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Adsorption of Heavy Metal from Recovered base Oil using Zeolite

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Abstract: Recovery of used lubricating oil by extraction produced organic sludge and recovered base oil, but this oil has metallic content such as magnesium and zinc. In this study, purification of recovered base oil by using adsorption process to remove heavy metals was performed. Zeolite was used as an adsorbent. The parameters studied were contact time, amount of zeolite, temperature and their interactions. The results showed that zinc removal was higher than that of the magnesium. The optimum magnesium and zinc removal obtained were 50 and 62%, respectively. The most influential parameter affecting the magnesium and zinc removal was the time of adsorption. Further investigations on the optimum conditions will be performed.

Key words: Adsorption, factorial design, heavy metal, recovered base oil, zeolite

INTRODUCTION

Lubricating oils must be replaced on a regular basis in all operating equipment due to contamination from dirt, water, salts, metals, incomplete products of combustion, antifreeze, or other materials. Additives to lubricating oils may also break down under use, adding contamination.

The large number of contaminants potentially contained in used lubricating oil complicates the selection of appropriate treatment methods. Among the treatment methods proposed during recent years, solvent extraction process has received considerable attention (Reis and Jernimo, 1988; Chementator, 1996; Sherman, 1993; Elbashir, 1998; Saunders, 1996; Reis, 1991). This process replaces successfully the classic acid-clay treatment whereby a useful organic sludge is produced instead of the toxic acidic sludge (Elbashir *et al*., 1997).

Single or composite solvent can be used in extraction process. Solvent extraction process is capable of removing about 10-14% of the used oil as contaminants, which corresponds roughly to the amount of additives and impurities normally found in used oil (Sherman, 1993). Nevertheless, the base oil recovered with this composite solvent still contains metals (Jesusa *et al*., 2005).

Metallic compounds are other important used oil components that should be removed to obtain base oil suitable for the formulation of new lubricants. Removal of heavy metals such as magnesium and zinc from recovered base oil was the main focus of this study, particularly on the effect and interaction of parameters.

The experimental study was carried out using a 2^3 factorial design in order to examine the main factors affecting the adsorption and their interactions.

MATERIALS AND METHODS

Materials: The materials used were lubricating oil (collected from garages, mixed in a single container), 2-propanol 99.7% (R and M Chemicals), methyl ethyl ketone 99.8% (R and M Chemicals), potassium hydroxide 85% (R and M Chemicals) and zeolite with size less than 45 μm (Fluka, Sigma-Aldrich).

Equipment: The experiments were carried out with equipments such as; for extraction used beaker and electric stirrer (IKA Stirring motor with propeller 4 bladed R 1342), for separation extracted oil from sludge used method of gravity settling, for separation solvent from recovered base oil used rotary evaporator (Yamato RE 300) and recirculation chiller (Büchi), for adsorption process used beaker, water bath (Yamato BM 510) and electric stirrer and last for separation adsorbent and re-recovered base oil after adsorption used centrifuge.

Experimental procedure:

- Metal content in used lubricating oil were analyzed by ICP according to ASTMD-D5185

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- Used oil was then mixed with the composite solvent (2-propanol/ MEK 3 g g^{-1}, solvent/oil 7 g g^{-1}) and a flocculating agent, potassium hydroxide (KOH) 2 g kg^{-1}, using an electrical stirrer at 200 rpm for 30 min
- The mixture of steps 3 was allowed to settle. A layer of asphalt was generated. This layer was removed
- The solvent and base oil were then put in the rotary evaporator
- The heavy metals in recovered base oil were analyzed by ICP according to ASTMD-D5185
- The recovered base oil was then treated via adsorption process using zeolite at conditions such as experimental design
- Adsorption product from the previous step was separated by centrifuge (500 rpm, 10 min) to remove the zeolite and any suspended solids
- The re-recovered base oil was analyzed for metal content by ICP according to ASTMD-D5185
- The heavy metals analyzed were magnesium and zinc

The flow chart for experimental procedure is shown in Fig. 1.

Removal of heavy metals: The removal of heavy metal values or yield (Y) was calculated from the following equation:

$$Y = \frac{C_0 - C_e}{C_e} \times 100 \quad (1)$$

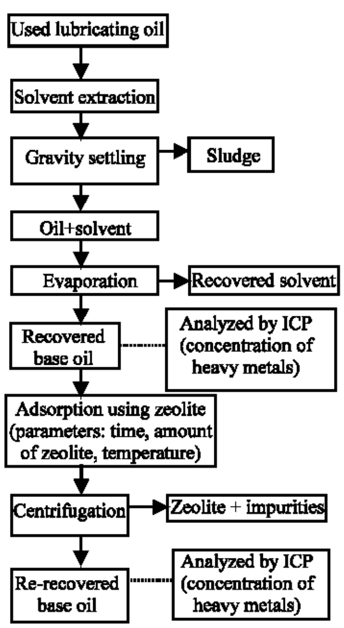

Fig. 1: Flow chart

where, Y is yield (%), C_e (ppm) is the equilibrium concentration of heavy metal at equilibrium and C_0 (ppm) is the initial heavy metal concentration.

Factorial design analysis: The parameters involved in the adsorption experiments were analyzed by full factorial design. The levels (-) and (+) in the adsorption of heavy metal for amount of zeolite (S), temperature (T) and time (t) are given in Table 1.

On the study of the effects of the three variables on the heavy metals removal, a two-level factorial design of experiments was adopted. The variables studied were amount of zeolite (1 and 10 g, respectively), temperature (30 and 70°C, respectively) and the adsorption time (5 and 60 min). The number of experiments required for quantifying all the effects is given by $a^k = 2^3 = 8$, where a is the number of levels and k is the number of factors. The two levels assigned to each variable were expressed in coded forms as + and - (Ragonese *et al.*, 2000; Massumi *et al.*, 2002).

Three variables with two levels for each variable experimental design matrix, 8 possible combinations with the adsorption of heavy metals are tabulated in Table 2. The design matrix for analysis of main and interaction effect are tabulated in Table 3.

Principal/main and interaction effect values were calculated from factorial design results. Both types of effects were calculated using the Eq. 2 (Box *et al.*, 2005):

$$\text{Effect} = \bar{R}_{+,i} - \bar{R}_{-,i} \quad (2)$$

Table 1: Factors and levels used in factorial design

Level	S Amount of zeolite (g)	T Temperature (°C)	t Time (min)
Upper (+)	10	30	5
Lower (-)	1	70	60

Table 2: Design of trial runs for heavy metals removal

Run	S	T	t
1	-	-	-
2	+	-	-
3	-	+	-
4	+	+	-
5	-	-	+
6	+	-	+
7	-	+	+
8	+	+	+

Table 3: Design matrix for main and interaction effect analysis

Run	Y_S	Y_T	Y_t	Y_{ST}	Y_{St}	Y_{Tt}	Y_{STt}
1	-	-	-	+	+	+	-
2	+	-	-	-	-	+	+
3	-	+	-	-	+	-	+
4	+	+	-	+	-	-	-
5	-	-	+	+	-	-	+
6	+	-	+	-	+	-	-
7	-	+	+	-	-	+	-
8	+	+	+	+	+	+	+

where, $\bar{R}_{+,i}$ and $\bar{R}_{-,i}$ are average values of Y for the high (+) and low (-) levels of each factor. For principal or main effects, the above averages simply refer to the results at the high (+) and low (-) levels of the factor whose effect is being calculated independent of the levels of the other factors. For binary interactions $\bar{R}_{+}$ is the average of results for both factors at their high and low levels whereas $\bar{R}_{-}$ is the average of the results for which one of the factors involved is at the high level and the other is at the low level. In general, high-order interactions were calculated using the above equation by applying signs obtained by multiplying those for the factors involved + for high and - for low level.

RESULTS AND DISCUSSION

The yield of heavy metals removal is shown in Fig. 2. The main effect analysis (Table 4), interaction effect analysis (Table 4), Pareto chart (Fig. 3-4) and contour plot (Fig. 5-10) were obtained by using statistical software (STATISTICA 6.0, USA).

Removal of heavy metals: Figure 2 shows that in all trial runs, the yield of zinc removal was higher than that of magnesium. The range of zinc removal was 45 to 62% and that for magnesium was 10 to 50%. These results meant that the adsorptive affinity of Mg was less than that of Zn to the zeolite surface.

Main effect and interaction effect: In Fig. 3 and 4, the biggest value of standardized effect, for magnesium and zinc was -3.0 and -6.6, respectively when varying the time of adsorption. Therefore, it can be said that the most influential parameter affecting the adsorption process was the adsorption time.

Table 4: Comparison of effect for Mg and Zn

Effect	Mg	Zn
Main effect		
Amount of zeolite (S)	5	2.25
Temperature (T)	5	-0.75
Time (t)	-15	-8.25
Interaction effect		
S-T	-10	-3.75
S-t	10	3.75
T-t	10	2.75

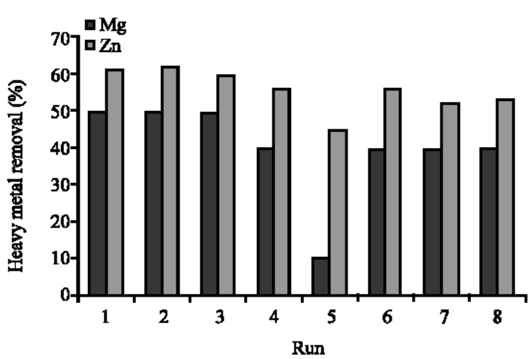

Fig. 2: Yield of magnesium and zinc removal

The best range for parameters in heavy metals removal: From Fig. 5, the optimum of magnesium removal was 50% for the range of parameters such as time of 5 min, temperature of 35 to 45°C and the amount of zeolite between 0-5 g. The optimum range of parameters in the magnesium removal obtained is shown in Table 5.

The optimum range of magnesium removal obtained, relatively, for time between 0 to 10 min, temperature of 30 to 50°C and for amount of zeolite between 0 to 5 g. The magnesium removal was 50%.

From Fig. 8, the optimum of zinc removal obtained was 62% for the range of parameters of time of 5 min, temperature between 30 to 50°C and the amount of zeolite in between 0 to 3 g. The optimum range obtained is presented in Table 6.

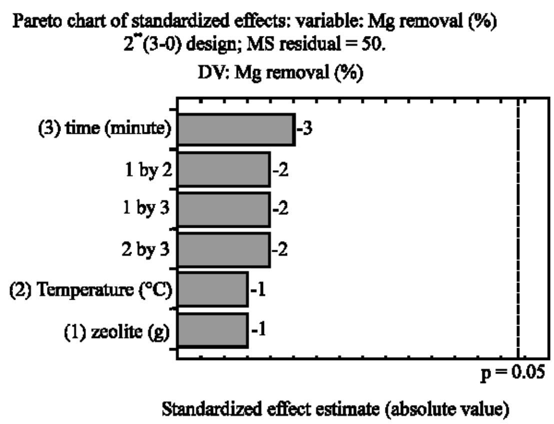

Fig. 3: Pareto chart for magnesium removal

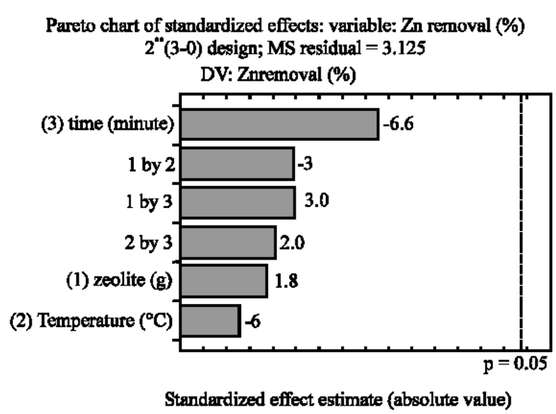

Fig. 4: Pareto chart for zinc removal

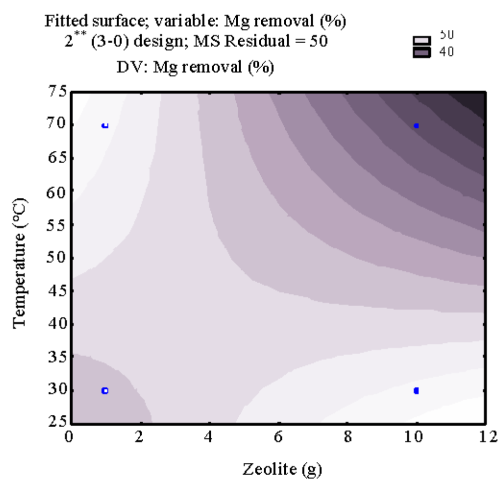

Fig. 5: Magnesium Removal Contour Plot (t = 5 min)

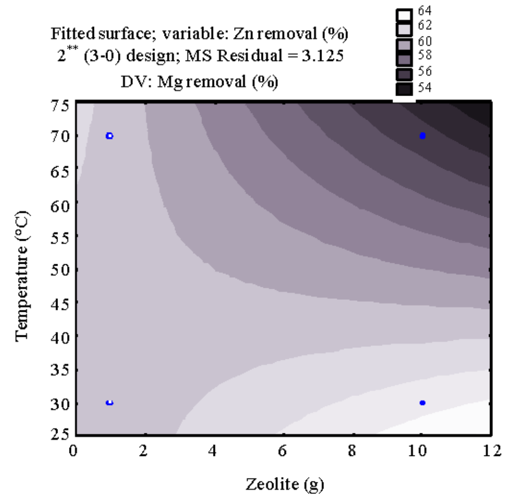

Fig. 8: Zinc removal contour plot (t = 5 min)

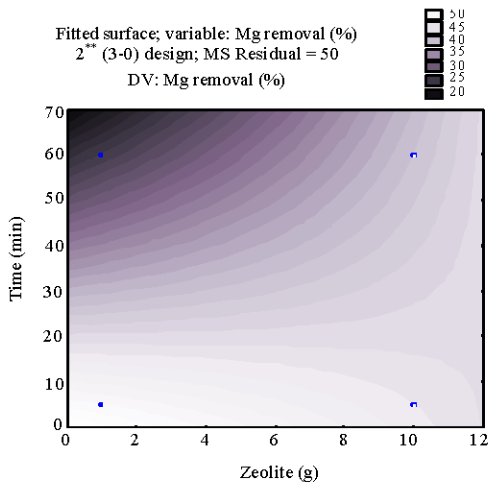

Fig. 6: Magnesium removal contour plot (T = 50°C)

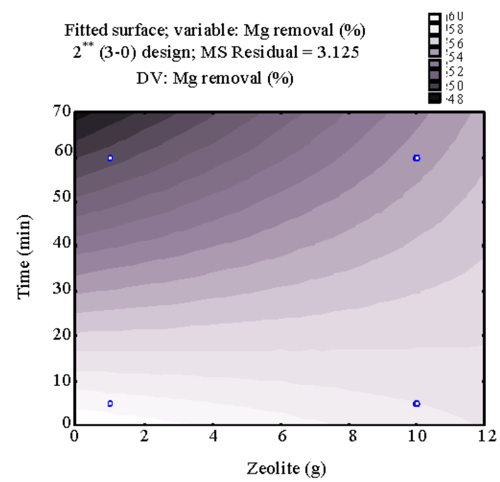

Fig. 9: Zinc removal contour plot (T = 50°C)

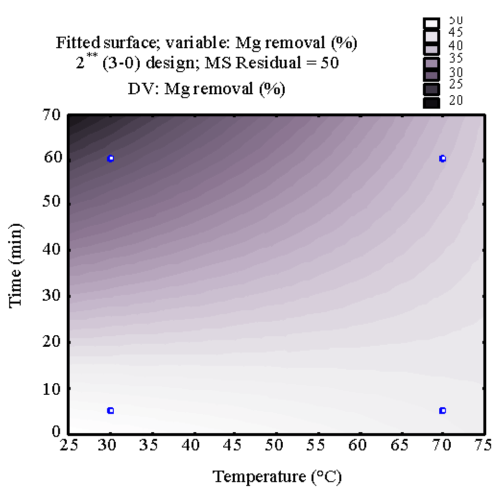

Fig. 7: Magnesium removal contour plot (Zeolite=5 g)

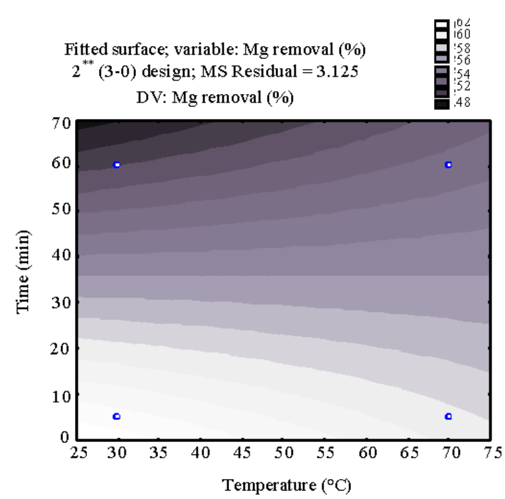

Fig. 10: Zinc removal contour plot (Zeolite = 5 g)

Table 5: The optimum range of parameters in magnesium removal

Figure	Parameter	Optimum range
5	Time	5 min (fixed)
	Temperature	35-45°C
	Zeolite	0-5 g
6	Time	0-10 min
	Temperature	50°C (fixed)
	Zeolite	0-5 g
7	Time	0-10 min
	Temperature	25-45°C
	Zeolite	5 g (fixed)

Table 6: The optimum range of parameters in zinc removal

Figure	Parameter	Optimum range
8	Time	5 min (fixed)
	Temperature	30-50°C
	Zeolite	0-3 g
9	Time	0-10 min
	Temperature	50°C (fixed)
	Zeolite	0-5 g
10	Time	0-10 min
	Temperature	30-50°C
	Zeolite	5 g (fixed)

The optimum range of zinc removal found were in the range of adsorption time of 0 to 10 min, temperature of 30 to 50°C and the amount of zeolite in the range of 0 to 5 g. The zinc removal was 62%.

CONCLUSION

From this study, the following conclusions can be made:

- In the adsorption of heavy metals from recovered base oil, the yield of zinc removal was higher than that of magnesium
- The most influential parameter affecting the magnesium and zinc removal was the time of adsorption
- The optimum parameters for the removal of zinc and magnesium obtained were in the range time of 0 to 10 min, temperature of 30 to 50°C and the amount of zeolite 0 to 5 g
- The optimum of magnesium and zinc removal obtained were 50 and 62%, respectively

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