

# Selective Nickel Recovery From Spent Catalyst\*

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## ABSTRACT

Nickel recovery from spent nickel catalyst that involved two consecutive steps i) extracting the metals from solid spent catalyst using acid leaching and ii) selective precipitation of nickel from other metals by an addition of sodium hydroxide was studied. In leaching process, it was found that sulphuric acid as a leaching agent resulted significantly higher nickel concentration compared to nitric and hydrochloride acids, and the amount of nickel extracted increases with increasing in acid concentration used. It was also found that the concentration of nickel in spent catalyst is about 8.15 % (dry weight). In the selective precipitation, sodium hydroxide was used to adjust the pH of the solution. The results show that nickel starts to precipitate at higher pH probably as  $(\text{NiOH})_2$  or  $3\text{Ni}(\text{SO}_4) \cdot 4\text{Ni}(\text{OH})_2$  than Al, Zn, and Fe that is, at the optimum pH, more than 80 % of nickel recovery was obtained.

### Keywords

Selective, Recovery, Spent Catalyst, Leaching, Precipitation

## 1. INTRODUCTION

Nickel containing catalysts or nickel catalysts are well known in process industries. They have been used in many reactions such as hydrogenation reaction, alkylation reactions, hydroalkylation reactions and cracking reactions. The initial composition of nickel catalyst depends on reaction involved for example catalyst for hydroalkylation reaction containing other metals such as tungsten besides nickel itself. Initially, these catalysts perform at high activity but by time they become less active and reach at a point where they are not no more effective to be used in a commercial process.

Considerable research has been carried out on developing the process for regenerating the spent catalyst since nickel is expensive to be discarded and more over, the safe disposal of waste nickel requires consideration of environmental hazard. However, the regenerating process of a nickel catalyst from spent catalyst is tedious, time consuming, and careful attention on detailed procedures. Therefore there is a continuous afford needed to developing methods of recovering nickel from spent catalyst that provides inexpensive recovery of nickel in a usable form. Several methods have been described in literature regarding recovering nickel not only from spent catalyst but also from other solid and liquid wastes such as using electrowinning, fluidised bed and burning process [1,2]

This paper will present results of the selective recovery nickel from spent catalyst using leaching and precipitation process.

## 2. MATERIALS AND METHODS

### 2.1. Materials

All chemical used in this work are analytical grade from various suppliers. Sample spent nickel catalyst was obtained from Palm Oleo Sdn. Bhd., Rawang, Selangor and Pan Century Edible Oil Sdn. Bhd., Pasir Gudang, Johor. Nickel Catalyst was also kindly provided by Pan Century Edible Oil Sdn. Bhd., Pasir Gudang, Johor.

### 2.2. Methods

#### a) Water content determination

Water content in the sample was measured by heating 1 gm sample in oven at 100 °C until a constant weight was achieved. At 110 °C water expected to be evaporated. The difference in weight of sample before and after heating will give the weight of water in the sample.

#### b) Organic substances determination

To determine the organic substances in the sample, the previous sample from water content determination sample was then placed in the furnace at 450 °C overnight. At this temperature, all organic matters

\* Paper presented at World Engineering Congress and Exhibition 1999 (WEC'99), Kuala Lumpur, July 19-22, 1999.

expected to be burned. Again, the difference in weight of sample before and after burning will give the weight of organic matter in the sample.

### c) Elemental composition determination

The elemental composition of the sample was carried out using energy dispersive x-ray analysis (EDAX) model Philips PV 9900. The sample was grinding to form a fine powder and subsequently compressed into pellet form with 13 mm in diameter and 1.5 mm of thickness. The pellet was then treated with gold sputtering to ensure a high conductivity of the samples. The composition of each element would be displayed as weight percentage.

### d) Leaching experiment

One gram of burning catalyst (after burning at 450 °C) was placed in 100 ml beaker, and then 50 ml of acid at specified concentration was added with vigorous stirring. The solution was then filtered using filter paper in order to separate undissolved solid. The filtrate was transferred to 100 ml volumetric flask and by using double distilled water, the solution was then make up to 100 ml. Similar procedure was carried out for other acids and concentrations. The solution was used to determine the metal concentration using Atomic Absorption Spectrophotometer (A.A.S) before it was used for precipitation experiment.

### e) Precipitation experiment

50 ml of solution obtained from leaching process was used in precipitation studies. The pH of the solution was adjusted to required pH using concentrated NaOH solution. Any significant change in solution volume due to pH adjustment has to be avoided. During the NaOH addition, the solution needs to stir vigorously to avoid any pH gradient and solution temperature was maintained at 50-55 °C using water bath. The solution was then left overnight before filtered of the precipitate. The filtrate was then used to determine metal concentration using Atomic Absorption Spectroscopy (A.A.S). The difference between initial and filtrate metal concentrations will give the amount of metal precipitation.

All experiments were carried out at ambient temperature and pressure unless specified in the procedures.

## 3. RESULTS AND DISCUSSION

### 3.1. Characterisation of spent catalyst

Table 1 shown the composition of the spent catalyst obtained from EDAX analysis results (Figure1). It was found that the nickel concentration is about 8.15 % (dry weight) which is accounted to be 66.7 % of total metal

in fresh nickel catalyst sample was also analysed and it found to have about 19.5 %. This percentage falls within in the range of specified composition of nickel in the fresh nickel catalyst given by the supplier [3]. This decrease in nickel concentration comparing to fresh catalyst might due to increase in other components in spent catalyst such as organic substances and metals or due to loss of nickel during the process. Table 2 shown the composition of the fresh nickel catalyst.

Table 1: The Elemental Composition of Spent Nickel Catalyst.

Components	% (Dry Weight)
Organic substances	63.70
Silicon	21.60
Nickel	8.15
Aluminium	1.54
Ferrum	1.40
Zinc	0.94
Silver	-0.10
Copper	0.08
Others	2.47

Table 2: The Composition of Fresh Nickel Catalyst [3].

Components	% (Dry Weight)
Nickel	5-20 %
Nickel Oxide	10-20 %
Diatomaceous earth	10-20 %
Triglyceride	50-60 %
Metal Oxides	<5 %

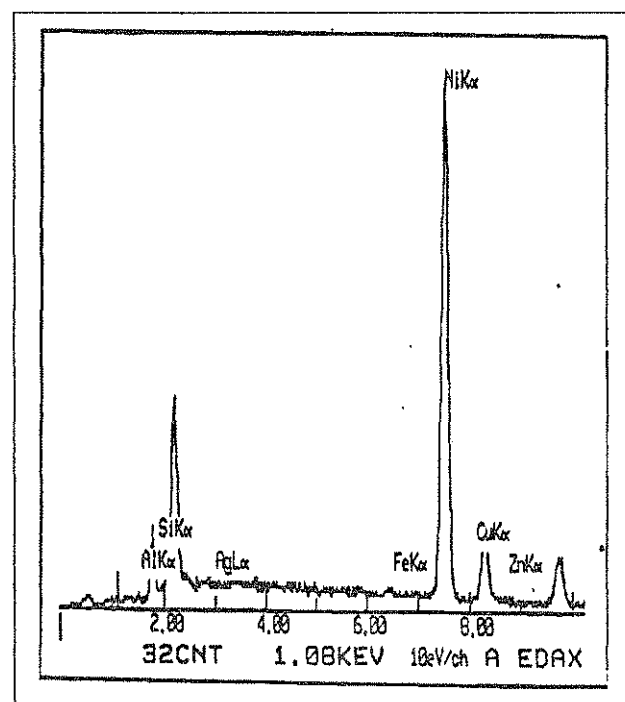


Figure 1: EDAX Elemental Spectrum of the Sample

### 3.2. Effect of acid types and concentration on leaching process

The effect of  $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$  and  $\text{HCl}$  at various concentrations on leaching process was studied. Figure 2 shown the extent of nickel extracted increases with increase in acid concentrations even though the highest extraction was observed for  $\text{H}_2\text{SO}_4$ . This might due to relative strength of the acids and their metal salts solubility. It was observed that the colour of leached solutions is light green for  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$  but for  $\text{HCl}$  is yellowish green. This slightly difference in colour of the leached solutions might be due to different types of salt formed for different types of acid used.

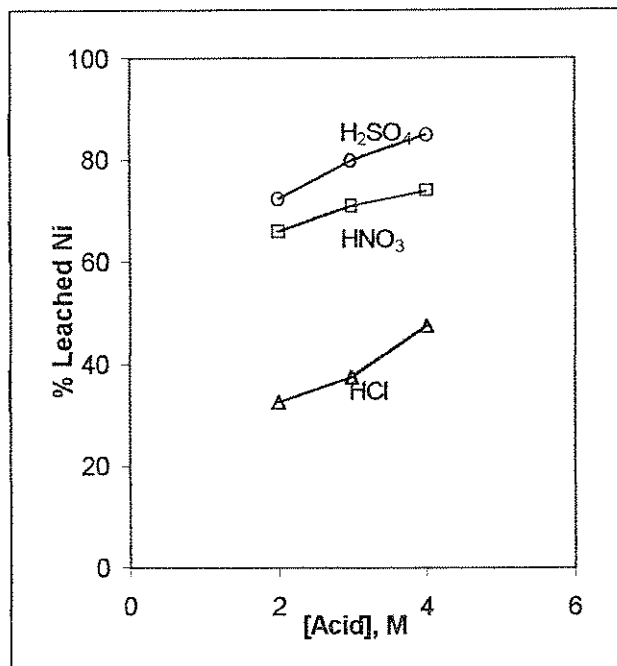


Figure 2: Effect of Acid Types and Concentrations on Leaching Process.

### 3.3. Effect of pH on metal's precipitation

Leaching solution from sulphuric acid leaching experiment was used in precipitation studies. By adding the sodium hydroxide to leaching solution, one would expect the formation of metal hydroxides that have low solubility than metal sulphates. Thus, the formation of precipitate was observed. If there is no interaction between one metal to another, the formation of sample metal hydroxide precipitates should follow the following order:  $\text{Fe}(\text{OH})_3 > \text{Al}(\text{OH})_3 > \text{Cu}(\text{OH})_2 > \text{Ni}(\text{OH})_2 > \text{Fe}(\text{OH})_2$  according to an order of increasing in  $K_{sp}$ :  $2.5 \times 10^{-39} < 4.6 \times 10^{-33} < 2.2 \times 10^{-20} < 3.3 \times 10^{-17} < 6.2 \times 10^{-16} < 3.3 \times 10^{-14}$  [4]. Figure 3 shown the effect of pH on the extent of metal ion's precipitation that indicates the precipitation process occurred according to the following order:

simply simple metal hydroxides as postulate earlier. Petkova et al. [5] showed that Zn, Fe and Ni can also precipitate in the form of  $\text{FeSO}_4 \cdot 2\text{Fe}(\text{OH})_2$ ,  $\text{ZnSO}_4 \cdot \text{Zn}(\text{OH})_2$ , and  $3\text{NiSO}_4 \cdot 4\text{Ni}(\text{OH})_2$  respectively. In order to get better understanding in term of which form of precipitates formed, further studies will be require. However, the results shown that one can recover more 80 % of nickel at pH higher than 9.92.

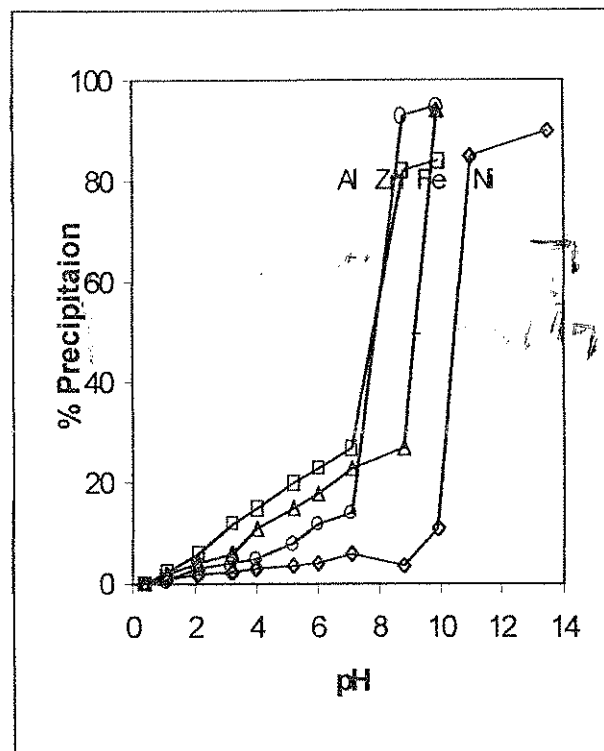


Figure 3: Effect of pH on Metal Ions Precipitation

## 4. CONCLUSIONS

Spent nickel catalyst was successfully characterised. Nickel content in spent catalyst is about 8.15 % (dry weight). It was found that  $\text{H}_2\text{SO}_4$  is the best leaching agent. Nickel recovery from leaching solution is possible since the difference in solubility products of metal hydroxides exist in the solution. The selective recovery of nickel can be achieved at pH than 9.92.

## 5. ACKNOWLEDGEMENTS

The authors would like to acknowledge the financial support from Universiti Teknologi Malaysia that make this work possible. We would like to thank Mr. Tayalan for his excellent technical assistance.

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