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CHEMICAL ENGINEERING

DISCOVERING THOUGHTS AND INVENTING FUTURE

HEHLIGHTS

Diesel Engine Emission

Fixed Bed Column Study

Removal of Barium, Zinc

Aquatic Environment by NCRH

The Noble Gases



Issue 3





GLOBAL JOURNAL OF RESEARCHES IN ENGINEERING: C Chemical Engineering

GLOBAL JOURNAL OF RESEARCHES IN ENGINEERING: A Chemical Engineering

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Fixed Bed Column Study for the Removal of Copper from Aquatic Environment by NCRH

By Upendra Kumar & Jyotikusum Acharya

National Institute of Technology, Silchar Assam, India

Abstract - This paper reports the results of the study on the performance of low-cost adsorbent such as NCRH in removing copper. The adsorbent materials adopted were found to be an efficient media for the removal of heavy metals in continuous mode using fixed bed column. The fixed bed column experiment was conducted in a column having a diameter of 2 cm with 10 mg/l Cu(II) solution at a bed depth of 10 cm maintaining a constant flow rate of 10 ml/min. The breakthrough and exhaust time were found to be 3.583 and 10.500 h, respectively. Height of adsorption zone was found to be 10.21 cm and the rate at which the adsorption zone was moving through the bed was 1.48 cm/h. The percentage of the total column saturated at breakthrough was found 44.95%. The value of adsorption rate coefficient (K) and adsorption capacity coefficient (N) were obtained as 0.056 l/(mg h) and 1623 mg/l, respectively.

Keywords : copper, NCRH, breakthrough curve, adsorption rate coefficient, adsorption capacity coefficient.

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FIXED BED COLUMN STUDY FOR THE REMOVAL OF COPPER FROM ADUATIC ENVIRONMENT BY NCRH

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Fixed Bed Column Study for the Removal of Copper from Aquatic Environment by NCRH

Upendra Kumar & Jyotikusum Acharya

Abstract - This paper reports the results of the study on the performance of low-cost adsorbent such as NCRH in removing copper. The adsorbent materials adopted were found to be an efficient media for the removal of heavy metals in continuous mode using fixed bed column. The fixed bed column experiment was conducted in a column having a diameter of 2 cm with 10 mg/l Cu(II) solution at a bed depth of 10 cm maintaining a constant flow rate of 10 ml/min. The breakthrough and exhaust time were found to be 3.583 and 10.500 h, respectively. Height of adsorption zone was found to be 10.21 cm and the rate at which the adsorption zone was moving through the bed was 1.48 cm/h. The percentage of the total column saturated at breakthrough was found 44.95%. The value of adsorption rate coefficient (K) and adsorption capacity coefficient (N) were obtained as 0.056 l/(mg h) and 1623 mg/l, respectively.

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I. INTRODUCTION

ater pollution due to toxic heavy metals has been a major cause of concern. The industrial and domestic wastewater is responsible for causing several damages to the environment and adversely affecting the health of the people. Metals can be distinguished from other toxic pollutants, since they are nonbiodegradable and can accumulate in living tissues, thus becoming concentrated throughout the food chain¹. As one of the important toxic heavy metals, copper finds its way to the water stream from industries like electroplating, mining, electrical and electronics, iron and steel production, the non-ferrous metal industry, the printing and photographic industries and metalworking and finishing processes ^{2, 3}. Moreover, copper sulfate has been used in eutrophic lakes to kill the algae since the early 1990s and is still widely used today. Trace amounts of copper are essential to human and many other living species. However, the intake of excessively large doses of Cu(II) by human may lead to severe mucosal irritation, a central nervous system irritation, possible necrotic changes in the liver and kidney, etc., recommended maximum and the acceptable concentration of Cu(II) in drinking water by the World Health Organization(WHO) is 1.5 mg/L. As per U.S. Environmental Protection Agency (EPA) maximum acceptable concentration in drinking water is 1.3 mg/L⁴.

Various treatment techniques have been employed to eliminate or reduce heavy metals in wastewater including precipitation, adsorption, ion exchange and reverse osmosis. As of now, adsorption by activated carbon is accepted to be the best available technology for the reduction of heavy metals, except that its manufacturing cost is quite high. Hence, a search is on worldwide for a low-cost alternative. Research in the recent years has indicated that some natural biomaterials including agricultural products and by-products can accumulate high concentration of heavy metals. Adsorbent generated from these biomass are cost effective and efficient. Low-cost agricultural products and by-products have been reported to be effective in removing heavy metals⁵. Adsorption process of heavy metals present in aqueous solution by low-cost adsorbents from plant wastes can be carried out with or without chemical modifications. In general, chemically modified plant wastes exhibit higher adsorption capacities than unmodified forms ⁶.

In our continued study on the use of lowcost material for the removal of organic and organic pollutants from water and wastewater we investigated rice husk as a sorbent for the removal of Cu(II). Some simple and low-cost chemical modifications resulted in increasing the sorption capacity of raw rice husk. The highly efficient low cost and the rapid uptake of Cd(II) by NCRH indicated that it could be an excellent alternative for the removal of heavy metal by sorption process ^{1,7}.

Agricultural products and by-products have been reported to be effective in removing copper. Rice husk consists of cellulose (32.24%), hemicelluloses (21.34%), lignin (21.44%) and mineral ash (15.05%) as well as high percentage of silica in its mineral ash, which is approximately 96.34%. Rice husk is insoluble in water, has good chemical stability, has high mechanical strength and possesses a granular structure, making it a good adsorbent material for treating heavy metals from wastewater ⁶. Rice husk, an abundant agricultural product, is capable of removing heavy metals and can be considered as an efficient and low-cost adsorbent for heavy metals. The present study has been undertaken to report the Cu(II) adsorption in the fixed bed column process. In recent years, attention has been taken on the utilization of unmodified or modified rice husk as a sorbent for the removal of pollutants ^{8, 9, 10}.

II. MATERIALS AND METHODS

a) Equipments and Chemicals

All chemicals used were of analytical grade (BDH, India). Stock solutions of 100 mg/l were prepared

using metal nitrate salts, which were diluted with distilled water to prepare working solutions. Cyberscan 510 model pH meter was used for the measurement of pH of the solution. A peristaltic pump (Miclins India Limited, PP 30) was also used for providing constant flow of metal and desorbing solution in fixed bed column. The metal ion concentrations in the solution were analyzed using atomic absorption spectrophotometer (AA-6650, Shimadzu).

b) Preparation of adsorbent

Fresh rice husk was obtained from a local rice mill and was passed through different sieve size. The fraction of particle between 425 and 600 micron (geometric mean size: 505 micron) was selected. The rice husk was washed thoroughly with distilled water. It was dried at 60°C. The sorbent thus obtained was designated Raw Rice Husk (RRH). Rice husk was treated with 0.1 M sodium carbonate solution at room temperature for 4 h. Excess of sodium carbonate was removed with distilled water and the material was dried at 40oC. This material was designated as sodium carbonate treated rice husk (NCRH)¹.

c) Experimental Studies

Fixed bed column study for Cu(II) removal from wastewater by NCRH was conducted using a column of 2 cm diameter and 55 cm length. The column was packed with NCRH between two supporting layers of pre-equilibrated glass wool. The bulk density of NCRH packed in the column was 0.267 g/cm³. The column bed depth was kept 10 cm. The schematic diagram of column study is shown in Fig. 1. The study was conducted at temperature of 28 \pm 2oC and the pH of the Cu(II) solution as 6.0 \pm 0.2 for the present study. The column was charged with Cu(II) bearing wastewater with a volumetric flow rate of 10 ml/min (\sim 2.10 $m^{3}/m^{2}/h$). The initial concentration of Cu(II) was 10 mg/l. The samples were collected at certain time intervals and were analyzed for Cu(II) using atomic absorption spectrophotometer (AA-6650, Shimadzu).

III. Results and Discussions

a) Behavior of adsorption column

The fixed bed column experiment was conducted with 10 mg/l Cu(II) solution at a bed depth of 10 cm maintaining a constant flow rate of 10 ml/min. The breakthrough curve of S-shaped was obtained as shown in Fig. 2. The breakthrough

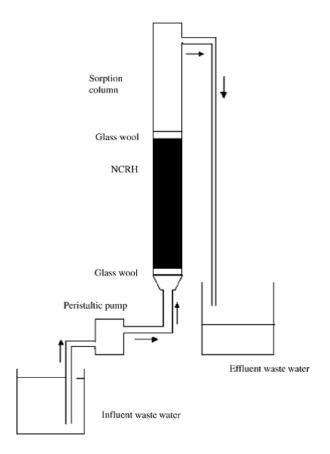


Fig. 1 : Schematic diagram fixed bed column experimental set up

time (corresponding to C/Co = 0.1) and exhaust time (corresponding to C/Co = 0.9) were found to be 3.583 and 10.500 h respectively. The corresponding volumes of wastewater treated were 2.15 and 6.30 liters respectively. About 8.39 gm of NCRH were used in the 10 cm column. The volume of metal effluent treated and the requirement of NCRH up to breakpoint have b shown in Table 1. The market price of activated carbon for industrial grade is US \$ 20-22 (Rs. 1000 - 1100) per kg depending on the quality¹¹. The cost of NCRH is in India was estimated as only Rs. 4 per Kg (US \$ 0.08). Hence, by cost comparison the cost, NCRH is about 250 times cheaper than activated carbon. Cost for volume of metal effluent treated up to breakpoint per kg of NCRH has been calculated and has been presented in Table 6.2.

The formation and movement of the adsorption zone has been described mathematically ^{12, 13}. The time required for the exchange zone to move the length of its own height up/down the column once it has become established is

$$t_z = \frac{V_E - V_B}{Q_w} \tag{1}$$

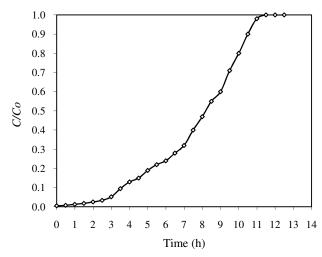


Fig. 2: Breakthrough curve for Cu(II) using NCRH.

Where,

 V_{E} = volume of wastewater treated to the point of exhaustion (I)

 $V_{\scriptscriptstyle B}$ = volume of wastewater treated to the point of breakthrough (I)

 Q_w = wastewater flow rate (I/h)

The time required for the exchange zone to become established and move completely out of the bed is

$$t_E = \frac{V_E}{Q_w} \tag{2}$$

Rate at which the exchange zone is moving up or down through the bed is

$$U_z = \frac{h_z}{t_z} = \frac{h}{t_E - t_f}$$
(3)

Where,

h = height of exchange zone (cm)

h = total bed depth (cm)

 $t_{\rm f} =$ time required for the initially form (h)

Rearranging Eq. 3 provides an expression for the height of the exchange zone as given below.

$$h_z = \frac{h(t_z)}{t_E - t_f} \tag{4}$$

The value of $t_{\rm f}$ can be calculated as follows.

$$t_f = (1 - F)t_z \tag{5}$$

At breakthrough the fraction of adsorbate present in the adsorption zone still possessing ability to remove solute is

$$F = \frac{S_{z}}{S_{\text{max}}} = \frac{\int_{V_{B}}^{V_{E}} (C_{o} - C) dV}{C_{o} (V_{E} - V_{B})}$$
(6)

Where,

 $C_0 =$ initial solute concentration (mg/l)

 S_z = amount of solute that has been removed by the adsorption zone from breakthrough to exhaustion S_{max} = amount of solute removed by the adsorption zone if completely exhausted

The percentage of the total column saturated at breakthrough is

% saturation =
$$\frac{h + (F - 1)h_z}{h} \times 100$$
 (7)

The values of the important design parameters were calculated using the data from breakthrough curve. Height of adsorption zone was found to be 10.21 cm and the rate at which the adsorption zone was moving through the bed was 1.48 cm/h. The percentage of the total column saturated at breakthrough was found to be 44.95 %.

b) Evaluation of adsorption column design parameters

Data collected during laboratory and pilot plant tests serve as the basis for the design of fullscale adsorption columns. A number of mathematical models have been developed for the use in design. In the present research work the fixed bed column was designed by logit method ^{14, 15}. The logit equation can be written as:

$$\ln\left[\frac{C/C_o}{1-C/C_o}\right] = -\frac{KN_oX}{V} + KC_ot$$
(8)

Where,

C = concentration at any time t

 $C_{o} = initial Cu(II)$ concentration (10 mg/I)

V = approach velocity (210 cm/h)

X = bed depth (10 cm)

K = adsorption rate constant (l/mg-h)

 $N_{\rm o}$ = adsorption capacity constant (mg/l)

Rearranging Eq. 8

$$\ln\left[\frac{C}{C_o - C}\right] = -\frac{KN_o X}{V} + KC_o t \tag{9}$$

Plot of ln C/(C₀ – C) vs. t gives a straight line with slope KC_o and intercept – KN₀ X /V from which K and N_o could be calculated. Plot of ln C/(C₀ – C) vs. t was shown in Fig. 3. The values of adsorption rate constant (K) and adsorption capacity constant (N_o) were obtained as 0.056 l/mg.h and 1623 mg/l (1.623 kg/m³) respectively. These values could be used for the design of adsorption columns. The adsorption capacity was found to be good. Hence, it could be concluded that NCRH is effective for Cu(II) removal.

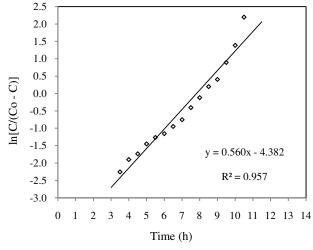


Fig. 3 : Linearized form of logit model.

IV. Conclusions

NCRH was found to be efficient media for the removal of Cu(II) from aquatic environment. The column with 2 cm diameter, and bed depth 10 cm could treat 2.15 liters of Cu(II) at breakthrough, at initial concentration 10 mg/l. About 3.90 g of NCRH was required per liter of Cu(II) treatment. Height of adsorption zone was found to be 10.21 cm and the rate at which the adsorption zone was moving through the bed was 1.48 cm/h. The values of adsorption rate constant (K) and adsorption capacity constant (N_o) were obtained as 0.056 l/mg.h and 1623 mg/l respectively.

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Table 1 : Volume of effluent treated and the mass of NCRH required up to breakthrough

Bed depth	10% Breakthrough	Treated volume	Total mass of	Mass per litre
(cm)	time (hr)	(lts)	NCRH (gm)	(gm/l)
10	3.583	2.15	8.39	3.90 gm

Table 2 : Cost for volume of metal effluent treated up to breakpoint per kg of NCRH

Metal	Volume of metal effluent treated	Cost per kg NCRH		
	per kg of NCRH	@ Rs. 4	@ US \$ 0.08	
Cu(II)	256 litres	64 litres / Rs.	3200 litres / US \$	



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Removal of Barium, Zinc and Mercury from Drill Cuttings Using Activated Palm Kernel Shell and Husk

By Ijeoma. A. Chukwu & Elijah T. Iyagba

University of Port Harcourt, Nigeria

Abstract - Palm kernel shell and Palm kernel husk have been used to remove Barium, Zinc and Mercury from drill cuttings. Batch adsorption studies were carried out as function of pH, contact time and Carbon dosage. Barium, Zinc and Mercury were found to be pH dependent with optimum pH of 9 for all activated Carbon materials. Barium and Zinc APKS was 150mins, while Barium and Zinc APKH was 120mins. For Mercury both APKS and APKH attained maximum adsorption at 60mins. For maximum adsorption, the adsorbent loading was 5g for Barium and Mercury APKS, 3g for Zinc and 4g for Barium, Zinc and mercury APKH. Although Barium and Zinc did not exceed the regulatory limit, the equilibrium experimental data were found to best fit the Freundlich Isotherm model for APKH with R2 = 99.84% for Ba, 85.66% for Zinc and 89.92% for Mercury. The intensity of adsorption for Barium was 0.9420, 0.0710 for Zinc and 0.2935 for Mercury. Although their was ion adsorption using Palm kernel shell and husk is not very favorable for the removal of Barium, Zinc and Mercury from drill cutting.

Keywords : drill cuttings, heavy metals removal, palm kernel shell, palm kernel husk, adsorption.

GJRE-C Classification : FOR Code: 030301, 030304

REMOVAL OF BARIUM, ZINC AND MERCURY FROM DRILL CUTTINGS USING ACTIVATED PALM KERNEL SHELL AND HUSK

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Removal of Barium, Zinc and Mercury from Drill Cuttings Using Activated Palm Kernel Shell and Husk

Ijeoma. A. Chukwu^a & Elijah T. Iyagba^o

Abstract - Palm kernel shell and Palm kernel husk have been used to remove Barium, Zinc and Mercury from drill cuttings. Batch adsorption studies were carried out as function of pH, contact time and Carbon dosage. Barium, Zinc and Mercury were found to be pH dependent with optimum pH of 9 for all activated Carbon materials. Barium and Zinc APKS was 150mins, while Barium and Zinc APKH was 120mins. For Mercury both APKS and APKH attained maximum adsorption at 60mins. For maximum adsorption, the adsorbent loading was 5g for Barium and Mercury APKS, 3g for Zinc and 4g for Barium, Zinc and mercury APKH. Although Barium and Zinc did not exceed the regulatory limit, the equilibrium experimental data were found to best fit the Freundlich Isotherm model for APKH with $R^2 = 99.84\%$ for Ba, 85.66% for Zinc and 89.92% for Mercury. The intensity of adsorption for Barium was 0.9420, 0.0710 for Zinc and 0.2935 for Mercury. Although their was ion adsorption of heavy metal ions at low concentration, the low intensity values below unity indicates that adsorption using Palm kernel shell and husk is not very favorable for the removal of Barium, Zinc and Mercury from drill cutting.

Keywords : drill cuttings, heavy metals removal, palm kernel shell, palm kernel husk, adsorption.

I. INTRODUCTION

n many operations of gas or oil wells, the drill cuttings give rise to an increasing problem with respect to their handling and disposal. If oil and gas exploration rigs and production installations are allowed to dump drilling wastes unchecked, the effects on marine life can be widespread (Frode and Gray, 1995, Okpokwasili and Nnubia, 1996, Wills, 2000). The ecological effects extend for several kilometers, smothering seabed life and remaining toxic for many years.

Cuttings that contain significant levels of heavy metals require special handling since repeated disposal can lead to accumulation of high molecular weight compounds. At high concentrations, these nonbiodegradable constituents can increase soil water repellency and render the land unfit without treatment or amendment (Callahan et al, 2002, Cordah, 2001). In a review Bell et. Al., 1998, underlined the dangers of allowing cuttings to accumulate. Various measures to obtain zero discharge have called for appropriate drill cutting treatments prior to disposal in order to meet standard conditions. Treatment of drill cuttings is vital because it has lots of application (Page et al., 2003, Reuben and Miebaka, 2008, Opete et al., 2010).

Living organisms require varying amounts of heavy metals, while excessive levels can be damaging to organisms. Some are dangerous to health (e.g.: Hg, Cd, As, Pb, Cr) and some cause corrosion (eg:Zn, Pb) (Sullivan, 1991, Ayotamuno et al., 2007). Numerous processes exist for removing dissolved heavy metals. The use of alternative low cost materials as potential sorbents for the removal of heavy metals is very important (Abel-Ghania and El-Chaghaby, 2007, Ademuluyi et. al., 2009). Disposal of agricultural wastes is currently a major economic and ecological issue, and the conversion of these agro-products to adsorbents such as activated carbon represents an alternative (Itodo et al., 2010).

To reduce the hazardous nature of these drill cuttings, international legislations have been imposed. The oil and gas exploration and production activities inevitably generate these drill cuttings which must be treated prior to disposal. Drill cuttings are treated thermally in Nigeria. This only takes care of hydrocarbons leaving heavy metals above regulatory limits. The heavy metal, if considerably reduced to a tolerable level reduces the hazardous effects to the environment (Okporanma and Ayotamuno, 2008).

In an earlier study, the feasibility of using activated carbon from two readily available agricultural wastes, namely; Palm Kernel Shell (PKS) and Palm Kernel Husk (PKH) to remove chromium and lead from drill cuttings was carried out (lyagba and Opete, 2009). This work is a continuation of the previous study and is aimed at the removal of Barium, Zinc and Mercury from drill cuttings, using PKS and PKH as well.

II. MATERIALS AND METHODS

a) Preparation of adsorbent

Palm kernel shell and husk were collected, washed with clean water and sun dried for 48 hours. They were crushed in mortar, sieved and carbonized at 300°C and 250°C for palm kernel shell and husk

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respectively. They were subsequently activated using concentrated $H_{3}\text{PO}_{4}$ and oven dried.

b) Preparation of heavy metal extract

Heavy metal extract from the thermally treated drill cuttings were prepared using the ASTM-D-3974 method.

c) Batch equilibrium studies

Batch equilibrium experiments were conducted by adding a known quantity of activated carbon shell and husk to 20 ml of the heavy metal extract and shaken vigorously.

d) pH

20ml of the heavy metal extract was adjusted to different pH values of 1,3,5,7 and 9 by adding 0.1M HCl or NaOH accordingly. The resulting solution at different pH levels were treated with 1.5g of APKS and APKH each and shaken. These were shaken and filtered and their concentrations determined using the Atomic Adsorption Spectrophotometer (GBC AVANTA Model).

e) Contact time

1g of the APKS and APKH each was weighed into a beaker. 20ml of the heavy metal extract was added to the activated carbon and shaken. This was allowed to stand for 30, 60, 90,120 and 150 mins and filtered. The filtrate was analyzed for Barium, Zinc and Mercury using an Atomic Adsorption Spectrophotometer

f) Carbon Dosage

Keeping the pH of the heavy metal extract constant (pH of 9) following the earlier results obtained, 1,2,3,4 and 5g of APKS and APKH were each added to 30ml of the heavy metal extract and allowed to stand for 60mins-Mercury, 120 mins-Barium and Zinc for APKH and 150 mins-Barium and Zinc for APKS. The resultant solution with the adsorbents was shaken and filtered. The Barium, Zinc and Mercury concentrations were determined spectrophotometrically.

The mass of Barium, Zinc and Mercury adsorbed were calculated using the formulae;

$$X = \left(c_i - c_f\right)V$$

where $c_{\rm i}$ and $c_{\rm f}$ are initial and final Barium, Zinc and Mercury concentrations, while V is the volume of extract used.

III. Results And Discussion

Analysis of the experimental data obtained from the batch studies were carried out. The study showed the following;

a) Effect of pH

The optimal pH of Barium, Zinc and Mercury is 9 (Figures 1,2 and 3). At these pH values, Barium removal for APKS and APKH both attained over 94%, Zinc removal for APKS and APKH were both 97% while for Mercury APKS and APKH were both 89%. At low pH, the removal of Ba²⁺, Zn²⁺ and Hg²⁺ was low. This finding is at variance with chromium and lead removal that took place optimally at 3 and 5 with APKS and APKH, respectively (lyagba and Opete, 2009). Metals have small number of electrons in excess of a stable, closedshell electronic configuration and as such have tendency to loose these extra ions to attain stability (Wikipedia). This should favour increased metal uptake at low pH. More metal removal at low pH may also be due to high proton present making metal bonding sites become positively charged thereby repelling the cations e.g. Cr³⁺ and Pb²⁺ and attracting anions such as Cl⁻ and OH -. However, oxides of Barium and Zinc are well known solid bases whereas oxides of trivalent Al³⁺, Cr³⁺ tend to be acidic (lyagba and Schutz, 2007). The result therefore shows that although neutral or highly acidic conditions are favorable for metals uptake, Ba²⁺, Zn²⁺ and Hg²⁺ uptake is enhanced in relatively strong alkaline media, as supported by Ansari and Raofie, 2006.

b) Effect of contact time

For APKS, varying the contact time of the adsorbent with the adsorbate shows a gradual increase in Barium and Zinc APKS, while there was a rapid increase for Barium and Zinc uptake for APKH. The highest adsorption for the uptake of Barium and Zinc in APKS occurred at 120 mins. These can be seen in figures 4 and 5, while the adsorption of Mercury on APKS and APKH was highest at 60 mins.(Figure 6).

From the plot, further increase in contact time for Barium and Zinc increases with rate of adsorption while for Mercury, further increase in contact time after 60 mins reduces the rate of adsorption; this may be attributed to the volatile nature of Mercury.

c) Effect of carbon dosage

The amount of carbon used in the process was found to affect the adsorption process. Figures 7 to 9 show that percentage removal of Barium, Zinc and Mercury increased with increasing carbon dosage.

The maximum adsorption of APKH in Barium, zinc and Mercury was attained at a particular dose (4g) of the adsorbent, whereas the maximum for APKS varied. Barium and Mercury uptake on APKS attained maximum adsorption at 5g, while Zinc adsorption attained the maximum at 3g. The variations in carbon dosage and rates of adsorptions are due to ability of contaminants to be adsorbed on the exterior carbon surface; move into the carbon powder and be adsorbed into the interior walls of carbon. These variations are due to varying iodine number, surface area, pore space and particle size distribution in APKS and APKH.

d) Adsorption Isotherm Studies

The Langmuir isotherm model is expressed as (Henderson et al., 2009, Igoni et al., 2009):

$$\frac{c}{(q)} = \frac{1}{ab} + \frac{1}{a}(c)$$

where q = mass of solutes adsorbed per mass of adsorbent, c = concentration of adsorbate in solution inequilibrium with the adsorbate adsorbed, a and b areconstants obtained by plotting c/q against c. Theslope is 1/a while the intercept is 1/ab. Figures 10 to 15show the Langmuir isotherm plots for the adsorption ofBarium, Zinc and Mercury in Palm kernel shell and huskrespectively.

The Freundlich isotherm model is given by the following equation:

$$q = K_f c^{\frac{1}{n}}$$

where $K_{\rm f}$ and n are constants. The linearised form of this equation becomes:

$$\ln q = \ln K_f + \frac{1}{n} \ln c$$

By plotting In q versus In C, the constants K_f and n are obtained. The slope a=1/n while the vertical axis intercept $b=In K_f$, therefore n=1/a and $K_f=e^b$ The plot of In q versus In c are given in figures 16 to 21 for the adsorption of Barium, Zinc and Mercury using palm kernel shell and husk respectively.

Table 1 shows the comparison between the Langmuir and Freundlich regression coefficients. It was observed that the experimental data fitted the Freundlich APKH isotherm model best. The Langmuir model was far from unity while the Freundlich isotherm model was closer to unity, with the Freundlich APKH better than Freundlich APKS.

Table 2 shows the comparison in values of regression coefficient, adsorption intensity and adsorption constant (k) for various studies; Ayotamuno and others (2007), Opete. (2008) and this study. The adsorption intensities of Barium, Zinc and mercury were the lowest, showing that low amounts of heavy metals are adsorbed at low concentrations of adsorbate and this varies with its regression coefficients and their constants respectively.

IV. Conclusion

Palm kernel shell and husk have been shown to have an average capacity for the removal of Barium, zinc and mercury present in thermally treated drill cuttings used in the oil and gas industry. This is with respect to its low values of n and k^f. Removal of Barium zinc and mercury are pH, contact time and carbon dosage dependent with optimal pH for Barium, zinc and mercury being 9. For APKS, maximum percentage adsorption for Barium and Zinc uptake occurred at 150 mins, while Barium and Zinc APKH occurred at 120 mins. Mercury APKS and APKH maximum occurred at 60 minutes. The maximum carbon dosage for Barium, zinc and mercury uptake in APKH were attained with 4g, whereas Barium and Mercury uptake in APKS was 5g and zinc uptake in APKS 3g. The equilibrium adsorption data obtained showed moderate adsorption favorable to

Freundlich Isotherm for palm kernel husk preferably. This work showed that with futher research, the readily available agricultural waste –Palm Kernel shell and Husk can be used to effectively adsorb Barium, zinc and mercury, thereby curbing disposal of toxic heavy metals.

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Tables

Table 1 : A Comparism of the Langmuir and Freundlich Regression Coefficients.

	BARIUM		ZINC		MERCURY	
	1	2	1	2	1	2
Langmuir	36.16	71.47	45.53	65.14	4.41	81.12
Freundlich	61.34	99.84	70.09	85.66	25.1	89.92

• *1: APKS

• *2: APKH

Table 2 : A	Comparism of the	Langmuir and	Freundlich Regression	Coefficients for various studies.

	Heavy Metal	Adsorbent	R²(%)	n	К
1.	Cr	PAC	98.1	1.3200	7.68 x 10 ⁻¹
2.	,,	APKS	91.94	1.3755	1.372 x 10 ⁻¹
	3.3	APKH	86.39	1.5511	2.056 x 10 ⁻¹
	Pb	APKS	89.37	1.5087	9.2 x 10 ⁻²
	3.3	APKH	96.74	1.6199	1.2 x 10⁻¹
3.	Ва	APKS	61.34	0.0683	2.72 x 10 ⁻¹
	3.3	APKH	99.84	0.9420	6.727 x 10 ⁻¹
	Zn	APKS	70.09	0.6249	2.408 x 10 ⁻¹
	,,	APKH	85.66	0.0710	7.055 x 10 ⁻¹⁶
	Hg	APKS	25.1	0.4421	2.8 x 10 ⁻⁴
	,,	APKH	89.92	0.2935	4.059 x 10 ⁻⁶

* Ayotamuno and others (2007)

* Opete O. (2008)

* This study

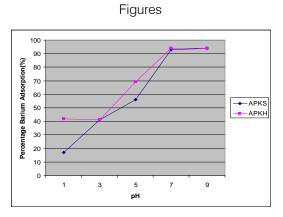


Figure 1 : Plot of pH against percentage Barium adsorption with APKS and APKH

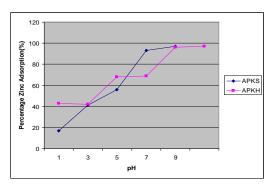


Figure 2 : Plot of pH against percentage Zinc adsorption with APKS and APKH

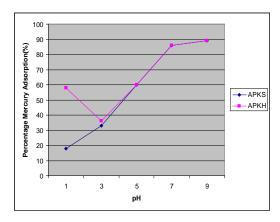


Figure 3 : Plot of pH against percentage Mercury adsorption with APKS and APKH

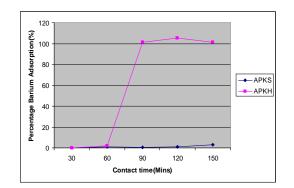


Figure 4 : Plot of contact time against percentage Barium adsorption with APKS and APKH

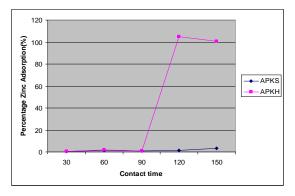


Figure 5 : Plot of contact time against percentage Zinc adsorption with APKS and APKH

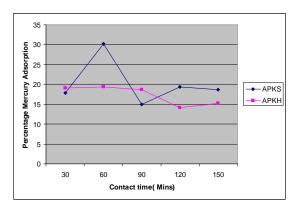


Figure 6 : Plot of contact time against percentage Mercury adsorption with APKS and APKH

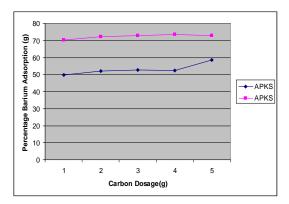


Figure 7 : Plot of Carbon Dosage against percentage Barium adsorption with APKS and APKH

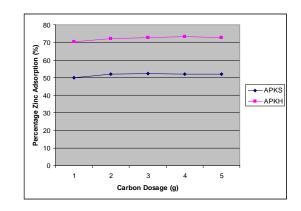


Figure 8 : Plot of Carbon Dosage against percentage Zinc adsorption with APKS and APKH

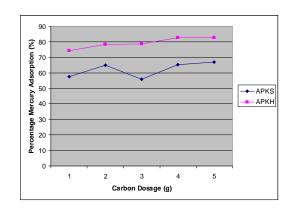


Figure 9: Plot of Carbon Dosage against percentage Mercury adsorption with APKS and APKH

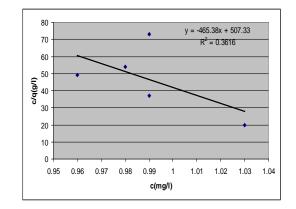


Figure 10 : Plot of Langmuir Isotherm for Barium Adsorption with APKS

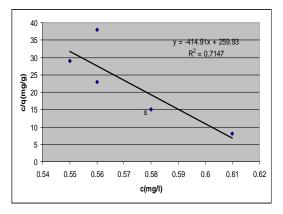


Figure 11 : Plot of Langmuir Isotherm for Barium Adsorption with APKH

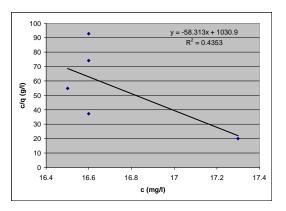


Figure 12 : Plot of Langmuir Isotherm for Zinc Adsorption with APKS

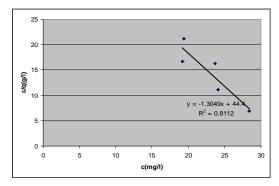


Figure 15 : Plot of Langmuir Isotherm for Mercury Adsorption with APKH

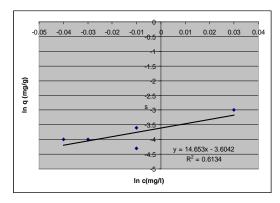


Figure 16 : Plot of Freundlich Isotherm for Barium Adsorption with APKS

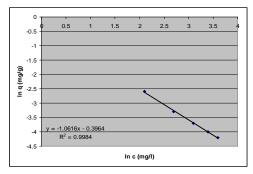


Figure 17: Plot of Freundlich Isotherm for Barium Adsorption with APKH

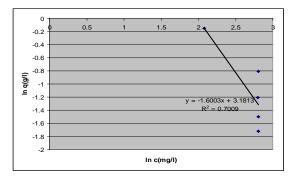


Figure 18 : Plot of Freundlich Isotherm for Zinc Adsorption with APKS

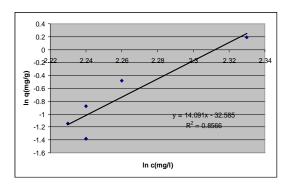


Figure 19 : Plot of Freundlich Isotherm for Zinc Adsorption with APKH

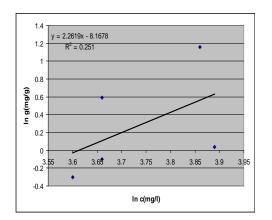


Figure 20 : Plot of Freundlich Isotherm for Mercury Adsorption with APKS

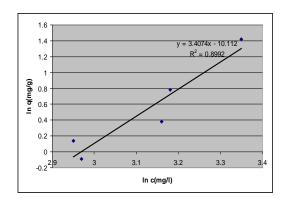


Figure 21 : Plot of Freundlich Isotherm for Barium Adsorption with APKS

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The Effect of High Water Content of Fuel on Diesel Engine Emission

By Abdulaziz H. El-Sinawi, Kifah Takrouri, Omar Osta & No'man Haimour King Faisal University

Abstract - Introducing water with fuel in diesel engines has been proved to be powerful and economical method for reducing combustion pollutant emissions from the engine. Most studies available in literature discuss the effect of adding water to the fuel in the range 5 to 10% water. In this paper, results of modeling study are presented where the effect of relatively high water content in fuel has been investigated. The fuel used was a surrogate mixture composed of 70% n-Heptane and 30% Toluene with two water contents of 25% and 35% by volume. The modeling study was performed using the commercially available software CHEMKIN at diesel engine-relevant conditions. The results show that water, even at high percentages, still has the tendency to reduce pollutant emissions as its concentration increases. Also, fuel consumption was found to decrease by increasing water content. However, the tradeoff with CO and Unburned Hydrocarbons UHC emissions was maintained.

Keywords : water-in-fuel emulsion, pollutants reduction, diesel, surrogate fuel, combustion.

GJRE-C Classification : FOR Code: 030599



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The Effect of High Water Content of Fuel on Diesel Engine Emission

Abdulaziz H. El-Sinawi $^{\alpha}$, Kifah Takrouri $^{\sigma}$, Omar Osta $^{\rho}$ & No'man Haimour $^{\odot}$

Abstract - Introducing water with fuel in diesel engines has been proved to be powerful and economical method for reducing combustion pollutant emissions from the engine. Most studies available in literature discuss the effect of adding water to the fuel in the range 5 to 10% water. In this paper, results of modeling study are presented where the effect of relatively high water content in fuel has been investigated. The fuel used was a surrogate mixture composed of 70% n-Heptane and 30% Toluene with two water contents of 25% and 35% by volume. The modeling study was performed using the commercially available software CHEMKIN at diesel enginerelevant conditions. The results show that water, even at high percentages, still has the tendency to reduce pollutant emissions as its concentration increases. Also, fuel consumption was found to decrease by increasing water content. However, the tradeoff with CO and Unburned Hydrocarbons UHC emissions was maintained.

Keywords : water-in-fuel emulsion, pollutants reduction, diesel, surrogate fuel, combustion.

I. INTRODUCTION

he exhausted gaseous phase from diesel engines is usually composed of hundreds of chemical compounds and pollutants. High engine temperatures are usually involved in the exothermic reactions of fuel with air. With the existence of nitrogen and oxygen from air or fuel, nitrogen oxides NO_x and carbon oxides COx are among the major pollutants emitted by the combustion process. To a lesser extent, Unburned Hydrocarbons UHC and Volatile Organic Compounds VOC can also be emitted as well as many trace amounts of hazardous air pollutants. Due to the impact of these emissions on the environment, many countries are gradually strengthening the regulations related to these emissions. As a result, diesel engine designers are challenged with two often conflicting goals: reducing the engine emissions and at the same time improving its efficiency.

Recently, intensive research has been done worldwide on developing methodologies that reduce the diesel engine pollutant emissions. Many of these methodologies are focused on improvements in engine design. However, such improvements are usually costly and difficult to apply.

One method for pollutants reduction is to decrease the engine high temperature by bringing water into the combustion chamber. Water can be injected to the engine inlet manifold, directly injected to the combustion zone or can be injected in the form of water-in-diesel emulsion. Among these technologies, the use of water-in-diesel emulsions was reported to be the most efficient to reduce pollutant emissions as, in addition to other effects, water is injected directly into the combustion zone causing a larger decrease of the combustion temperature [1, 2]. Water-in-diesel emulsions can also be applied with virtually no additional costs.

The main reason for the reduction in pollutants emission in the use of water-in-diesel emulsion is the reduction in the combustion temperature. The reduction in the temperature itself is due to (1) vaporization of liquid water which decreases the internal energy and (2) increasing heat capacity due to having higher trapped mass of vapor. Moreover, water provides its improvements physically by enhancing mixing within the engine and chemically by reacting with the combustion gases. Also, water, which has a much lower boiling point than that of the surrounding diesel in the emulsion, suddenly and dramatically expands upon vaporization. This expansion increases turbulence and enhances the mixing between oxygen and fuel. This process is usually referred to as the micro-explosion process. Several researches reported that the emulsion combustion reactions are not simply kinetically controlled, but the turbulence inside the combustor has a considerable effect.

The water-in-diesel emulsions emit reduced amounts of particulate matters and soot. These emulsions also show better burning efficiency and therefore reduce fuel consumption without engine modifications. However, one limitation is the potential increase in CO and UHC production.

In practice, extensive research has been done on the use of water-in-fuel emulsions. Some water-infuel emulsions, such as the Aquazole formulation developed by TOTAL, are already in use today on a large number of vehicles in France and other countries in Europe. It has been claimed that this formulation brings about a reduction of up to 30% in NO_x and of up to 80% in soot [3]. Water-in-fuel emulsions have also been used on autobuses in some areas of China.

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In literature, high water content in diesel emulsions (more than 10%) was rarely discussed and needs to be thoroughly investigated [4, 5].

This study provides modeling analysis of emissions reduction obtained by using water-in-fuel emulsions with water ratios of 25% and 35% in volume. Surrogate fuel was used in this study as detailed kinetic and thermodynamic data can be available for selected surrogate fuels but not available for conventional diesel. Conventional diesel fuels are blends of several hundreds of individual components [6]. Surrogate fuels can be reasonably considered as simpler representatives of the chemically complex conventional diesel fuel in terms of performance and emissions behavior [6]. As one-component surrogate fuel may not accurately reflect the behavior of real diesel, surrogate fuels of more than one component are usually employed. However, a surrogate fuel should be prepared with the smallest possible number of components that provide the desired representation of the conventional diesel. Surrogate fuel composed of 70% n-Heptane and 30% Toluene was used in this study.

In most of the studies conducted with regard to water-in-fuel emulsions only NO_x and particulate matter were the emissions of concern [7]. Some recent studies investigated the CO and UHC emissions. In this study, emissions of CO, UHC, VOC and other compounds like propargyl, benzene and others were looked at as well as emissions of n-Heptane; one component of the surrogate fuel used.

II. LITERATURE REVIEW

This literature review is focused on the combustion studies where relatively high water content in fuel (20% and above) was used. There are inconsistent results reported in different studies where high water contents were used which reveal the need for further research on combustion of such emulsions. Lif et al. [3] reported that the typical water content of a diesel emulsion is between 10 and 20%. However, Nazha et al. increased the amount of water up to 50% and reported a 60% reduction in NO_x but noticed a slight increase in smoke emissions [9].

Park et al. [8] used water up to 40% in combustion tests and the general picture of pollutants reduction was maintained. However, small effects on CO and UHC emissions were reported. In another study [3], the NO_x and soot levels were significantly reduced for water contents between 15–45% but the CO and UHC emissions increased. Canfield [10] reported a significant pollutant emissions reduction with up to 45% water, by volume, in diesel. He reported that of six technologies evaluated for pollutants reduction, the water-in-diesel emulsion promises the easiest solution.

Matheaus et al. [6] used diesel having 20% water by mass in an experimental study to investigate

the effect of water on the engine emissions and fuel consumption rate. He reported a 19% reduction in NO_x and 16% reduction in particulate matter. However, there was 42% increase in CO and 28% increase in UHC.

III. Modeling, Results and Discussion

In this study, the commercially available code CHEMKIN was used to investigate the combustion of the selected surrogate fuel (70% n-Heptane and 30% Toluene) with water content of 25 and 35% by volume. CHEMKIN contains an extensive database of temperature dependent properties and a built-in chemical reaction kinetics solver which is appropriate for combustion detailed calculations. CHEMKIN computation is performed using 393 species and 1,925 reactions. The Partially Stirred Reactor PaSR modeling approach was used in this analysis. PaSR can assess the extent of interactions between turbulence and chemical kinetics and therefore can provide information on how turbulence intensity inside a combustor will affect combustion [11].

The initial temperature and pressure for the computations are 900 K and 39 bar; respectively, with air as the oxidizer. The temperature and pressure values agree with the actual parameters of a real engine [6]. The fuel/air equivalence ratio was 0.8.

Fig. 1 shows typical profiles of some major exhaust emissions resulting upon combustion of the surrogate fuel with 25% water. Fig. 2 shows the emissions plotted against water mole fraction. Most of the data of this paper are plotted against water mole fraction in the exhaust gaseous phase being a better representing parameter.

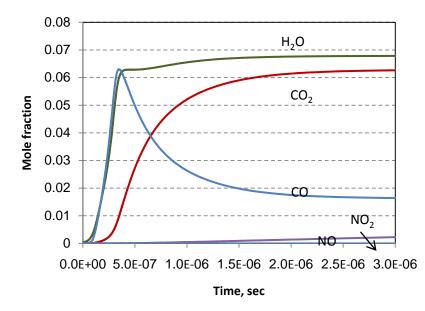


Fig. 1 : Typical NO_x , CO_x and water mole fraction profiles for 25% water content in emulsion of 70% n-Heptane and 30% Toluene.

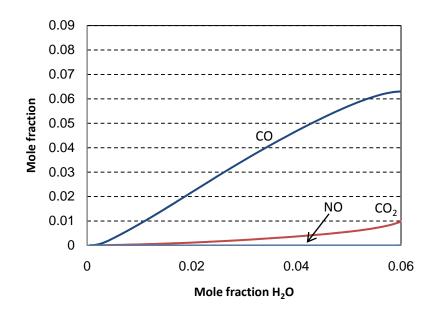


Fig. 2: Typical plots of CO_x and NO mole fractions for 25% water content in emulsion of 70% n-Heptane and 30% Toluene.

Fig. 3 shows the calculated combustion temperature for the two water contents. As shown in the figure, the peak temperature decreases with increasing water content.

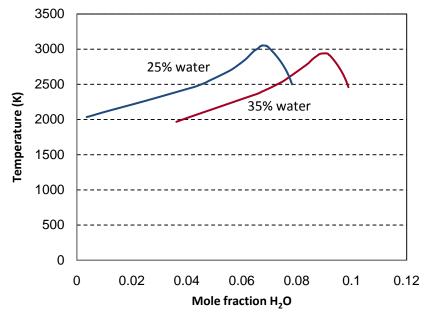


Fig. 3 : Calculated temperatures for 25% and 35% water in a surrogate fuel of 70% n-Heptane and 30% Toluene.

Fig. 4 shows the water mole fraction profile for the two emulsions. As expected, the water concentration in the exhaust gas is higher for the emulsion with higher water content.

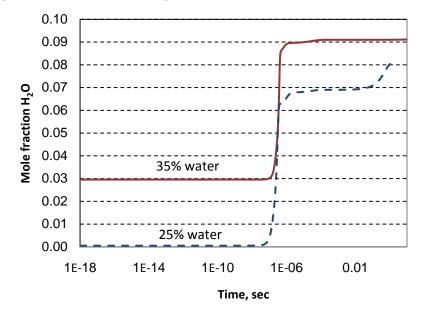


Fig. 4 : Water mole fraction profile for emulsions with water contents of 25% and 35% in a surrogate fuel of 70% n-Heptane and 30% Toluene.

Fig. 5 shows the mole fraction of n-Heptane for the two emulsions. The emulsion with the higher water content results in a higher n-Heptane concentration which is consistent with findings of other studies that increasing the water content may decrease the fuel consumption [3, 10, 12].

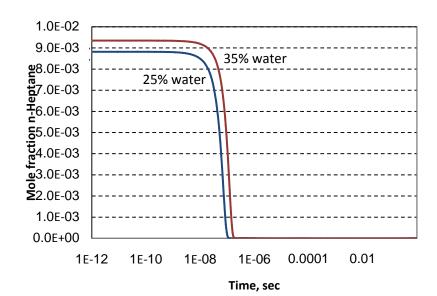


Fig. 5: n-Haptane mole fraction for emulsions with water contents of 25% and 35% in a surrogate fuel of 70% n-Heptane and 30% Toluene.

Fig. 6 shows the effect of water content on CO, where CO and UHC are produced as a result of incomplete combustion. At high temperatures with more sufficient time, these two products further oxidize and form carbon dioxide and water.

As shown in Fig. 6, the final CO concentration increases with increasing water content in the emulsion. Water reduces the temperature inside the combustion chamber to a level where CO oxidation is inhibited. The reaction rates of CO with O, O_2 and OH decrease with decreasing temperature. Therefore the production rate of CO is increased. CO production rates for the two water contents are shown in Fig. 7 where the peak CO production rate increases as the water content increases.

The increase of CO level with increasing water content was experimentally validated by several researchers [13].

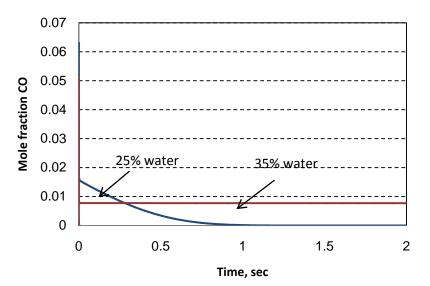


Fig. 6 : CO mole fraction for 25% and 35% water content in emulsion with 70% n-Heptane and 30% Toluene.

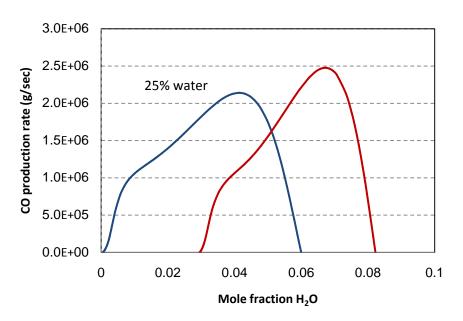


Fig. 7: CO production rate (g/sec) for 25% and 35% water content in emulsion with 70% n-Heptane and 30% Toluene.

Fig. 8 shows the effect of water content on the UHC and VOC. As shown in the figure the amount of UHC increases as the water content increases. This result agrees with the findings of Milton and Carter, who showed for tests simulating a typical city driving cycle that UHC tend to increase by water injection [14], and with the findings of Prakash et al. [16].

UHC emissions are direct result of incomplete combustion of fuel. As water content in the emulsion increases, longer ignition delay and lower combustion temperature are usually experienced which may lead to the emission of more partially oxidized hydrocarbons [14, 16]. Therefore, while benefits to several pollutant emissions occur as a result of water addition to the fuel, these may be offset by the increase in CO and UHC emissions. However, researchers used emulsions of water with heavy oil [13] and biodiesel [2] reported a decrease in UHC emissions with increasing water content and thus the effect of water on UHC emissions needs further thorough investigation. Fig. 8 also shows the effect of water content on VOC emissions which like the UHC, increase with increasing water content. VOC are typically not acutely toxic, but instead may have compounding long-term health effects.

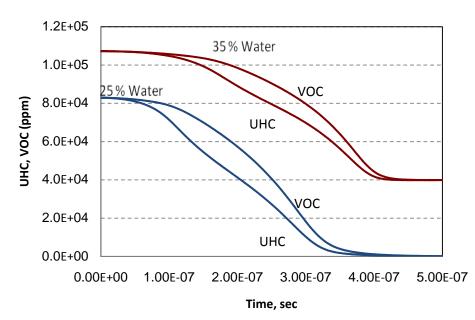


Fig. 8: UHC and VOC for 25% and 35% water content in emulsion with 70% n-Heptane and 30% Toluene.

The effect of water content on propargyl C_3H_3 formation is shown in Fig. 9. C_3H_3 concentration decreases with increasing water content. C_3H_3 is a major precursor to form Polycyclic Aromatic Hydrocarbons PAH as two molecules of C_3H_3 can combine together to form Benzene ring C_6H_6 . For low water contents, ElSinawi [15] found that the formation of C_3H_3 decreases with increasing water content up to 5% then it increases when higher amounts of water were added with surrogate fuel composed of 80% n-Heptane and 20% Toluene.

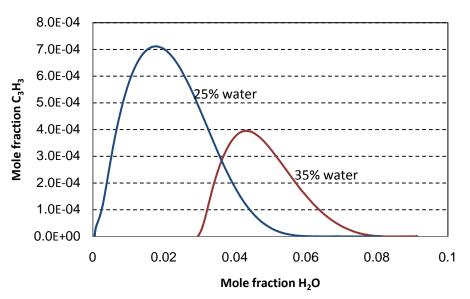


Fig. 9 : C₃H₃ mole fraction for 25% and 35% water contents in emulsion with 70% n-Heptane and 30% Toluene.

The effect of water content on Benzene formation is shown in Fig. 10. Benzene concentration decreases with increasing water content. Benzene can

combine with Toluene and therefore has an important role in PAH and soot formation [4].

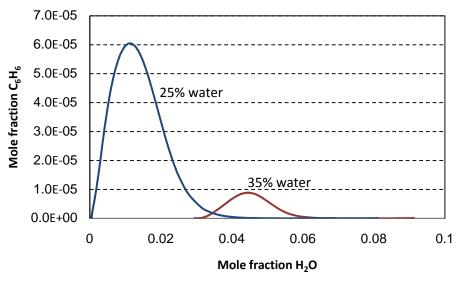




Fig. 11 shows the effect of water content on the concentration of fulvene, which is a toxic compound. Increasing water content decreases fulvene emissions.

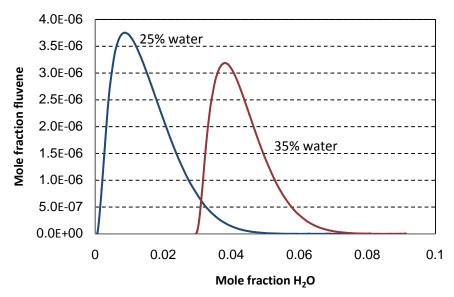


Fig. 11 : Fulvene mole fraction for 25% and 35% water contents in emulsion with 70% n-Heptane and 30% Toluene.

The concentration of nC_3H_7 group is shown in Fig. 12. nC_3H_7 can lead to the formation of formaldehyde which is very toxic and corrosive. The concentration of

 $nC_{\rm 3}H_{\rm 7}$ decreases as water content in the emulsion increases.

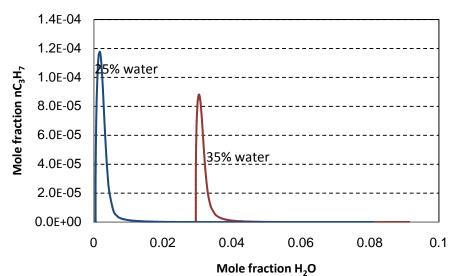


Fig. 12 : nC₃H₇ mole fraction for 25% and 35% water content in emulsion with 70% n-Heptane and 30% Toluene.

IV. Conclusions

The effect of adding a relatively high percentage of water to surrogate fuel composed of 70% n-Heptane and 30% Toluene on the formation of pollutants was discussed. The use of water-in-fuel emulsion is an effective technique to reduce pollutant emissions from surrogate fuel even at relatively high water contents of 25% and 35%. It was found that as water content increases the reduction of the emissions studied increases and the fuel consumption slightly decreases. However, the tradeoff with CO and UHC emissions was maintained.

V. Abbreviations

CO CO	Carbon Monoxide Carbon Dioxide
COx	Carbon Oxides
NO	Nitrogen Monoxide
NO ₂	Nitrogen Dioxide
NO _x	Nitrogen Oxides
PAH	Polycyclic Aromatic Hydrocarbons
UHC	Unburned Hydrocarbons
VOC	Volatile Organic Compounds

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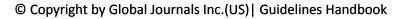
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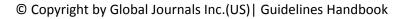
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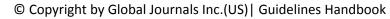
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Abstract	Clear and concise with appropriate content, Correct format. 200 words or below	Unclear summary and no specific data, Incorrect form Above 200 words	No specific data with ambiguous information Above 250 words
Introduction	Containing all background details with clear goal and appropriate details, flow specification, no grammar and spelling mistake, well organized sentence and paragraph, reference cited	Unclear and confusing data, appropriate format, grammar and spelling errors with unorganized matter	Out of place depth and content, hazy format
Methods and Procedures	Clear and to the point with well arranged paragraph, precision and accuracy of facts and figures, well organized subheads	Difficult to comprehend with embarrassed text, too much explanation but completed	Incorrect and unorganized structure with hazy meaning
Result	Well organized, Clear and specific, Correct units with precision, correct data, well structuring of paragraph, no grammar and spelling mistake	Complete and embarrassed text, difficult to comprehend	Irregular format with wrong facts and figures
Discussion	Well organized, meaningful specification, sound conclusion, logical and concise explanation, highly structured paragraph reference cited	Wordy, unclear conclusion, spurious	Conclusion is not cited, unorganized, difficult to comprehend
References	Complete and correct format, well organized	Beside the point, Incomplete	Wrong format and structuring

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