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Innovative Method for Sludge Oil Extraction and Characterization Using Nano Electrodes

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Petroleum industry generated significant amounts of oily sludge, which considered as oil wastes. Due to the increment generation of the petroleum industry wastes around the world and their hazardous nature, the treatment of oil wastes attracted widespread attention. The aim of the present work is using an innovative method for sludge oil extraction and characterization using a designed nano electrodes and ultrasonic method. Four ultrasonic powers of 25, 50, 75 and 100 W used with five test periods of; 2, 4, 6, 8, 10 and 60 min at sludge/water weight ratios of; 0.125, 0.25, 0.5, 0.75 and 1.0 and test temperature ranges from 25 to 100 ºC. The recovered oil, the recovered oil salt, wastewater hydrocarbons and wastewater salts obtained at various test conditions. Two heating conditions are used, the first through ultrasonic method and the other through conventional heating of electric furnace. The increased heating temperature enhances the oily sludge extraction. The higher ultrasonic power rapidly increases the oily sludge extraction and the sludge temperature.

Keywords: Ultrasonic method, Nano electrodes, Recovered oil, Recovered oil salt, Wastewater hydrocarbons, Waste water salt.

INTRODUCTION

From the environmental point of view, disposal of industrial waste are very important [1]. Roozbehani *et al.* [2] and Hemmingsen *et al.* [3] reported that environmental pollution affects the search and technological developments. Eguavoen *et al.* [4] and Larsson [5] stated that the discharge of polluting sludge from industrial sources is a real problem in several countries.

Tkachenko *et al*. [6] considered lipid as a source of biodiesel, while Abo-Dief *et al.* [7] found that non-used corn oil produces bio-diesel with an efficiency reaching 98.5 %. Bharathi *et al.* [8] explained that biodiesel has become more attractive because of its environmental benefits. Similarly, Abo-Dief and Mohamed [9] reported the biodiesel weight % from six fresh vegetable oils. Abo-Dief *et al.* [10] also discussed the effect of the disposed sour oil on the corrosion of oil pipelines. Jamaly *et al.* [11] used contaminants such as oil, fats, grease, and inorganics for oily wastewater treatment.

Petroleum oily sludge contains hazardous waste, high concentration of toxic components and required proper management, [12-14]. Abo-Dief *et al.* [15] deduced that 1 gallon of waste oil could foul a million gallons of drinking water, similarly the presence of H_2S in crude oil is one of the most frequent contaminants at hazardous-waste sites [16]. In addition, Chorom *et al.* [17] and El Naggare *et al.* [18] proved that the oil industry has large effect on air, soil, water and human health.

Orabi *et al.* [19] used Schiff base (SH) and Schiff/nano Ni composites of different percentages to adsorb H_2S and oil disposals. Hua *et al.* [20] concluded that pump failures, pipeline ruptures and oil draining from tanks and operation units lead to oil sludge accumulation. Some researchers [21,22] evaluated the effect of fabricated iron oxide/activated carbon composite adsorption of sludge sulphur compounds. Orabi *et al.* [23] treated automotive engine oil using a microwave-induced pyrolysis process. Islam [24] found that oily sludge has 40 % recoverable oil. However, Kumar and Raj [25] found that oily sludge contained 55.13% water, 23.19% of light hydrocarbons with smaller amounts of sediments, asphaltenes and wax. Revellame *et al.* [26] showed that transesterification of dried sludge is better than that of wet activated sludge.

Sludge recovery through various methods, including solvent extraction, centrifugation, pyrolysis, microwave heating and ultrasonic irradiation [27]. Gozan [28] used low temperature (40-70 ºC) for extracting oil sludge. Jadoon *et al.* [29] separated the sludge using centrifuge machine. Li *et al.* [30] used ultrasonic, freeze/thaw and the co-pyrolysis methods for the oily sludge extraction with wood waste.

Mohamed [31] and Mohamed & Mohamed [32] studied the effect of the higher ultrasonic frequency on fretting wear behavior of 1100 aluminium, copper and austenitic stainless steel and the fatigue of aluminium and brass. Shibata *et al.* [33] enhanced the microbubble froth separation by ultrasonic irradiation for colloidal slurry. Almasi *et al.* [34] digested the anaerobic of oil refinery sludge using the ultrasound method. Xu *et al.* [35] concluded that at 40 ºC, 0.10 MPa and 28 kHz are the optimal conditions for oil recovery from sludge. Zhang *et al.* [36] recovered 80% oil using ultrasonic probe system.

EXPERIMENTAL

Oily sludge collected from oil refinery plant in eastern Egypt. The oily sludge characteristics are listed in Table-1.

TABLE-1
SLUDGE CHARACTERISTICS

Variable	Result
Density	0.9514
Specific gravity	0.9505
API gravity	17.37
Kinematic viscosity (cSt)	257.87
Total acid number (mg KOH/g)	00.33
Nitrogen content (wt. %)	00.04
Pour point (°C)	00.00
Flash point (°C)	45.00
Water content (vol. %)	08.00
Ash content (wt. %)	00.13
Carbon residue (wt. %)	09.49
Salt content (Ptb)	150.00

The ultrasonic generator controlled ultrasonic power device (Jio-technic U10) and test period is used in the present study. Conventional heating for the oily sludge carried out using a heating furnace at various test parameters. The ultrasonic power, test period, sludge/water (S/W) ratio and the test temperature effects on oil recovery and salt contents were investigated. Four test temperatures 25, 50, 75 and 100 ºC selected in the ultrasonic tests. The effect of both ultrasonic and furnace oily sludge treatment investigated.

Nano-carbon electrode: A nano-carbon electrode manufactured in the lab to calculate the salt contents in both recovered oil and wastewater. A nano-carbon electrode used to reduce background noise while maintaining large sampling volume. Nano carbon-electrode arrays used to detect down to 0.25 ng/mL.

Procedure: The heated sludge/water mixture contained three layers, recovered oil on the top, wastewater in the middle and sediments in the bottom. The concentrations of recovered oil separated using a separator funnel. The wastewater analyzed using the nano carbon electrodes. The recovered oil hydrocarbon concentration dissolved in 15 mL cyclohexane, shaken at 150 rpm for 1 h. Then, packed with silica gel and anhydrous sodium sulfate for cleanup, collected and evaporated to remove the solvent and concentrate the hydrocarbons. Wastewater extracted using cyclohexane (volume ratio of 8:3), with the remaining procedures being similar to those used for measuring the hydrocarbons in the oil layer sample.

RESULTS AND DISCUSSION

Effect of ultrasonic power on temperature: Fig. 1 illustrates the effect of ultrasonic power on the test temperature at various test periods of the sludge/water slurry. As the ultrasonic power increases, the sludge/water slurry temperature increases up to 10 min test period, after that all the power trends show a very slight increase. The water layer was not clear indicated that it might contain unwanted impurities such as fine solids and dispersed petroleum hydrocarbons, which requires proper treatment before discharging into the environment.

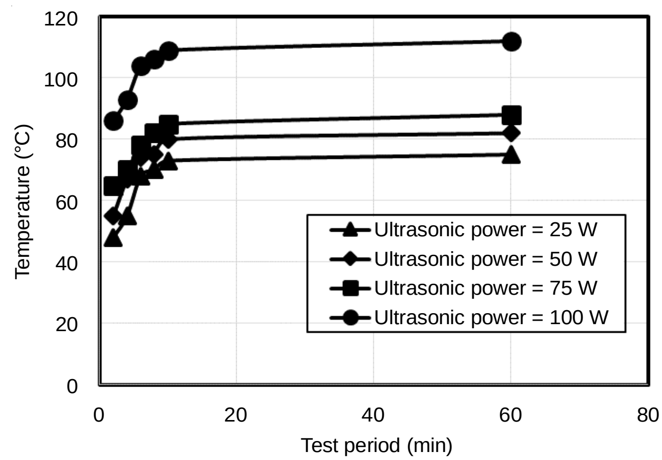

Fig. 1. Effect of ultrasonic power and test period on temperature

Effect of test temperature on sludge products: Temperature increment reduces the viscosity and accelerates the water separation from oil. Fig. 2a illustrates the variation of recovered oil % at various tested temperatures and ultrasonic power. The oil recovery % increased with both temperature and ultrasonic power increment with a peak of 73 % at 75 ºC. As the ultrasonic power increases, the percentage of oil recovery increases. Also, the furnace heating sludge has the lowest percentage of oil recovery. Fig. 2b shows the variation of recovered oil salts with tested temperatures at various ultrasonic powers and furnace heating. It is clear that as the test temperature increases, the recovered oil salt content decreases in all conditions and ultrasonic powers. Therefore, the temperature increment effect leads to oil recovery increment and oil recovery salt decrement. The furnace-heating trend found higher. Also, the ultrasonic power increment leads to the recovered oil salt decrement. Figs. 2c and 2d showed the variation of both waste oil hydrocarbons and waste oil salts with test temperature at various ultrasonic powers. In Fig. 2c, it is clear that as ultrasonic power increases, waste water hydrocarbons decreases because most of the hydrocarbons coupled with the recovered oil. While the wastewater salt has reversible trends, as the ultrasonic power increases, the wastewater salts increases. In both cases, the furnace heating trends found higher compared to the ultrasonic trends. At higher temperatures, the ultrasonic oil recovery rate becomes greater compared to the furnace oil recovery rate, which attributed to its cavitation effect [12,28,33]. Due to insufficient disintegration of the wastewater activated oil sludge structure flow, furnace heating is insufficient. During the ultrasonic heating, the generated hydro-mechanical shear forces caused disturbances

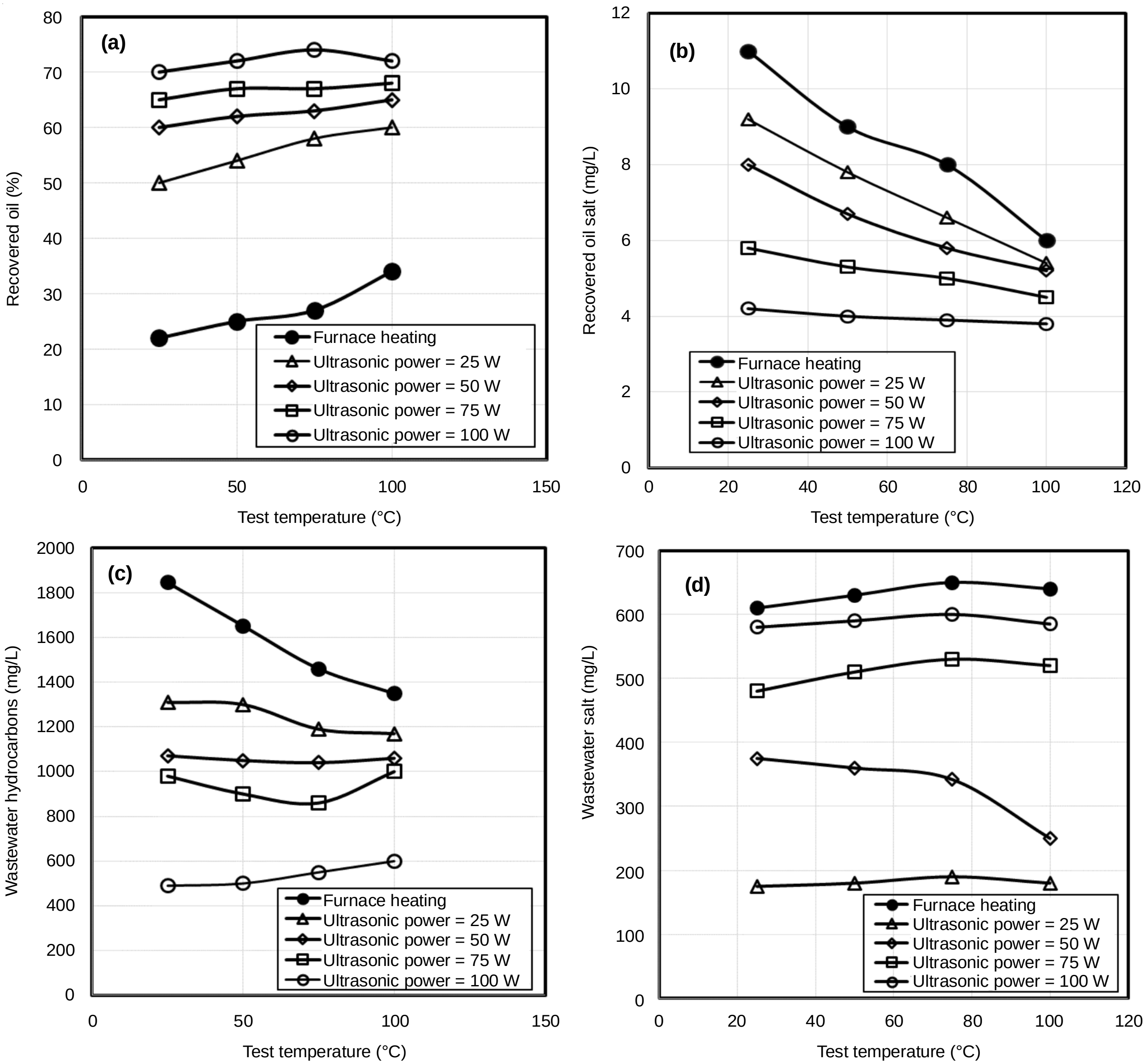

Fig. 2. Effect of test temperature on recovered oil percentage (a), recovered oil salt (b), wastewater hydrocarbons (c) and wastewater salt (d) at various ultrasonic powers

in the liquid sludge and introduced a drag effect on its droplets. So, the liquid sludge small droplets moved faster than the larger droplets which increases the collision frequency, leading to an increased chance of forming larger water droplets, which would then enhance the separation of water and oil. Fig. 2b showed that the recovered oil salt content decreased with increasing treatment duration and ultrasonic power which became more significant with 25 W ultrasonic power. With increasing the tested temperatures from 25 to 100 ºC, the salt content decreased from 11.2 to 6.3 mg/L with furnace heating. While, the salt content in recovered oil decreased from 4.2 to 3.8 mg/L at 100 W ultrasonic power at the tested temperatures.

Effect of tested period on sludge products: Fig. 3a shows the variation of the percentage of oil recovery with the tested period at various ultrasonic powers. It is clear that the percentage of oil recovery increases with test period increment up to 10 min. After 10 min, there is no percentage of oil recovery increment is observed for all ultrasonic power values. After 2 min of ultrasonic treatment, the percentage of oil recovery was 50.2, 60.1, 65.4 and 70.0 % at an ultrasonic power of 25, 50, 75 and 100 W, respectively. After 10 min, the percentage of oil recovery nearly reached its maximum values of 90.1, 80.2, 73.2 and 60.4 % at an ultrasonic power of 100, 75, 50 and 25 W, respectively. For all ultrasonic power values, Fig. 3b showed that as tested period increases, the recovered oil salt values increases up to 4 min, followed by rapid decrement up to 10 min. After 10 min, recovered oil salt values have a very slight decrement. Higher demulsification efficiencies of oily sludge lead to less salt content in the separated oil phase (Fig. 3c) and higher salt content in the separated wastewater (Fig. 3d).

Effect of sludge/water ratio: Fig. 4a showed that as the sludge/water (S/W) ratio increases, percentage of recovered

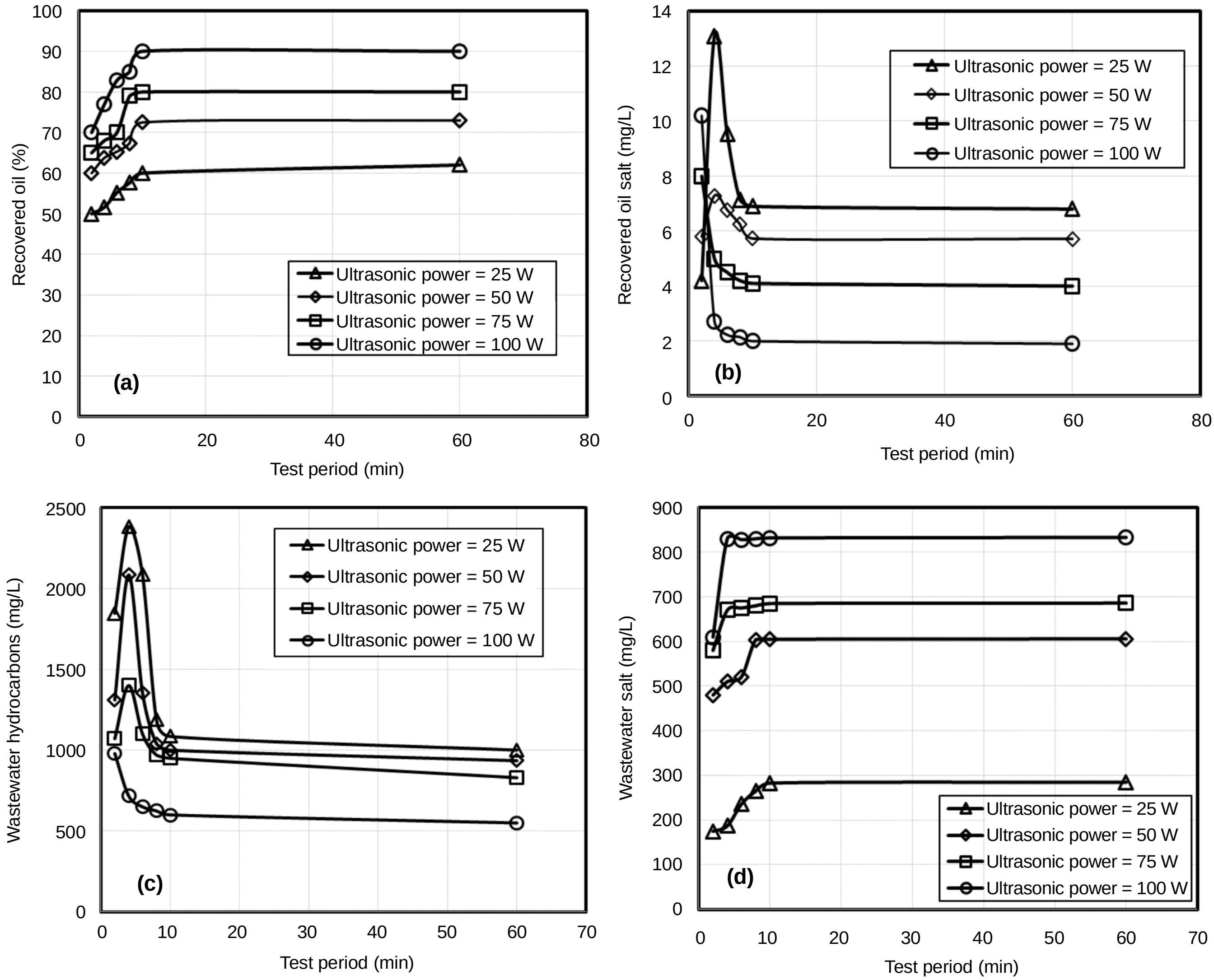

Fig. 3. Effect of test period on recovered oil percentage (a), recovered oil salt (b), wastewater hydrocarbons (c) and wastewater salt (d) at various ultrasonic powers

oil decreases at 75 and 100 W, while at ultrasonic powers of 25 and 50 W, recovered oil percentage trends increases up to 0.25 sludge/water ratio followed by a decrement at all other sludge/ water values. As the ultrasonic power increases, the percentage of recovered oil increases for all sludge/water ratios. A reverse effect is shown in Fig. 4b, where the recovered oil salt amounts increases at all ultrasonic power values at sludge/water ranges from 0.25 to 1.00. The recovered oil salt amounts trend of 25 W ultrasonic power is shown higher followed by 50, 75 and 100 W, respectively. Fig. 4c illustrated that as sludge/water increases, the wastewater hydrocarbons amounts decreases for all ultrasonic power values up to S/W = 0.25 except the ultrasonic power of 25 W. After S/W = 0.25, the waste water hydrocarbons amounts increases for all ultrasonic power values, where 25 W trend found higher followed by 50, 75 and 100 W, respectively. Low water content resulted in the oily sludge viscosity increment leading to the weakened sonication effect on oil recovery and desalting that resulted in an increment of waste salt hydrocarbons, Fig. 4c. and increment of waste water salts with ultrasonic power increment (Fig. 4d). High water content in the sludge reduces the ultrasonic irradiation treatment effect. So, the sludge to water ratio of 0.125 selected as the optimal value for oily sludge treatment. So, recovered oil %, recovered oil salt, waste water hydrocarbons and wastewater salt amounts decreases for all ultrasonic power values.

Conclusions:

- Oily sludge proven to be a new source of oil. Almost 90 % of oily sludge recovered by ultrasonic method.
- The increased heating temperature enhances the oily sludge extraction.
- The higher ultrasonic power rapidly increases the oily sludge extraction and the sludge temperature.
- Investigations on the effects of the ultrasonic power showed that it has an important influence on oil extraction.
- The designed and fabricated nano-carbon electrode computed the salt contents successfully.
- As the test temperature increases, both the recovered oil % and the salt contents in the waste water increases with ultrasonic power increment, while recovered oil salts and waste water hydrocarbons decreases with ultrasonic power decrement. The maximum recovered oil % of 73 % acheived at 75 ºC.

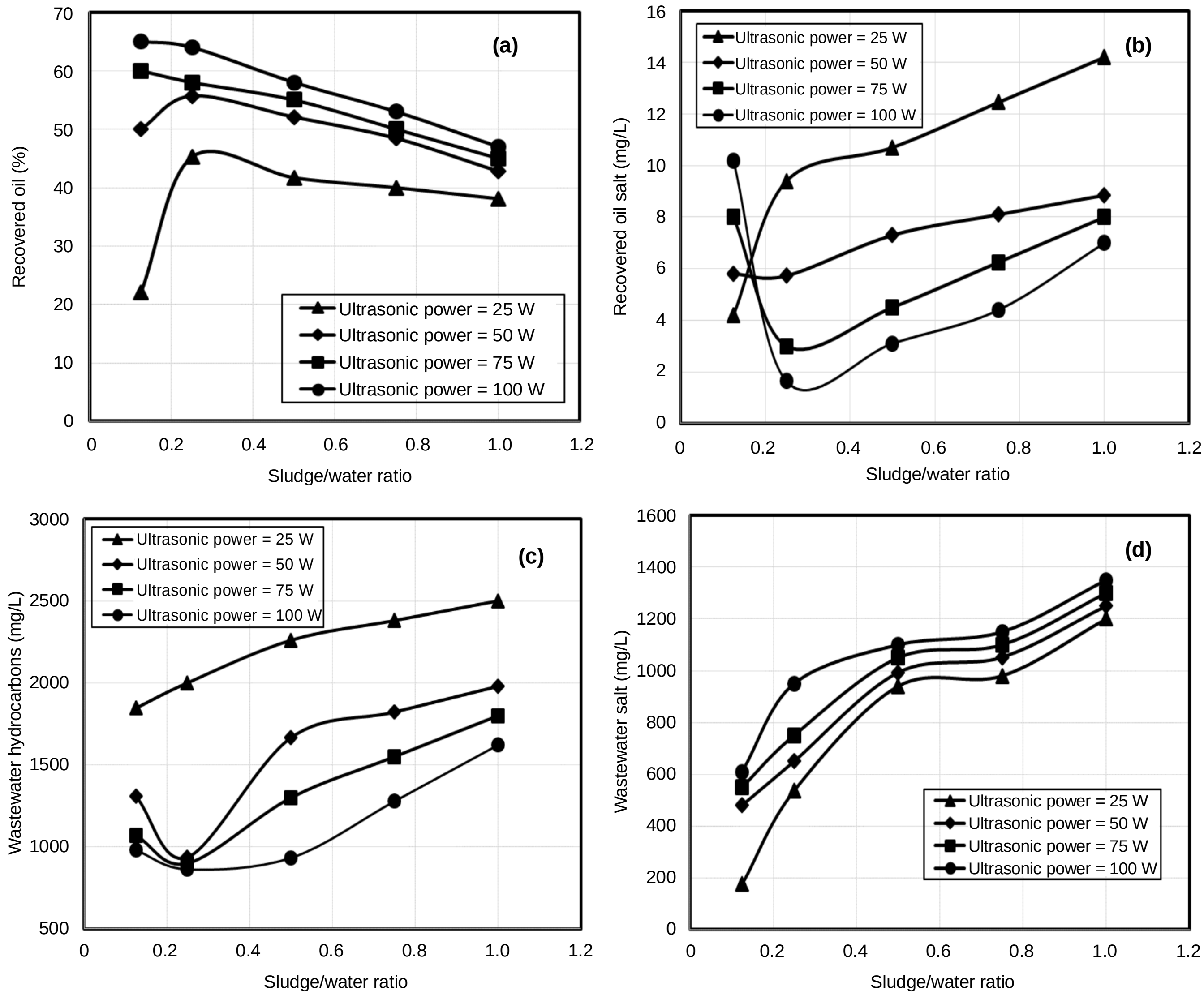

Fig. 4. Effect of sludge/water ratio on recovered oil percentage (a), recovered oil salt (b), wastewater hydrocarbons (c) and wastewater salt (d) at various ultrasonic powers

- As the tested period increases, both the recovered oil % and the salt contents in waste water increases with ultrasonic power increment, while recovered oil salts and wastewater hydrocarbons decreases with ultrasonic power decrement. The treatment duration extended from two to 10 min and then it approached an asymptote, which indicates that further extension of treatment duration is unnecessary. The maximum recovered oil percentage is 90.1 % at 10 min.
- As the sludge/water ratio increases, both the recovered oil % and the salt contents in the waste water increases with ultrasonic power increment, while recovered oil salts and waste water hydrocarbons decreases with ultrasonic power decrement. The maximum recovered oil percentage of 66.2 % at sludge/water ratio of 0.125:1.0.

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CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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