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Regeneration of Wind Power Waste Lubricating Oil by a Combination Use of Chitosan, Sodium Carbonate and Silica Gel

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ABSTRACT

Waste lubricant oil can be regenerated by flocculation and adsorption to get rid of impurities. In this work, the performance of flocculants (chitosan, sodium carbonate and sodium silicate) and sorbents (carclazyte and silica gel) were evaluated experimentally. The results showed that the combination of chitosan and sodium carbonate had beat results in removal of acid value and mechanical impurities. Silica gel contacting adsorption had the best colour removal. Thermal precipitation (80°C, 24h) was adopted as primary step before flocculation to wipe off heavier particulate matter. The best ratio of chitosan, sodium carbonate and silica gel was 1:1:1, and the best dosage of chitosan was 2% compared to oil. After the re-refining process, viscosity decreased from 186.34 (mm^2/s, 40°C) to 155.21 (mm^2/s, 40°C), the acid value from 0.26 (mg KOH/g) to 0.06 (mg KOH/g), mechanical impurities content from 3.62% to 1.41%, water content from 0.45% to 0.06%, and light absorbance from 4.31 to 1.17.

INTRODUCTION

With the dwindling of fossil energy and increasing demand for electricity, wind power has become one of the most important renewable energy technologies. This power source features many advantages such as accessibility, inexhaustibility and sustainability. According to Bandoc et al. (2018), wind power accounted for 16%~25% of the global total renewable capacity in the year 2017, and the total wind power capacity in China has reached 18000 MW in 2017 (Li et al. 2018). The rapid development of wind power industry has caused abundant waste lubricant problems (Ramasamy et al. 2007).

Lubricant is made of base oil (71.5~96.2 wt%) and chemical additives (metallic detergent, ashless dispersant, oxidation inhibitor, antioxidant wear, friction modifier, pour point depressant, antifoam and so on), playing a dual role in the heat generation reduction and friction reduction (Mohammed et al. 2013). The major difference between fresh lube oil and the used oil is the contaminant contents such as dust, water, particles and oxidative aromatic compounds (Jafari et al. 2015). The important source of oil contamination during use is the chemical disrupt of additives and their interaction to generate corrosive unfavourable components. Compared with other spent lubricating oil generated by vehicles and fuel machines, wind power waste lubricating oil is much less polluted owing to short replacement period, as shown in Fig. 1.

Furthermore, the heavy metal, the hydrocarbon oxidation and PCBs (polychlorinated biphenyls) caused by the friction of the engine device are harmful to environment and human health if it was not disposed appropriately (Scapin et al. 2009). Re-refining or reclamation is the most important method for recycling used lubricating oils to avoid environmental pollution and recycle base-oil. Vacuum distillation, solvent extraction (Kamal et al. 2009, Osman et al. 2018), adsorption (Chen et al. 2018), hydrogenation and pyrolysis are the widespread technologies. One of the main problems associated with distillation is the formation of deposits and hot-spots caused by resinous materials. These deposits may induce coking and fouling of the internal surfaces, which makes continuous operation difficult and producing low-quality base oils (Pinheiro et al. 2018). Solvent extraction may lead to extraction agent residue, leading to lower property of recovery oil. Hydrogenation and pyrolysis need equipment to operate at high temperature and very high vacuum, requiring high investment and operating costs (Rincón et al. 2003). On the other hand, flocculation and adsorption have been reported as a simple, efficient, low energy consumption and economical technology (Scapin et al. 2009).

To date, a number of flocculants are commercially available, but some of them have been received attention for their negative environmental consequences and potential health impacts. For example, the use of inorganic salts, such as aluminium chloride or aluminium sulphate, is

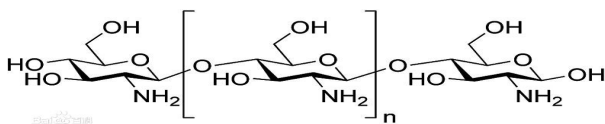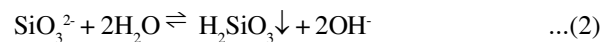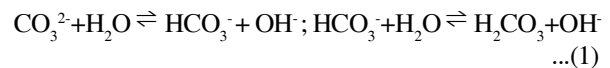Fig. 1: Vehicles machines (a) and wind power (b) waste lubricating oil.

Fig. 2: Chemical structure of chitosan.

now disputed due to their potential contribution to Alzheimer's disease and large usage (Divakaran et al. 2001). Other drawbacks of inorganic coagulants still include highly sensitive to pH, inefficient towards very fine particles and applicable only to a few disperse systems (Bratskaya et al. 2006). Chitosan has attracted enormous attention, because of its biocompatibility, diversity in molar mass and charge density (Feng et al. 2017, Agbovi et al. 2018). Chitosan, obtained by deacetylation of chitin is now available as a commercial product, manufactured from crab and prawn shells. The chemical structure of chitosan resembles that of cellulose very much. While cellulose is a polymer of d-glucose, chitosan is a polymer of d-glucosamine with an NH_2 group in place of the OH group on carbon-2 of d-glucose (Fig. 2) (Du et al. 2018). It is described as a cationic polyelectrolyte and is expected to coagulate negatively charged suspended particles (Haufe et al. 2017). The physical-chemical properties of chitosan related to the presence of amine functions (acid-base properties, solubility,

cationicity) make it very efficient for binding metal cations in near neutral solutions. Chitosan has been widely used for the coagulation-flocculation of mineral colloids (Feng et al. 2017), in biomedicine (Guibal et al. 2007) and dye-containing solution (Rehna et al. 2013).

To improve the capacity of adsorbing positive particles, strong alkali salts, such as KOH, NaOH, were commonly added. Strong nucleophile OH^- can break polar bonds, causing metal precipitation. Meanwhile, addition of OH^- can result in a compression of the electrical double layer surrounding each suspended particle, resulting in coagulation (Pinheiro et al. 2017). In addition, OH^- promotes saponification reactions between long chain fatty acids or esters and the alkali, yielding soap. Soaps have a thickening effect on the oil, leading to gel-type oil (Martín-Alfonso et al. 2009). The production of a gel during separation steps reduces the lubricant's fluidity, causing blocking problems in the regeneration process. To reduce the saponification reactions, weak acid root salts such as Na_2CO_3 and Na_2SiO_3 were studied in the work. Weak acid root salts occur hydrolysis reaction with water, yielding OH^-, as Eqs. (1) and (2).

$$CO_3^{2-}+H_2O \rightleftharpoons HCO_3^- + OH^-; HCO_3^- +H_2O \rightleftharpoons H_2CO_3+OH^- \quad ...(1)$$

$$SiO_3^{2-} + 2H_2O \rightleftharpoons H_2SiO_3\downarrow + 2OH^- \quad ...(2)$$

To reach better physical separation and enhance the chemical properties goal of spent oil, the combination of flocculation and adsorption was used in the work. The flocculation efficiency of chitosan, Na_2CO_3 and Na_2SiO_3 was studied. Carclazyte and silica gel, as the common adsorbents in industrial manufacture, were also studied in the work to remove colour. The best conditions of the better flocculant and adsorbent were investigated by orthogonal test. The results should provide a sound basis for developing an environmental friendly and economically competitive method for used lubricant oil treatment.

MATERIALS AND METHODS

Materials Used

Waste lubricants were gathered from a gearbox of the wind power station in Yumen city (Gansu province, China). The oil samples were used as such without any further pretreatment. The physicochemical properties of the waste

Table 1: The properties of waste wind power lubricant oil.

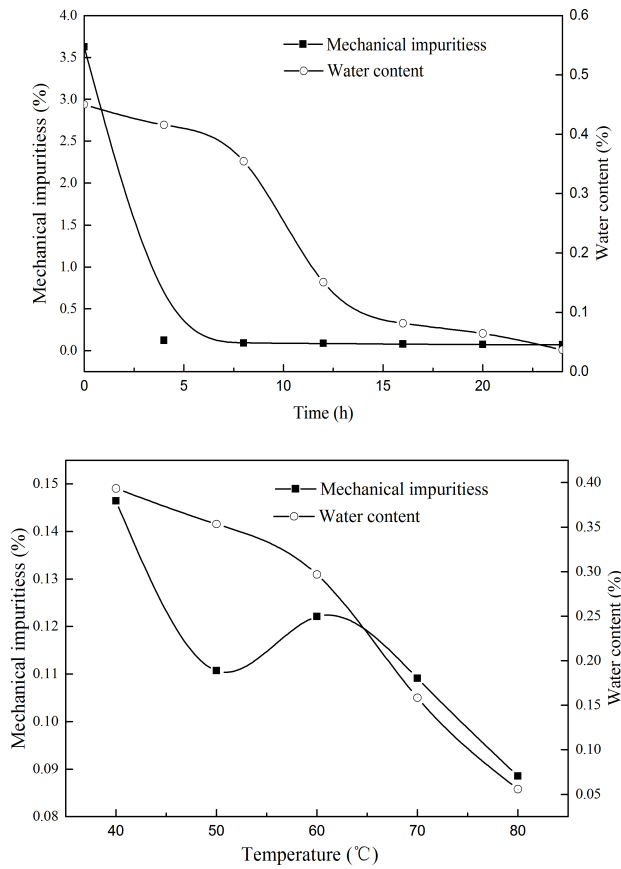| Acid value (mgKOH/g)	Viscosity (40°C, mm²/s)	Viscosity (100°C, mm²/s)	Impurity (%)	Water (%)	UV-Vis absorbance
0.258	186.336	32.84	3.625	0.45	4.310

Fig. 3: Mechanical impurities content and water content change under different precipitation time (a) and temperature (b).

wind power lubricant oil are presented in Table 1.

All reagents (petroleum ether, 95% ethyl alcohol, anhydrous sodium carbonate, sodium silicate, chitosan, cresol red, potassium hydroxide, anhydrous toluene, Karl Fischer reagent) were of analytical grade. Activated clay (activity degree: 85 mmol/kg) and solvent naphtha (SH004) were purchased from Tianjin Guangfu Fine Chemical Research Institute. The size of the silica gel was 30~50 mesh.

Procedure

Thermal precipitation: To improve the efficiency of flocculation and adsorption, a thermal precipitation test re-

moving part of water and larger particles was conducted. 200 g waste lubricating oil was heated under a controlled temperature from 40°C to 80°C. The precipitation time was also tested.

Flocculation: The pre-thermal precipitated waste lubricating oil was then mixed with a fixed amount of 1wt% of different flocculants in a beaker. The mixture was then heated to 70°C in water bath, kept the temperature and stirred for 10 min at 1000 r/min. Then the mixture was kept for gravity setting for 6 h. After that, the recovered oil was separated from the mixture. The yield of oil was calculated on the basis of initial mass of used oil taken and then analysed for different physical properties. After choosing the best flocculant, the best using condition was studied by an orthogonal experiment.

Adsorption: The spent oil after flocculation was then mixed with a fixed amount 1wt% of different adsorbents in a beaker. The mixture was then stirred for 30 min at a frequency of 1000 r/min. The beaker was then put into a drying oven at a controlled temperature of 120°C. Meanwhile, the oil was treated by adsorbents filtration to provide a reference. After choosing the better adsorbents and the better way, the best treating condition was studied further.

Analysis of Samples

Viscosity: The viscosity mentioned in work was kinematic viscosity (40°C), determined by Chinese national standard "Petroleum products-Determination of kinematic viscosity and calculation of dynamic viscosity" (GB265-88).

Acid value: Acid value was measured by Chinese national standard "Petroleum products-Determination of acid number" (GB264-88).

Water content: Water content was measured by Karl Fischer Moisture Titrator (Metrohm 787 KF Titrino).

Mechanical impurities content: Mechanical impurities content was measured by Chinese national standard "Petroleum products and additives-Determination of mechanical" (GB/T511-88).

Colour change: The colour changing for different treatment was measured by UV/VIS spectrophotometer (EVO-

Table 2: Prediction models of thermal precipitation.

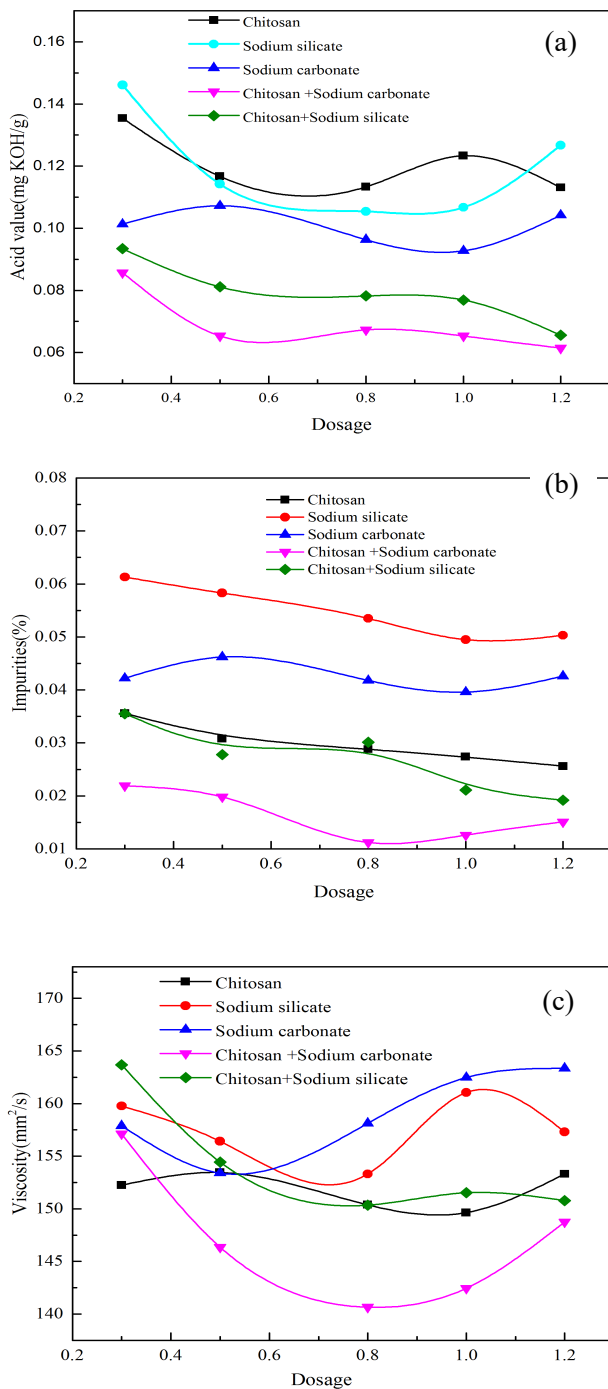| Dependent variable (%)	Independent variable	Prediction models	R^2	Significance
Impurities removal	Time (h)	$y = e^{(4.588 - 0.069/x)}$	0.987	<0.001
Water removal	Time (h)	$y = e^{(5.066 - 12.63/x)}$	0.951	0.001
Impurities removal	Temperature (°C)	$y = 89.453x^{0.019}$	0.777	0.048
Water removal	Temperature (°C)	$y = x^{2.874}$	0.990	<0.001

Fig. 4: Dosage (wt%, flocculant to oil) to acid value (a), impurities content (b), and viscosity (c).

LUTION 300) at the maximum absorption wavelength of 329 nm.

Statistical analysis: Statistical analysis was carried out using SPSS software.

RESULTS AND DISCUSSION

Thermal Precipitation

Mechanical impurities content and water content decreased by increasing settling time and settling temperature (Fig. 3). Rate of settling was maximum during the initial 4 hours. Then the mechanical impurities removal rate nearly kept in a constant while the time increased. It may be caused by the density of particles. Heavier particles were wiped out completely in the initial 4 hours, while smaller and lighter particles formed colloform. Colloform was difficult to remove. A spent small oil drop and water drop can lead to stable emulsion, which can be broken by heating. Higher temperature destabilized the colloidal system, promoting the aggregation of the particles by Van der Waals forces. After settling 8 h at 70°C, water content decreased from 0.45% to 0.15%, and mechanical impurities decreased from 3.63% to 0.11%. The efficiencies were 66.67% and 96.96%, respectively.

To further study the relationship between responses (mechanical removal and water removal) and independent factors (time and temperature), predicated models were made (Table 2). It was concluded that impurities removal was obviously influenced by time, while water removal was influenced by temperature in the thermal precipitation process. In other words, to get better impurities removal, precipitation time should be extended. And to get better water removal, temperature of the oil should be appropriately heated.

Flocculation

Selection of flocculant: Flocculation was used to collide and adhere small droplets to form larger flocs for the improvement of sedimentation. The interaction between flocculants and particles may be induced by interparticle bridging, charge neutralization and patch adsorption. As shown in Fig. 4, it can be seen that in general, combination of chitosan and sodium carbonate had the best efficiency in acid value, viscosity decreasing, and impurities removal. The graphs showed that after the presence of the best condition (lowest acid value, viscosity and impurities) a further increase in the amount of flocculant to oil ratio would result in increasing the acid value, viscosity and impurities.

From Fig. 4, it can be concluded that different flocculants had the different influence on acid value, viscosity and impurities. The difference between them was related to the coagulation capacity of these flocculants.

Chitosan as a cationic polyelectrolyte mainly coagulated negatively charged suspended particles while sodium carbonate and sodium silicate as negion polyelectrolyte

Table 3: Prediction models of flocculation process while the combination of chitosan and sodium carbonate as flocculant.

Dependent variable (%)	Independent variable	Prediction models	R^2	Significance
Acid value removal	Ratio	$y = 68.565 - 0.601/x$	0.725	0.067
	Dosage	$y = 78.727 - 1.711/x$	0.952	**0.005**
	Stirring time	~	~	~
	Stirring temperature	$y = e^{(4.488 - 5.674/x)}$	0.711	0.07
	Sedimentation temperature	$y = e^{(4.264 - 0.824/x)}$	0.726	0.067
	Sedimentation time	~	~	~
Viscosity decreased percentage	Ratio	$y = e^{(2.874 - 0.056/x)}$	0.894	0.015
	Dosage	~	~	~
	Stirring time	$y = 12.6 + 1.182x - 0.044x^2$	0.934	0.066
	Stirring temperature	$y = 12.394e^{0.004x}$	0.675	0.088
	Sedimentation temperature	$y = e^{(3.379 - 20.923/x)}$	0.775	**0.049**
	Sedimentation time	$y = e^{(3.17 - 1.029/x)}$	0.861	**0.023**

Note: significance values in bold are statistically significant. '~' means significance value > 0.1, there is no relation.

mainly coagulated positively charged suspended particles. Charge neutralization occurred when a positively charged molecule adsorbed particles carrying negative charges. However, as the average distance between charged chitosan segments was much smaller than that between negatively charged surface sites, positively charged segment of chitosan neutralizing each negatively charged surface site was difficult. So, it formed 'patches', an aggregation of positively and negatively charged particles. Increasing the dosage of chitosan, the positive charge was excessive, leading to repulsive interaction. Flocs were broken, leading to increase in acid value, viscosity and impurities.

Because the acidity of H_2CO_3 was higher than H_2SiO_3, hydrolysis constant of CO_3^{2-} was higher than SiO_3^{2-}. In other words, when the amount of Na_2CO_3 and Na_2SiO_3 was equal, Na_2CO_3 yield more OH^-, which lead to stronger flocculation ability. So, lower acid value and impurities were observed in sodium carbonate treatment, as displayed in Figs. 4 a and 4b.

Single factors changing: Chitosan and sodium carbonate were combined and used in the treatment. To further study the relationship between responses (acid value removal and viscosity decreased percentage) and independent factors, a series of single factors changing experiments were made. Independent factors were ratio (sodium carbonate: chitosan, w/w), dosage (the amount of chitosan and sodium carbonate to oil, wt%, among them, the ratio of two flocculants was 1:1), stirring time (min), stirring temperature (°C), sedimentation temperature (°C), and sedimentation time (h). Predicated models were made as Table 3.

Orthogonal test: Because a high flocculation efficiency is desirable, it was very important to identify the optimum parameters, while the combination of chitosan and sodium

carbonate as flocculant, to achieve high efficiency. However, the study of the effects of changing single factors on flocculation efficiency was not sufficient to judge that, which parameter values were optimum because of interaction between different factors. Therefore, the optimum parameters should be obtained using orthogonal test design and mathematical analysis.

Six controllable variables, namely, ratio (chitosan: sodium carbonate), dosage (the percentage of chitosan to spent oil), stirring time (min), stirring temperature (°C), sedimentation temperature (°C) and sedimentation time (h), were selected for optimization, each at five levels. With reference to the experimental design theory, the orthogonal array L_{25} was selected to arrange the test program. The test results are listed in Table 4. As the results indicated, in our study range, the ranking of the six influence factors on the acid value removal was sedimentation time > stirring temperature > ratio > stirring time > dosage > sedimentation temperature and on the viscosity decrease was ratio > dosage > sedimentation time > sedimentation temperature > stirring temperature > stirring time. The optimum flocculation condition to acid value removal was achieved using 3:1 ratio, 2% dosage, stirring 20 min at 60°C, and then precipitating 5 h at 70°C. The optimum flocculation condition to viscosity decrease percentage was achieved using 3:1 ratio, 1% dosage, stirring 10 min at 60°C, and then precipitating 9 h at 70°C.

Adsorption

Selection of adsorbent: Carclazyte and silica gel as low cost and eco-friendly adsorbents are popular in the industry. The whole colour removal process can be divided into

Table 4: Results of L25 orthogonal experiment while the combination of chitosan and sodium carbonate as flocculant.

Sample numbers	Ratio	Dosage	Stirring time (min)	Stirring temperature (°C)	Sedimentation temperature (°C)	Sedimentation time (h)	Acid value removal (%)	Viscosity decreased percentage (%)
1	1:3	0.50%	5	40	40	4	82.24	17.89
2	1:3	1.00%	10	50	50	6	80.70	19.50
3	1:3	1.50%	15	60	60	8	81.46	21.11
4	1:3	2.00%	20	70	70	10	81.65	19.50
5	1:3	2.50%	25	80	80	12	65.45	14.67
6	1:2	0.50%	10	60	70	12	78.99	20.57
7	1:2	1.00%	15	70	80	4	80.12	22.18
8	1:2	1.50%	20	80	40	6	76.01	18.43
9	1:2	2.00%	25	40	50	8	79.69	19.50
10	1:2	2.50%	5	50	60	10	79.15	21.11
11	1:1	0.50%	15	80	50	10	72.85	16.28
12	1:1	1.00%	20	40	60	12	76.41	18.96
13	1:1	1.50%	25	50	70	4	76.63	18.43
14	1:1	2.00%	5	60	80	6	77.66	16.82
15	1:1	2.50%	10	70	40	10	77.26	17.89
16	2:1	0.50%	20	50	80	8	84.11	15.74
17	2:1	1.00%	25	60	40	10	81.40	18.96
18	2:1	1.50%	5	70	50	12	75.84	14.67
19	2:1	2.00%	10	80	60	4	81.27	17.35
20	2:1	2.50%	15	40	70	8	81.65	15.21
21	3:1	0.50%	25	70	60	6	79.15	21.11
22	3:1	1.00%	5	80	70	8	78.99	20.57
23	3:1	1.50%	10	40	80	10	80.12	22.18
24	3:1	2.00%	15	50	40	12	79.88	18.43
25	3:1	2.50%	20	60	50	4	83.57	19.50

Table 5: Prediction models of adsorption process.

Dependent variable	Independent variable	Prediction models	R^2	significance
A/A_0	Dosage	$y = 0.343 + 0.054/x$	0.882	**0.018**
	Stirring time	~	~	~
	Stirring temperature	$y = 0.077 + 0.016x + 0.001x^2$	0.991	**0.009**
	Sedimentation temperature	~	~	~
	Sedimentation time	~	~	~
Recovery efficiency (%)	Dosage	$y = 79.543 + 2.74/x$	0.808	**0.038**
	Stirring time	$y = 77.58 + 0.41x - 0.012x^2$	0.986	**0.014**
	Stirring temperature	$y = e^{(4.422 - 2.245/x)}$	0.758	**0.055**
	Sedimentation temperature	~	~	~
	Sedimentation time	~	~	~

Note: significance values in bold are statistically significant; '~' means significance value > 0.1, there is no relation.

three stages: physical adsorption stage, internal diffusion stage and chemical adsorption stage. There was no clear boundary among the three divided stages, and the adsorbent process can occur simultaneously and continually until the adsorbent structure was saturated and deactivated.

Carclazyte [$H_2Al_2(SiO_3)_4 \cdot nH_2O$] has known to have special adsorptive properties, because of the unique crystal structure and surface charge properties. The crystallographic structure suggested that kaolinate particles consisted of a silica tetrahedral surface and an aluminium hydroxide octahedral (Gupta et al. 2011). Because of the pH change, silica face and alumina face present negative charge or positive charge. Suzzoni et al. (2018) showed that the specific surface area of carclazyte was 21 m²/g, and the average diameter was 230 nm. Silica gel had great adsorption capability due to its big specific surface area (~500 m²/g) and numerous hydroxyl groups (Hua et al. 2017). Hydroxyl group of silica gel can interact with amine groups and carbonyl
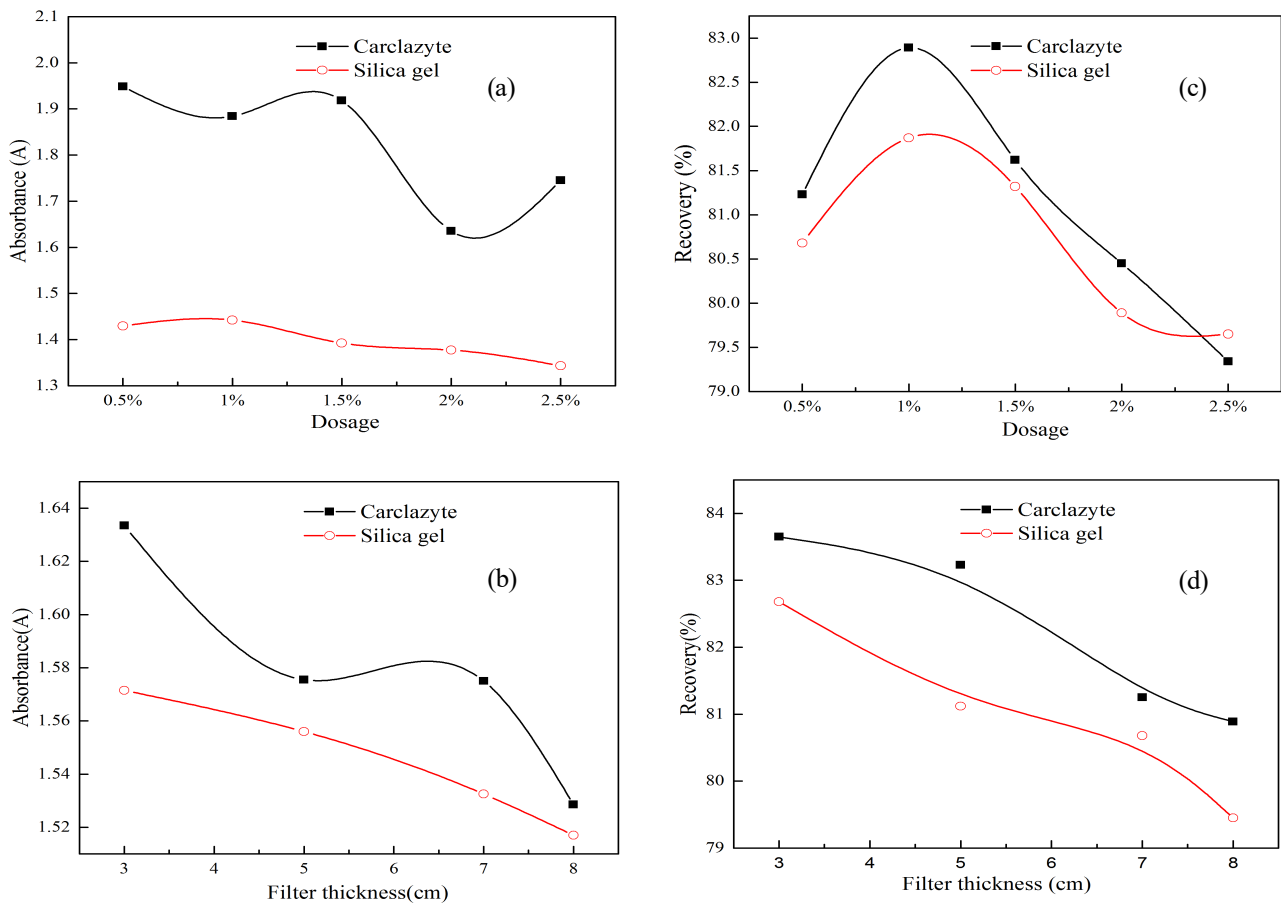
Fig. 5: Dosage (wt%) of particle dispersion technique (a) (c), and thickness of filter technique (b) (d) to recovery efficiency (%) and absorbance.

groups in spent oil. Because of higher surface area and stronger chemical adsorption, the adsorption capacity of silica gel was larger than carclazyte (Fig. 5 a). Silica gel was better than carclazyte in wind power waste lubricating oil colour removal.

Comparing the efficiency of contacting ways, as presented in Figs. 5a and 5b, particle dispersion technique had better colour removal efficiency than filter technique. This phenomenon may be because of the contacting surface of dispersion technique was larger than filter, providing more adsorption sites. Hence, it was observed that silica gel dispersion treatment had the lower absorbance than silica gel filter. From Figs. 5c and 5d, it was observed that silica gel had the lower recovery rate than carclazyte, which may be caused by the lager adsorption of silica gel.

Single factors changing: Silica gel dispersion treatment was used in the treatment. To further study the relationship between responses (A/A_0 and recovery efficiency) and inde-

pendent factors, a series of single factors changing experiments were made. Independent factors were ratio dosage (silica gel, wt%), stirring time (min), stirring temperature (°C), sedimentation temperature (°C), and sedimentation time (h). Predicated models were made as given in Table 5.

Orthogonal test: Five controllable variables, namely, dosage (the percentage of carclazy to spent oil), stirring time (min), stirring temperature (°C), sedimentation temperature (°C) and sedimentation time (h), were selected for optimization, each at four levels. The test results are listed in Table 6. As results indicated, the ranking of the influence of each of the five factors on the colour removal was stirring temperature > dosage > stirring time > sedimentation temperature > sedimentation time, and on the recovery efficiency was sedimentation time > stirring temperature > dosage > sedimentation temperature > stirring time. In other words, five controllable variables had a different influence on the colour removal and recovery efficiency.

Table 6: Results of L16 orthogonal experiment.

Sample	Dosage	Stirring time (min)	Stirring temperature (°C)	Sedimentation temperature (°C)	Sedimentation time (h)	A/A_0	Recovery efficiency (%)
1	0.50%	5	40	40	3	0.27	81.70
2	0.50%	10	50	50	5	0.28	80.90
3	0.50%	15	60	60	7	0.22	81.54
4	0.50%	20	70	70	9	0.27	76.98
5	1%	5	50	60	9	0.26	78.44
6	1%	10	40	70	7	0.30	73.99
7	1%	15	70	40	5	0.27	80.47
8	1%	20	60	50	3	0.25	81.26
9	1.50%	5	60	70	5	0.23	80.37
10	1.50%	10	70	60	3	0.30	75.69
11	1.50%	15	40	50	9	0.32	79.76
12	1.50%	20	50	40	7	0.33	76.47
13	2%	5	70	50	7	0.25	78.69
14	2%	10	60	40	9	0.23	80.34
15	2%	15	50	70	3	0.25	79.18
16	2%	20	40	60	5	0.27	84.89

The optimum adsorption condition to colour removal was achieved using 2% dosage, stirring 5 min at 60°C, and then precipitating 5 h at 70°C. The optimum adsorption condition to recovery efficiency was achieved using 2% dosage, stirring 15 min at 60°C, and then precipitating 5 h at 70°C. Overall consideration of colour removal and recovery efficiency, the optimum adsorption condition was 2% dosage, stirring 15 min at 60°C, and then precipitating 5 h at 70°C.

CONCLUSIONS

Thermal precipitation followed by flocculation and adsorption was one of the competitive process for recycling waste wind power lubricant oil. In this paper, organic flocculants and inorganic flocculants were studied, and found that their combined use had better effects. Silica gel was better than carclazyte in colour removal.

The best thermal precipitation condition was at 80°C temperature and sedimentation time of 24 h. The best ratio of chitosan, sodium carbonate and silica gel was 1:1:1, and the best dosage of chitosan was 2% compared to oil. After the re-refining process, viscosity decreased from 186.34 (mm²/s, 40°C) to 155.21 (mm²/s, 40°C), while the acid value decreased from 0.26 (mg KOH/g) to 0.06 (mg KOH/g), mechanical impurities content decreased from 3.62% to 1.41%, water content decreased from 0.45% to 0.06%, and light absorbance decreased from 4.31 to 1.17.

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