li, Y.D., Li, C.W., Wang, H.R., et al., 1999. Preparation of nickel ultrafine powder and crystalline film by chemical control reduction. Materials Chemistry and Physics. 59(1), 88-90.
DOI: https://doi.org/S0254-0584(99)00015-2

[16] Bai, L., Fan, J., Hu, P., et al., 2009. RF plasma synthesis of nickel nanopowders via hydrogen reduction of nickel hydroxide/carbonate. Journal of Alloys and Compounds. 481(1-2), 563-567.
DOI: https://doi.org/10.1016/j.jallcom.2009.03.054

[17] Ying, Z., Shengming, J., Guanzhou, Q., et al., 2005. Preparation of ultrafine nickel powder by polyol method and its oxidation product. Materials Science and Engineering: B. 122(3), 222-225.
DOI: https://doi.org/10.1016/j.mseb.2005.06.006

[18] Paserin, V., Baksa, S., Zaitsev, A., et al., 2008. Potential for mass production of nickel-based nanomaterials by carbonyl process. Journal of Nanoscience and Nanotechnology. 8(8), 4049-4055.
DOI: https://doi.org/10.1166/jnn.2008.AN44

[19] Michalcová, A., Svobodová, P., Nováková, R., et al., 2014. Structure and magnetic properties of nickel nanoparticles prepared by selective leaching. Materials Letters. 137, 221-224.
DOI: https://doi.org/10.1016/j.matlet.2014.09.012

[20] Agrawal, A., Bagchi, D., Kumari, S., et al., 2007. Recovery of nickel powder from copper bleed electrolyte of an Indian copper smelter by electrolysis. Powder Technology. 177(3), 133-139.
DOI: https://doi.org/10.1016/j.powtec.2007.03.032