THE REGENERATION TREATMENT OF WLO BY MEANS OF FLY-ASH ADSORPTION

Ping OUYANG[1*], Xianming ZHANG[1]

For studying the application with high additonal value by fly-ash adsorption in the regeneration of waste lubricating oil(WLO), both of X-ray diffractometer (XRD) and scanning electron microscope (SEM) were applied to exploring the phases and the overall morphology of fly-ash, the adsorption effect of fly-ash on WLO were examined by the comprehensive comparative experiments with activated clay, and the relative wettability of the two to oil was discussed by means of contact angle meter. The experimental results showed that the relative optimum technological conditions, for the acid value of WLO as the index of investigation, are as follows: 12 % of fly-ash dosage, 60 min of adsorption time, 90 ℃ of adsorption temperature and 950 r/min of agitation rate, and under this condition, the adsorption effect of fly-ash was equivalent to that of activated clay, and the surface energy of raw fly-ash could be improved by physical and chemical modifications to enhance its wettability and affinity to oil, so as to improve the adsorption effect of fly-ash on WLO.

Keywords: Fly-ash; adsorption; waste lubricating oil (WLO); regeneration

1. Introduction

China is the second largest consumer of lubricating oil (LO) on the earth after the United States, with apparent consumption of lubricants exceeding 6 million tons in 2018. In the context of globally accelerating energy shortage, the recycling and regeneration of WLO have become an important and urgent issue that must be solved [1,2]. At present, the regeneration processes of WLO are often divided into re-purification, re-refinement and re-distillation, but the adsorption method is the basis of these processes, natural or man-made adsorbents are used to remove various impurities in WLO, thus improving oil quality [3,4].

The commonly used adsorbents for regeneration of WLO are mainly activated clay, silica gel, molecular sieve, etc [5,7]. However, due to the high cost, the complex acquisition process (such as silica gel, molecular sieve, etc.), or the serious environmental pollution (such as clay), some adsorbents have restricted the adsorption method from being applied to the waste oil regeneration process

[1] Engineering Research Center for Waste Oil Recovery Technology and Equipment of Ministry of Education, Chongqing Technology and Business University, Chongqing 400067, China, e-mail: oyp9812@126.com

[8,9]. Therefore, it has become an important research subject to explore efficient and environmental adsorbents that can be acquired easily at low cost.

China is the largest coal consumer and producer in the world. In 2018, more than 6 million tons of fly-ash were produced in China. The accumulation of abundant fly-ash has led to serious pollution to the atmosphere, water and soil [10]. Fly-ash, formed in the process of coal combustion, is a tiny melted noncombustible substance. It exists in the form of spherical particle under the action of surface tension. It is characterized by high chemical internal energy and strong adsorption activity due to the rapid cooling in its generation process [11,12]. It has been widely applied in the adsorption treatment of wastewater, waste gas and microwave [13-21]. And in the treatment of WLO, the adsorption of fly-ash is considered to reduce the labor intensity and production cost of WLO regeneration [22-26], but there has barely been relevant systematic experimental research report.

Therefore, in this article, raw fly-ash was used as an adsorbent to regenerate WLO, XRD and SEM were used to study the structure and the surface properties of fly-ash. The comprehensive comparative experiments with activated clay were performed for investigating the adsorption effect of WLO by fly-ash. Also, a comparative experiment was made to dispose the WLO separately through fly-ash and activated clay, and powder contact angle tester was used to explore the relative wettability of both fly-ash and clay to oil, thus providing theoretical and experimental foundation for the concept of "Treating Waste with Waste" and the application with high additional value by means of fly-ash adsorption in the regeneration of WLO.

2. Materials and Methods

2.1. Materails and Instruments

Main materials: fly-ash (raw fly-ash from a heat-engine plant in the Province of Guizhou); activated clay (CS-1060, 75 microns from Wuhu Feishang Mining Development Co., Ltd in Anhui Province); WLO (from a factory recycling industrial waste oil in the City of Chongqing with an acid value of 2.892 mgKOH/g); potassium hydroxide, ethyl alcohol, methylbenzene, petroleum ether (30-60℃) and isopropanol, liquid paraffin (the analytical reagent from Chongqing Chuandong Chemical (Group) Co., Ltd with surface tension of 29.536 mN/m and dynamic viscosity of 2.2 CP); and distilled water (instantly consumed with surface tension of 72.6 mN/m and dynamic viscosity of 1 CP).

Main instrument: D/MAX-2500 X-ray diffractometer (Rigaku, Japan); VEGA Ⅱ LMU scanning electron microscope (TESCAN, Czech Republic);

titrando 905 potentiometric titrators (from Metrohm); JF99A powder contact angle tester (from Shanghai zhongchen digital technic apparatus Co., Ltd).

2.2. Experimental Method

2.2.1. Pretreatment of fly-ash and its characterization

The raw fly-ash came from a heat-engine plant in the Province of Guizhou. At first, large pieces of impurities were eliminated from the raw fly-ash by using a 250-micron test sieve, which was then washed for 2 or 3 times to get rid of the dust covered on the surface of fly-ash by the clean water. After that, the fly-ash was dried for 7h in an electrically heated drying cabinet and successively passed through the 100-micron, 75-micron and 40-micron seive. The fly-ash with 75-micron size was chosen and used in our experiments. Both of XRD and SEM were used for the phase analysis and overall morphological analysis respectively on the fly-ash acquired for experimental purpose.

2.2.2. Index selection and measurement on the treatment effect of WLO by the adsorption of fly-ash

Complex impurities, such as water, solid impurities and various oxidized, metamorphic and acidic compounds are contained in WLO, where the acids mainly originate from the lubricating additives with acid group, the organic acid produced by the oxidation of hydrocarbons and the acid generated after the burning of sulfurous fuels. Acids can be grouped into the following types, including the organic acid, the inorganic acid, the esters, the phenolic compounds, the heavy metal salts and the other salts of weak base. Also it can be produced by water oxidation in LO, the generation of heat by friction, and the thermal oxidation of impurities.

Therefore, besides indicating the acid content, acid value can also indirectly reflect the content of the other impurities in WLO. Compared with the other indexes, it’s a more representative index to measure the deterioration degree of LO in a machine. Also as an important physical and chemical index to evaluate the properties of LO, the acid value is the total content of inorganic acids and organic acids in LO, which can be expressed by the milligrams of potassium hydroxide required to neutralize 1 g of petroleum products, and it’s denoted by the unit of mgKOH/g. A decrease in acid value represents the less deterioration of LO, while an increase in acid value means the higher degree of deterioration of LO.

The determination of acid value in LO can be carried out by the 905 automatic potentiometric titrator of Metrohm, based upon the “Standard test method for acid value of petroleum products by potentiometric titration” of Chinese national standard GB/T 7304-2014 [27].

2.2.3. The adsorption treatment of WLO by fly-ash

50.0g WLO was accurately weighed into the three-necked flask which had been placed in a magnetic stirring device with heating function of oil bath, and a certain dosage of experimental flyash and activated clay was respectively added into the three-necked flask as required, and then the adsorption was carried out under a certain agitation rate, adsorption temperature and time. After the adsorption was made each time, the oil was cooled to the ambient temperature after the static settlement and then added with a proper amount of petroleum ether (30-60 ℃) for washing and settlement. Then the oil supernate was filtered and measured according to the China GB/T 7304-2014 Test Method for Acid Value of Petroleum Products in terms of the acid number after the solvent was distilled off.

2.2.4. The relative wettability between oil and two powders (fly-ash and activated clay)

Experimental fly-ash and activated clay were respectively put into sample tubes. Distilled water was chosen as perfect wetting liquid for comparison. With Washburn method, JF99A powder contact angle tester was used to measure advancing contact angle. Under the condition that the addition of sample powders to water was considered as perfect wettability, the corresponding system parameter β was obtained. Then, the relative wetting contact angles between oil and sample powders were measured on the basis of β under the same experimental conditions.

3. Results and Discussion

3.1. Analyses on Fly-ash Characterization

3.1.1. XRD analysis

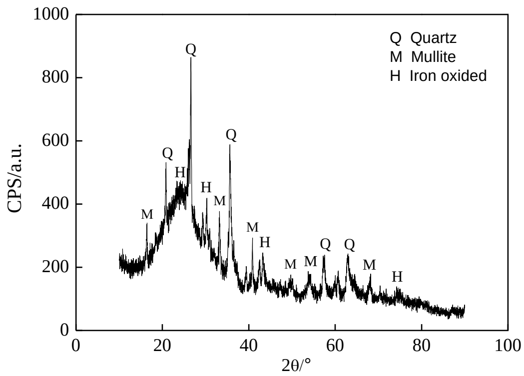
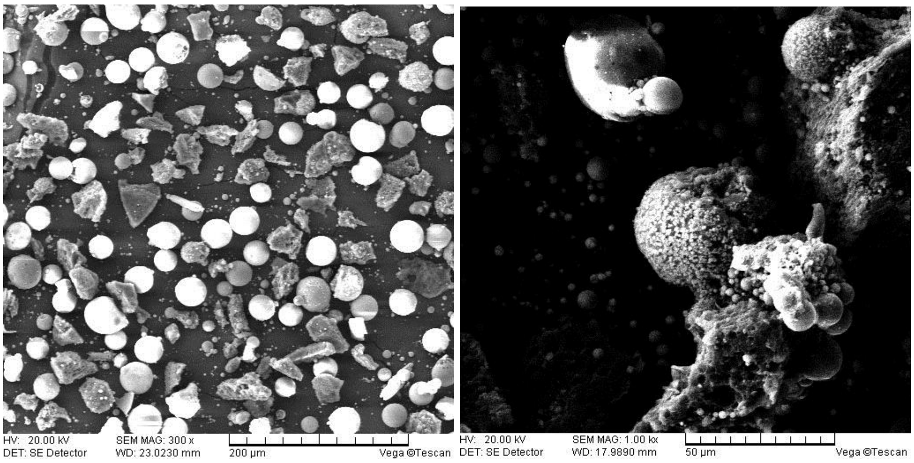


Fig. 1. XRD pattern of fly-ash

The XRD pattern of fly-ash after the pretreatment is shown in Fig. 1, which can give the composition and content of the fixed structure phase composed of various elements. According to the diffraction angle value of the fly-ash diffraction peak, the mineral composition of fly-ash mainly included mullite (Al_2O_3•SiO_2), quartz and iron oxide. A broad characteristic diffraction peak appears within the region of 15°~30°, revealing that abundant Si-Al glasses rich in Si-O-Si bonds and Al-O-Al bonds can be contained in the fly-ash. Therefore, it should be rich [28] in the active constituents of Si and Al. Moreover due to the strong electronegative carbonyl functional groups, such as the alcoholic hydroxyl group, the aldehyde group, the ketone group and the carboxy group [29] on the surface of the Si-Al structure, fly-ash should have good surface activity, which constitutes the material basis of fly-ash with excellent adsorption property.

3.1.2. SEM analysis



(a) (b)

Fig. 2. SEM picture of fly-ash (a) amplification of 300X; (b) amplification of 1.00kX

The SEM picture for the material composition of raw fly-ash is shown in Fig. 2(a) with an amplification of 300X. It can be observed that fly-ash are constituted by circular beads and irregular crystals. Most of the circular beads are hollow glass beads, while the irregular crystals mainly consist of some irregular glass beads and the unburned carbon granules. Glass beads dominate in the fly-ash. The SEM picture for the surface topography of fly-ash constituents is provided in Fig. 2(b) with an amplification of 1.00kX. It can be seen that that no matter if it is the circular bead or the irregular crystal, all of them have a rough surface with abundant, loose pore structure and uniform pore distribution. However, it is just the uneven surface, the gap between the irregular crystals and the beads as well as the pores on the surface of the material, which constitutes the structural basis of fly-ash with good adsorption property.

3.2. Adsorption Treatment of WLO

3.2.1. Adsorption time

Adsorption time has an important effect on the adsorption of adsorbates in WLO. The effect of fly-ash and activated clay adsorption on the acid value of WLO is revealed in Fig. 3 under the following experimental conditions: 100 ℃ of adsorption temperature, 1000 r/min of agitation rate, 5 wt% of adsorbent dosage, and respectively for 30 min, 45 min, 60 min, 90 min, 120 min, 150 min of adsorption time .

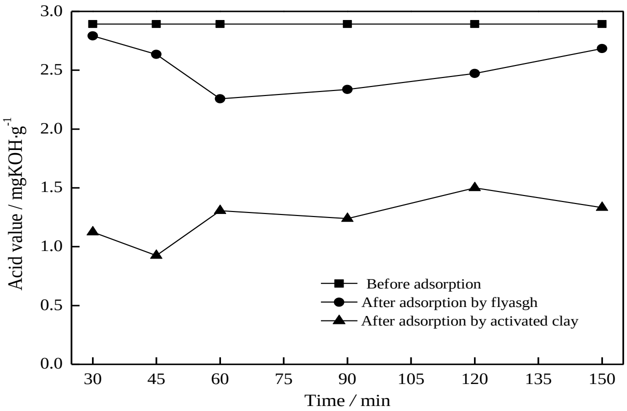


Fig. 3. Adsorption effect by fly-ash and activated clay with different adsorption time

The acid value of the WLO declines significantly by the treatment of fly-ash and activated clay after the adsorption time observed from the Fig. 3. However, the adsorption effect of activated clay is superior to that of fly-ash. The maximum difference between the effects of activated clay and fly-ash is about 2 mgKOH/g, while the minimum is around 0.8 mgKOH/g. Under this adsorption condition, the adsorption effect of fly-ash and activated clay is quite different. In addition, with the increase of adsorption time, the adsorption of fly-ash and activated clay reaches the optimum value, that is to say, the acid value of waste oil is the lowest relative value. This is because along with the adsorption, the active site of the adsorbent is covered by adsorbate, and the adsorption tends to be balanced, but too much time of stirring leads to friction of mixing blade, oil and adsorbent, producing heat or increasing oxidation. As a result, the further deterioration of the oil and the absorption of the adsorbate is promoted so as to enhance the acid value of oil. The operation time of subsequent precipitation and refining is likely to be lengthened.

3.2.2. Adsorption temperature

Adsorption temperature plays an important role in the adsorptive regeneration of WLO. The effect of fly-ash and activated clay adsorption on the acid value of WLO is shown in Fig. 4 under the following experimental conditions: 60 min of adsorption time, 1000 r/min of agitation rate, 5 wt% of adsorbent dosage, and respectively for 80 ℃, 90 ℃, 100 ℃, 110 ℃, 120 ℃, 130 ℃ of adsorption temperature.

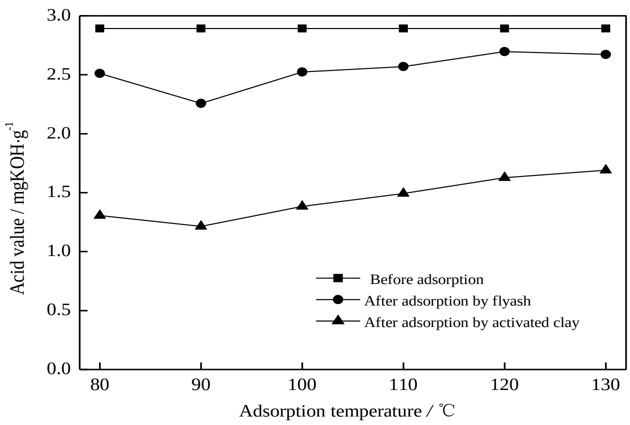


Fig. 4. Adsorption effect of fly-ash and activated clay with different adsorption temperature

The acid value of the WLO is reduced significantly by the treatment of fly-ash and activated clay at the adsorption temperature seen from the Fig. 4. The adsorption effect of activated clay is superior to that of fly-ash. The maximum difference between the effects of activated clay and fly-ash is about 1.4 mgKOH/g, while the minimum is around 0.6 mgKOH/g. Under the minimum difference condition, the difference between the effects of activated clay and fly-ash is becoming smaller and the fly-ash adsorption effect gets better. When the adsorption temperature is inferior to 90 ℃ for the adsorption regeneration of WLO by fly-ash and activated clay, the acid value of oil presents a decreasing tendency with the rise of adsorption temperature. However, when the adsorption temperature is superior to 90 ℃ for the adsorption regeneration, the acid value of oil will increase with the progressive improvement of temperature. It is maybe attributed to the desorption of adsorbates in the temperature range over 90 ℃. And with the progressive growth of temperature, the oil oxidizes and deteriorates further to enhance the acid value of oil.

3.2.3. Adsorbent dosage

The effect of fly-ash and activated clay adsorption on the acid value of WLO is illustrated in Fig. 5 under the following experimental conditions: 90 ℃ of adsorption temperature, 1000 r/min of agitation rate, 60 min of adsorption time, and respectively for 2 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%, 25 wt% of adsorbent dosage.

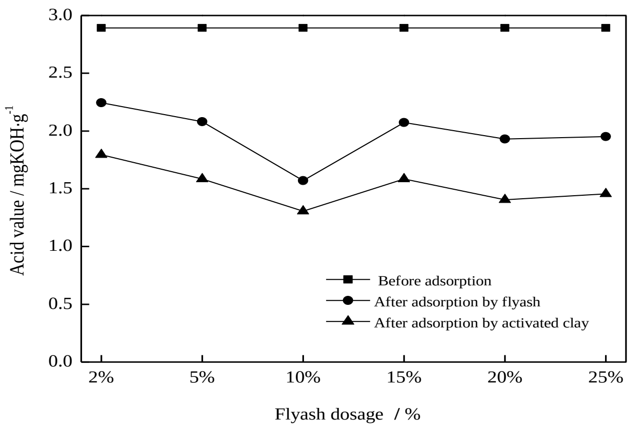


Fig. 5. Adsorption effect of fly-ash and activated clay with different adsorbent dosage

As illustrated in Fig. 5, the acid value of the WLO obviously decreases by the treatment of fly-ash and activated clay dosage. Although the adsorption effect of activated clay is superior to that of fly-ash, the difference is smaller. The maximum difference between the effects of activated clay and fly-ash is about 0.8 mgKOH/g, while the minimum is around 0.3 mgKOH/g. When adsorption time and temperature are the same, the difference between adsorption effect of fly-ash and activated clay is smaller at this dosage and the adsorption effect of fly-ash improves. In addition, with the increase dosage of adsorbent, the acid value of the WLO shows a decreasing trend when the fly-ash dosage is below 10 %. The minimum acid value of waste oil is reached at 10 % fly-ash dosage. When the fly-ash dosage is above 10 %, the acid value will go up with the improvement of fly-ash dosage. It is probably due to the amount of fly-ash dosage surpassing the optimal proportion obtained for the regeneration of WLO. Continuous dosage addition leads to high consumption of adsorbent, decreasing adsorption efficiency and desorption of adsorbate. Other problems, such as secondary pollution, difficulty in filtering and low recovery rate of WLO, are likely to happen. Thus the acid value increases.

3.2.4. Agitation rate

The effect of fly-ash and activated clay adsorption on the acid value of the WLO is indicateded in Figure 6 under the following experimental conditions: 10 % of adsorbent dosage, 60 min of adsorption time, 90 ℃ of adsorption temperature, and respectively for 200 r/min, 400 r/min, 700 r/min, 900 r/min, 1000 r/min, 1300 r/min of agitation rate.

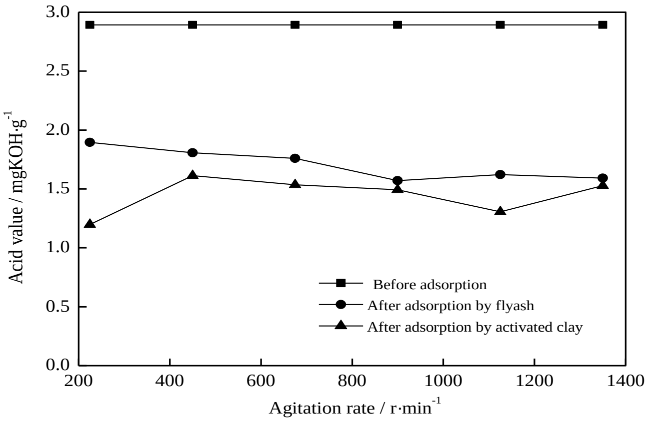


Fig. 6. Adsorption effect of fly-ash and activated clay with different agitation rate

The acid value of the WLO declines significantly by the treatment of fly-ash and activated clay at the agitation rate observed from the Fig. 6. Although the adsorption effect of activated clay is superior to that of fly-ash, the difference is not significant. The maximum difference between the effects of activated clay and fly-ash is about 0.7 mgKOH/g while the minimum is only less than 0.05 mgKOH/g. When adsorption time, temperature and adsorbent dosage keep the same, the adsorption effects of fly-ash and activated clay are nearly the same at this agitation rate and the adsorption effect of fly-ash improves. In addition, with the growth of agitation rate, the adsorption of fly-ash and activated clay both have the lowest value, respectively. This is because with the adsorption, agitation is beneficial to the dispersing of adsorbents in the waste oil, which is beneficial to the establishment of adsorption equilibrium. The insufficient agitation makes the contact between the waste oil and the adsorbent inhomogeneous, resulting in the decrease of the adsorption regeneration efficiency of the WLO, but too violent agitation may break the original adsorption equilibrium and the introduction of air in the oil to cause contact oxidation along with the desorption of adsorbate, increasing the acid value of the oil. The operation time of subsequent precipitation and refining are likely to be lengthened.

3.2.5. Multifactor experiment for the fly-ash regeneration of WLO

The multifactor experiments were performed by the $L_9(3^4)$ orthogonal experimental table to find the optimum adsorption conditions for the fly-ash regeneration of WLO, then the experimental table and results are both provided in Table 1**,** where K1, K2 and K3 are the experimental results for the various factors at the same level, while $\overline{K}1$, $\overline{K}2$ and $\overline{K}3$ represent the average value of the various experimental results for the corresponding factors at the same level. R is the range.

Table 1

Orthogonal experiment results L9(3^4)

Item Nmuber	Fly-ash: 1 Adsorption time/min		2 Adsorption temperature/ ºC		3 Fly-ash dosage / %		4 Agitation rate / r·min^{-1}		Acid value / mgKOH·g^{-1}
1	1	55	1	87	1	7	1	850	1.478
2	1	55	2	90	2	10	2	900	1.514
3	1	55	3	95	3	12	3	950	1.482
4	2	60	1	87	2	10	3	950	1.475
5	2	60	2	90	3	12	1	850	1.391
6	2	60	3	95	1	7	2	900	1.502
7	3	70	1	87	3	12	2	900	1. 463
8	3	70	2	90	1	7	3	950	1.460
9	3	70	3	95	2	10	1	850	1.479
K_1	4.474		4.416		4.44		4.438		
K_2	4.368		4.365		4.468		4.479		
K_3	4.423		4.463		4.336		4.417		
$\overline{K}_1$	1.492		1.472		1.48		1.479		
$\overline{K}_2$	1.456		1.455		1.489		1.493		
$\overline{K}_3$	1.467		1.488		1.445		1.472		
R	0.036		0.033		0.044		0.021		

As shown in Table 1, the optimum adsorption conditions for the fly-ash regeneration of WLO are as follows: 950 r/min of agitation rate, 12 % of fly-ash dosage, 90 ℃ of adsorption temperature and 60 min of adsorption time. Also, based upon the range analyses of Table 1, fly-ash dosage plays the greatest role in the fly-ash regeneration of WLO, the second is the adsorption time and temperature, the effect of agitation rate is the minimal. Therefore, in the practical operation of fly-ash regeneration for WLO, since the agitation rate has the least impact on the adsorption effect, the agitation rate can be appropriately reduced to slow down the contact oxidation caused by high-speed stirring of WLO in the adsorption regeneraton, and the power consumption of equipment can be reduced, but the adsorption treatment effect of WLO can be improved by selecting the relatively optimized amount of fly-ash added.

3.3. Comparative Experiment for the Adsorption of WLO by Fly-ash and Activated Clay

Based on the optimum processing parameters described above, comparative experiment was conducted for the adsorption of WLO by fly-ash and activated clay. The comparison on the adsorption effect was made from the following aspects, including acid value, kinematic viscosity, mechanical impurity and water content of the adsorbed WLO. The experimental results are provided in Table 2.

Table 2

Adsorption comparison of WLO by fly-ash and activated clay

Item	Acid value /mgKOH·g^{-1}	Mechanical impurity/ %	Water content / ug·g^{-1}	Kinematic viscosity / mm^2·s^{-1}
Original WLO	2.892	2.38	1518	63
WLO after adsorption by Fly-ash	1.382	2.12	1320	56
WLO after adsorption by activated clay	1.359	1.98	1194	52

As indicated in Table 2, after the WLO was absorbed separately by fly-ash and activated clay, the values of the various parameters decline significantly after the measurement. Also, from the perspective of the relevant parameters of the WLO after the adsorption, there is no big difference between the fly-ash and the activated clay regarding the adsorption effect. However, as activated clay is a solid adsorbent that must go through the various activation processes, including pre-heating, grinding, sulfuric acid activation, neutralization, washing and fine grinding, it's of high cost. But compared with the activated clay, fly-ash that is a solid waste from power plant has obvious advantage in cost. Also, it can further improve the adsorption effect after some modifications, including microwaving and ultrasonication as well as the acid and alkali treatment.

3.4. Relative Wettability of Fly-ash and Activated Clay to Liquid Paraffin Oil

When the solid contacts the liquid, the degree that the free energy of the system (the solid and the liquid) reduces is called wettability. The contact angle is an important measure of the wettability of the solid and the liquid, and is denoted by θ. The smaller the value of θ, the better the wettability of the solid and the corresponding liquid is; otherwise, the worse. $\theta = 90°$ can be defined as boundary between wetting and non-wetting. The boundary can be wetted at $\theta < 90°$ while is not wetted when $\theta > 90°$ [30]. The wettability of solid surface in solid-liquid contact can reflect the surface energy and microstructure. The greater the contact angle of the liquid on the solid surface, the worse the affinity of the solid is to the liquid, the less likely it is to be wetted by the liquid, the smaller the surface energy of the solid is, and the smaller the adhesion is [31]. Therefore, in order to compare the sensitivity of the adsorbent to the LO, the non-polar liquid paraffin was selected as the wetting liquid to investigate the relative wettability of the fly ash and activated clay.

Under the condition that the addition of sample powders to water was considered as perfect wettability. JF99A powder contact angle tester was used to obtain the corresponding system parameter β. Then the relative wetting contact angles between oil and sample powders were measured on the basis of β under the same experimental conditions. The experimental results are indicated in Table 3.

Table 3

Relative wettability experiments of fly-ash and activated clay to liquid paraffin oil

Item	β	Cosθ	θ°
the relative wettability of activated clay to liquid paraffin oil	77.71	0.70	45.46
the relative wettability of fly-ash to liquid paraffin oil	242.05	0.15	81.48

As found in Table 3, the contact angle of the fly-ash to the liquid paraffin oil $\theta_{fly\text{-}ash}$ = 81.48° while the contact angle of activated clay to the liquid paraffin oil $\theta_{activated\ clay}$ = 45.46°. Both of them are less than 90°, indicating that these two adsorbents and the liquid paraffin are wettable. $\theta_{activated\ clayx}<\theta_{fly\text{-}ash}$ indicates that compared with raw fly-ash, activated clay has better wettability and affinity to the non-polar liquid paraffin oil, that is to say, activated clay has more micro surface-active structure and higher surface energy than raw fly-ash. It also explains that activated clay in 3.2 section has a better absorption influence in treating WLO than raw fly-ash. Therefore, the physical and chemical modification of raw fly-ash, such as mechanical grinding, microwave, ultrasonic, high temperature roasting, acid and alkali salt, can be used to enhance the wettability and affinity of fly-ash to the LO so as to enhance the adsorption effect of fly-ash on the WLO.

4. Conclusions

(1) The XRD and SEM analysis proves that fly-ash is rich in the active constituents of Si and Al, which accounts for more than 90 %. Meanwhile the uneven surface and the numerous pores have constituted the material and structural basis for the excellent adsorption property.

(2) Both of the single factor and the multifactor experiments were performed to obtain the optimum adsorption conditions for the fly-ash regeneration of WLO : the fly-ash dosage of 12 %, the adsorption time of 60 min, the adsorption temperature of 90 ℃ and the agitation rate of 950 r/min.

(3) The comparative experiments by fly-ash and activated clay indicate that fly-ash has good adsorption properties in treating WLO with the effect equivalent to that by activated clay. However, fly-ash has an obvious advantage in cost. Therefore, the regeneration of WLO by fly-ash adsorption has a broad application prospect.

(4) The experiments for the relative wettability of fly-ash and activated clay to liquid paraffin oil reveal the result of $\theta_{activated\ clay} < \theta_{fly\text{-}ash}$, which menas that compared with raw fly-ash, activated clay has better wettability and affinity to the

non-polar liquid paraffin oil. And the physical and chemical modification of raw fly-ash can be used to enhance the wettability and affinity of fly-ash to oil, so as to improve the adsorption effect of fly-ash on the WLO.

Acknowledgements

This work was supporteded by the Technology Innovation and Application Demonstration Project (cstc2018jscx-msybX0339) from Chongqing Science and Technology Burear, Chongqing Education Commission Project (KJQN201800822, KJZD-K201800801 and KJZD-M201900802).

R E F E R E N C E S

[1] *G. S. Wu, M. X. Tong, S. Yao,* "Discussion on the construction and management technology of lubricating oil application", Lubricating Oil, **vol. 34**, no.1, 2019, pp. 55-59.

[2] *L. Q. Ding, M. G. Li, L. L. Niang*, "Research progress on preparation of lubricant additives from vegitable oils", Acta Petrolei Sinca(Petroleum Processing Section), **vol. 35**, no.2, 2019, pp. 414-420.

[3] *N. W. Gao, W. Ke, Y. Q. Fan,* "Regeneration of wasting lubricating oil", Chemical Industry and Engineering Progress, **vol. 28**, no.2, 2009, pp. 414-418. (in Chinese)

[4] *J. L. Dai*, "Regeneration of waste lubricating oil (4th edition)", Beijing: China. Petrochemical. Press, 2009, pp. 1-15.

[5] *R. R. Mohammed, A. R. Inanaami, H. Tahaa,* "Waste lubricating oil treatment by extraction and adsorption", Chemical Engineering Journal, **vol. 220**, no.6, 2013, pp. 343-351.

[6] *N. M. Abdel-jabbar, H. A. Zubadidye, M. Mehrvar,* "Waste lubricationg oil treatment by adsorption process using different adsorbents", World Academy of Science, Engineering & Technology, **vol. 4**, no.2, 2010, pp. 9-12.

[7] *Y. L. Hsu, C. C. Liu,* "Evaluation and selection of regeneration of waste lubricating oil technology", Environmental Monitoring & Assessment, **vol. 176**, no. (1-4), 2011, pp. 197-212.

[8] *X. M. Zhang, X. P. Yang, Y. P. Ou,* "Research progress in regeneration of used lubricaing oil with flocculation", Modern Chemical Industry, **vol. 34**, no. 1, 2014, pp. 48-51. (in Chinese)

[9] *S. P. Liu, Q. Gao, X. L. Hu, et al,* "Development in oil-adsorptin materials", Journal of Wuhan Institute of Chemical Technolog, **vol. 35**, no. 12, 2013, pp. 27-34.

[10] *Y. Su, H. F. Fang,* "Research on fly ash policy in China", Clean Coal Technology, **vol. 22**, no. 4, 2016, pp. 52-55.

[11] *X. J. Ren, X. B. Zhang, X. Q. Liu, et al,* "Preparation of porous ceramic membranes from fly ash", Journal of Materials Science & Engineering, **vol. 24**, no. 4, 2006, pp. 484-488. (in Chinese)

[12] *W. Q. Li, J. P. Zhai, Y. C. Xu, et al,* "Study on the characteristics and genetic mechanism of microspheriods in CPFA", Environmental Engineering, **vol. 15**, no.4, 1997, pp. 51-56.

[13] *Z. M. Bai, J. Yang, H. Diing,* "Research progress on silicate solid wastes I-current developments in application of flyash", Journal of Chinese Ceramic Society, **vol. 35**, no. 1, 2007, pp. 172-179.

[14] *X. G. Zhao, Z. N. Liu, Y. Liu*, "Preparation of coal flyash based forming adsorbent and its adsorptive properties for methylene blue", Journal of Chinese Ceramic Society, **vol. 37**, no. 10, 2009, pp. 1683-1688.

[15] *H. S. Shi, M. Xia, X. L. Guo,* "Research development on mechanism of flyAsh-based

geopolymer and effect of each component", Journal of Chinese Ceramic Society, **vol. 41**, no.7, 2013, pp. 972-980.
[16] *Z. D. Wang, M. L. Zhang,* "The physical chemistry explanation of the capillary condensation and the circuit of adsorption-desorption", Xinjiang Petroleum Geology, **vol. 23**, no. 3, 2002, pp. 233-235. (in Chinese)
[17] *Z. P. Xu, Q. C. Liu, C. L. Yao,* "Oxidation capability of modified fly ash based adsorbent for mercury in coal-fired flue gas", Journal of Chongqing University, **vol. 35**, no. 11, 2012, pp. 81-85.
[18] *C. F. Cai, J. P. Xu, G. W. Tang, et al,* "Study on photocatalytic activities of fly ash in the mine power plant", China Coal Society, **vol. 31**, no. 2, 2006, pp. 227-231.
[19] *X. Y. Wang, J. Li, Y. L. Wang,* "Study on modified fly-ash for the removal of acid mist waste gas", China Journal of Environmental Engineering, **vol. 2**, no. 5, 2008, pp. 675-678.
[20] *Q. J. Mao, C. X. Yu, K. Y. Ge, et al*, "Microwave absorption characteristic of surface treated fly-ash cenosphere particles", Journal of Tsinghua University, **vol. 45**, no. 12, 2005, pp. 1672-1675.
[21] *Y. S. Dai, C. H. Lu, Y. R. Ni*, "Research on cement-based fly ash for wave-absorbing properties", Journal of Wuhan University of Technology, **vol. 31**, no. 20, 2009, pp. 30-33.
[22] *L. Yang, F. Cai,* "Study of Coal Ash utilization in waste lubricant regeneration", Comprehensive Utilization of Coal Ash, **vol. 6**, no. 2, 1997, pp. 19-22. (in Chinese)
[23] *W. Wang, B. P. Tian, S. P. Wei,* "Research of basic physical and mechanics properties of fly-ash", Journal of Sichuan University of Science and Engineering (Natural Science Edition), **vol. 24**, no. 1, 2011, pp. 121-124.
[24] *H. T. Yuan, Y. Liu, L. An,* "The preparation of modified fly ash adsorbent and adsorption study on the petroleum hydrocarbon", Shandong Chemical Industry, **vol. 47**. no. 10, 2018, pp. 180-183. (in Chinese)
[25] *N. Y. Peng, P. Yu, Y. B. Luo, et al,* "Research on a new kind of spherical adsorbent for the regeneration of transformer oil in the daya wann nuclear power station, High Voltage Enigeering, **vol. 27**, no. 3, 2001, pp. 55-57. (in Chinese)
[26] *Z. Li, M. H. Wang, Y. H. Qian, et al,* "Experimental study on the regeneration of deteriorated fire-resistant oil by flyash from power plant", Thermal Power Generation, **vol. 10**, no. 5, 2001, pp. 65-66. (in Chinese)
[27] "Standard test method for acid value of petroleum products by potentiometric titration", Chinese National Standard GB/T 7304, 2014, pp. 1-19. (in Chinese)
[28] *P. Ouyang, H. Y. Fan, X. M. Zhang,* "Research progress of the modification mechanism of flyash based on adsorption", Journal of Material Science & Engineering, **vol. 32**, no. 4, 2014, pp. 619-624.
[29] *B. J. Wang, M. Li, Q. Y. Zhao,* "Relationship between surface potential and functional groups of coals", Journal of Chemical Industry & Engineering, **vol. 55**, no. 8, 2004, pp. 1329-1334.
[30] *Z. N. Xu, F. C. Lv, H. T. Zhang,* "Influencing factors of silicone rubber static contact angle measurement", High Voltage Engineering, **vol. 38**, no. 1, 2012, pp. 147-156.
[31] *X. B. Li, Y. Liu,* "Contact angle model and wettability on the surfaces with microstructures", Materials Review: Research Papers, **vol. 23**, no. 12, 2009, pp. 101-103.