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ORIGINAL PAPER

Removal of oil and grease contamination from stream water using the granular activated carbon block filter

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Abstract Most sources of oil and grease (O&G) are insoluble in water. Because the specific gravity of O&G is lower than water, it floats on the top surface. The presence of O&G may have adverse impact on water resources management. Activated carbon can remove O&G from water by adsorption process. Still the use of physical models to adsorb O&G from stream water needs to be verified. This study proposes the mathematical models for adsorption of O&G from stream water using the granular activated carbon block filter (GACBF). The parameters in equations are all physically meaningful, and the experimental data validation shows that the equations are sufficiently accurate. The proposed models to calculate the accumulation rate, lifetime, and adsorption capacity for the adsorption of O&G onto GACBF from Ulu Pontian River water are presented to contribute to clean technology and environmental contamination investigation and assessment.

Keywords Adsorption · Granular activated carbon block filter · Mathematical model · Oil and grease · Ulu Pontian River water

List of symbols

a	Slope of curve $(t/C)^{1/3}$ versus t (dimensionless)
A	Area traversed (m^2)
b	Interception of curve $(t/C)^{1/3}$ versus t (dimensionless)
c	Accumulation quantity of O&G traversed through the GACBF (g/kg)
C	Amount of contaminant O&G that has already been accumulated in the GACBF (g/kg)
k_1	Accumulation rate coefficient (g/kg h^2)
L	Ultimate adsorption capacity of the GACBF (g/kg)
P	Available space of the GACBF to be filled by O&G (g/kg)
P_o	Total available space of the GACBF before any accumulation of O&G occurred (g/kg)
r	Accumulation rate (g/kg.h)
S	Concentration of O&G measured in the stream water (in g/m^3)
t	Accumulation time (h)
v	Water velocity (m/h)

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Introduction

Oil and grease (O&G) is a measure of a variety of substances, including fuels, motor oil, lubricating oil, hydraulic oil, cooking oil, and animal-derived fats and oils. It is one of the critical components of the contaminant load in urban storm water runoff that deleteriously affects the quality of receiving water bodies by carrying a significant load of various storm events. This will be facing the problems in water resource management: pollution and supply shortage. The O&G presented in raw produced water elapses the sand filter and reduces the water treatment efficiency, and is not readily removed by most conventional drinking water treatment processes. Because the excessive amount of O&G jeopardizes the quality of drinking water, it is crucial to human health. Long-term use of high doses of O&G can lead to loss of normal bowel response. Other dilemmas are nausea, abdominal cramps, vomiting, diarrhea, leakage of oil from rectum (commonly with a higher dose), and reduced absorption of nutrients from food. The significance of possible effects on human health because of O&G overdose in drinking water has been the subject of exhaustive reviews by several research institutions (Greenberg et al. 2003; Farmakia et al. 2007; Pawlak et al. 2005).

Granular activated carbons (GACs) are used extensively as adsorbent for separation of contaminants from gaseous and liquid phase due to their high surface area, high porosity and good surface reactivity (Bansal et al. 1988; Rodrígues-Reinoso et al. 1997; Fulazzaky 2011). The characteristics of GAC fabricated from different raw materials of such as wood, coconut shell, and charcoal are suitable for many filter applications. Surface chemical functions of adsorbent involved to the interaction with an adsorbate could be strongly attracted due to chemisorption and van der Walls forces. These unique characteristics make GAC very versatile materials, which have been studied not only as adsorbents, but also as catalysts and catalyst supports for different purposes such as the removal of pollutants from gaseous or liquid phase and the purification or recovery of chemicals (Derbyshire et al. 2001). The use of waste materials as low-cost adsorbents is attractive due to their contribution in the reduction of costs for waste disposal and is suitable for adsorption of certain contaminants from aqueous solution. Still the usability of GAC is more efficient for adsorbing a greater amount of the pollutants. Evidence suggesting that selection of cheaper raw material and/or appropriate method of production such as carbonization temperature and carbonization time can reduce manufacturing costs (Lafi 2001; Sudaryanto et al. 2006).

Adsorptions of organic and inorganic matter onto activated carbon (AC) in aqueous solution have been proposed for many applications. In fact, it is known that around 80% of the world production of AC is used in liquid phase. Although the development of the low-cost adsorbents using waste materials to remove specific pollutants of such as heavy metals and dyes from aqueous solution has been studied extensively (Shukla et al. 2002; Babel and Kurniawan 2003; Wang et al. 2003; Chuah et al. 2005; Ramesh et al. 2005; Crini 2006), the application of AC based on peat materials to remove the metals from waste streams has been proposed (Brown et al. 2000), and the powdered AC was investigated by developing adsorption isotherms for O&G using wastewater collected from a Ford plant (Mueller et al. 2003), the use of GAC to remove O&G from stream water is still not fully understood. The objectives of this study are (1) to investigate the possibility commercial GAC to directly remove O&G from steam water, (2) to propose the mathematical models for the dynamic adsorption of O&G onto GAC from stream water, and (3) to determine accumulation rate and adsorption capacity which will help us find out the period required for change of the filter.

The importance to monitor O&G for Ulu Pontian River

The Ulu Pontian River emerges from the Hill's Bukit Batu and traverses the western-east region of Johor state for some 18 km until it reaches the coast. The basin is approximately a 16 km long and 10 km wide and has an area of 159 km^2, or about 0.8% of the Johor region, inhabited by about 150,000 people in 2008. The annual average flows are 1.5 m^3/s observed at Bukit Batu near the water intake canal (WIC) and 2.0 m^3/s at the estuary. Even though the major use of water in the basin is for agricultural, agro-industrial, and domestic purposes, the effective river basin management maintains environmental flows, or water levels, sufficient to sustain all the elements of aquatic ecosystems. One of the important uses of Ulu Pontian River water provides for Bukit Batu water treatment plant (BBWTP). The operational BBWTP has been functioning since 1965 under administration of Syarikat Air Johor Holdings Sdn. Bhd. (SAJ) as local water supply company. The BBWTP supplies the drinking water to about 6,775 people of Felda Bukit Batu, Pekan Bukit Batu, and Ladang Kuala Kabong. This treatment plant uses the conventional technology of chemical and physical treatment and disinfection. The raw water source is from Ulu Pontian River abstracted through the WIC with a 150 m^3/h flow rate during the 13 h period for each day.

The sources of pollutants are due to point sources such as the pollutants from industrial and domestic wastewater and non-point sources such as pollutants from agricultural activities and erosion. A risk assessment was evaluated in a previous study by Zhang et al. (2003) for the persistent organic pollutants in the Minjiang River Estuary, China. Even if O&G's contaminant is not an issue of non-point sources, liquid and solid wastes discharged from industries have resulted in significant water pollution. Widespread lack of adequate disposal of waste O&G original from industries leads to contamination of watercourses and is one of the significant sources of water pollution. For example, one of the important cases of the stream water pollution (see Fig. 1) has occurred for Ulu Pontian River on October 14, 2009. There is due to the excessive amount of O&G was found in raw water, the use of conventional treatment system was ineffective to produce a good quality of drinking water. As consequence, the SAJ has decided to close the operational BBWTP from 15 to 16 October 2009 to prevent contamination of the community drinking water reserved in the BBWTP's storage tanks. Therefore, a

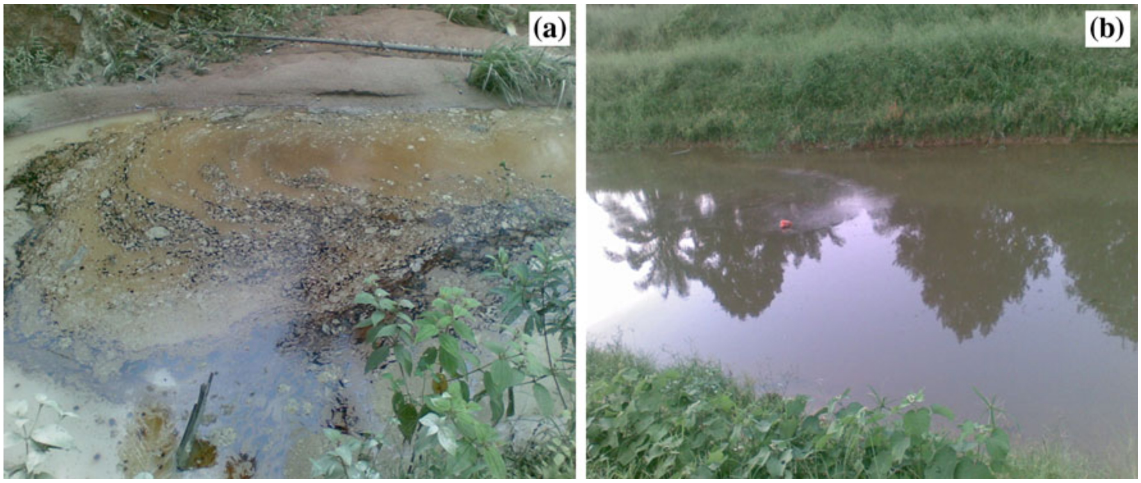

Fig. 1 Image of Ulu Pontain river water: **a** near the sources of pollutant and **b** near the WIC

simple technique as pre-treatment to remove O&G from the stream water is expected. A previous study by Valcárcel et al. (2011) proposed the need for water quality monitoring and research in urban rivers, as well as the need for improved water treatment techniques able to eliminate the pharmaceutically active compounds from the effluent waters as well as from drinking water sources.

Materials and methods

Models development

Some limitations such as concentration of O&G in stream water, available pore space and accumulation time can be proposed as variable to develop the linear equations. This study focused on the variability of accumulation time, due to lifetime of the granular activated carbon block filter (GACBF) installed at mouth of the WIC needs to be verified. It was the concern of the research project to understand a filter change most easily. The mass balance equation is written for the adsorption of O&G onto the GACBF to level of the system (macroscopic balance) in different conditions of its functioning using the following formula:

$$\frac{dP}{dt} = -r \tag{1}$$

where P is available space of the GACBF to be filled by O&G (in g/kg), t is accumulation time (in h), and r is accumulation rate (in g/kg h).

It is assumed that if r is first order, then rearrangement of Eq. 1 gives a continuous equation expressed in form of:

$$\frac{dP}{dt} = -k_1 t \tag{2}$$

where k_1 is accumulation rate coefficient (in g/kg h^2).

When we separate the variables, Eq. 2 can be integrated in form of:

$$P = P_o \times e^{-k_1 t} \tag{3}$$

where P_o is total available space of the GACBF before any accumulation of O&G occurred (in g/kg).

If we recognize that C is amount of O&G which has been already accumulated in the GACBF (in g/kg) and L is ultimate adsorption capacity of the GACBF (in g/kg) as schematized in Fig. 2, theoretically. This gives

$$L - C = P \tag{4}$$

By substituting Eq. 3 into Eq. 4 yields the equation, such that:

$$L - C = P_o \times e^{-k_1 t} \tag{5}$$

But $P_o = L$, rearrangement of Eq. 5 gives

$$C = L - L\left(e^{-k_1 t}\right) \quad \text{or} C = L\left(1 - e^{-k_1 t}\right) \tag{6}$$

A variety of methods may be used to determine k_1 and L from an experimental set of C data. The simplest and

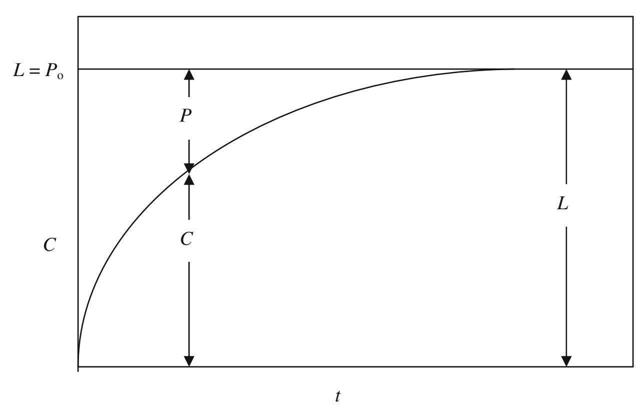

Fig. 2 Schematic of theoretical model for adsorption of O&G onto GAC

least accurate method is to plot C versus t. This results in a hyperbolic first-order curve of the form shown in Fig. 2. L is estimated from the asymptote of the curve. The rate equation is used to solve for k_1. It is often difficult to fit an accurate hyperbola to data that are frequently scattered. The usual graphical methods for first-order reactions cannot be used because the semilog plot requires knowledge of the total available space of the GACBF which, in this case, is one of the constants we are trying to determine, that is L. Methods that linearize the data are preferred. One simple method using in this study around this impasse is called Thomas' graphical method (Davis and Cornwell 2008). Rearranging Eq. 6 in form of the linear equation yields:

$$\left(\frac{t}{C}\right)^{\frac{1}{3}} = \frac{(k_1)^{\frac{2}{3}}}{\left(6L^{\frac{1}{3}}\right)} \times t + \frac{1}{(k_1 L)^{\frac{1}{3}}} \quad (7)$$

Equation 7 is analogous to the equation $y = a\,(x) + b$, where a is defined as slope and b is interception of curve $(t/C)^{1/3}$ versus t, y is $(t/C)^{1/3}$, and x is t. Accumulation rate coefficient (k_1) is constant and is thus expressed in the following relation that:

$$k_1 = 6\left(\frac{a}{b}\right) \quad (8)$$

Using Eq. 8 permits us to calculate the accumulation rate coefficient (k_1) if the values of the parameters (a; b) were verified analyzing the linear regression generated from the experimental data by plotting the curve $(t/C)^{1/3}$ versus t.

Ultimate adsorption capacity of the GACBF to remove O&G from stream water is defined as maximum quantity of O&G captured onto GAC and is constant. This expresses in the following relation that is

$$L = \frac{1}{6(a)(b)^2} \quad (9)$$

Using Eq. 9 permits us to calculate the capacity of the GACBF to remove O&G contaminants from stream water since the values of the parameters (a; b) were verified as for the calculation of k_1 in Eq. 8.

Experiment planning and laboratory analysis

Installation of the GACBF

This research project as shown in Fig. 3 was conducted in the WIC of BBWTP, ordered for adaptive problem solving to handle the case of O&G pollution in Ulu Pontian River. This is due to the stream water which provides the important water supply resource for the people. The GACBF was installed on 16 October 2009 at mouth of the WIC in effect filtering the water before it enters the BBWTP. Along the experiment, the stream water of 150 m^3/h flow rate regulated using a dedicated pump can continuously flow through the GACBF. The main characteristics of the GACBF are presented in Table 1. After passing the GACBF, the water is free from O&G and thus can be used as raw water for drinking water production. In a previous study by Mohan and Karthikeyan (2004) has presented the results pertaining to the adsorptive removal of reactive azo dye onto a low-cost coal-based adsorbent (charfines) and its efficiency in dye color sorption was compared with AC. This study used the commercial GAC as adsorbent originally delivered from Tay Scientific Sdn. Bhd. The 50 kg GAC (Fig. 3) was fitted with a fine mesh netting of 100 μm that preventing the GAC from getting out. Even if the GACBF must be changed quite often which adds to the expense of filter maintenance, the lifetime of the GAC is needed to be verified.

Fig. 3 The GACBF installed at mouth of the WIC

Table 1 Principle characteristics for the GACBF

No	Element	Unit	Dimension
1	Dimensions		
	Length	cm	120
	Width	cm	15
	Depth	cm	80
	Design volume	L	144
2	Effective volume	L	111
3	Flow rate	m^3/h	150
4	Weight of the GAC	kg	50
5	GAC density	kg/L	0.45

Water sampling

Water samplings in this study were classified into two phases. During phase 1, the monitoring of the river water was intended to verify level of the contaminant O&G in the stream water. The location of monitoring was selected in Ulu Pontian River near the WIC. The samplings were carried out hourly over a period of 6 h with the hourly monitoring from 08:00 am to 02:00 pm on October 14, 2009 and hourly over a period of 7 h with the hourly monitoring from 08:00 am to 03:00 pm on October 15, 2009. During phase 2, the monitoring of efficiency of the GACBF was intended to assess the possibility filtered water to be used as raw water for drinking water production. The monitoring of samples was conducted at inlet and outlet of the GACBF. The samplings were carried out hourly over a period of 15 h with the hourly monitoring from 07:00 am to 10:00 pm on October 17, 2009, two times at 10:00 am and 04:00 pm on October 19, 2009, and once at 00:30 pm on November 2, 2009. To analyze a volume of 2 L water sample for each time of monitoring was filled into the sampling bottle collected from each sampling location. All the water samples were sent to Central Laboratory of SAJ at Sri Gading, Batu Pahat, Johor, Malaysia for the measurement of O&G.

Laboratory analysis

The O&G analyses were performed on the same day of the sampling date. The water samples analysis was carried out using the partition-gravimetric method (APHA 1995) done by Chemical Analysts of Central Laboratory of SAJ.

Results and discussions

Monitoring the level of O&G in the river water

Figure 4 presents the results of water quality monitoring for Ulu Pontian River. During the first day monitoring on October 14, 2009, the stream water samples had concentrations in the range of 17.10 mg/L to 42.86 mg/L with an average value of 20.90 mg/L. Even if a concentration of 17.10 mg/L is the lowest O&G value, the use of Ulu Pontian River water as raw water source for drinking water production does not meet the Malaysia's Interim National Water Quality Standards (MINWQS). Maximum permissible concentrations or threshold level values for Class IIA standardized in the MINWQS are equal to 0.04 mg/L for mineral O&G and 0.7 mg/L for cooking oil. Uses designated on Class IIA waters include raw water for drinking water production, aquatic life other than fish, recreation, wildlife, industry, agriculture, and scenic value (DOE 2003). Thus, the quality of waters treated with a conventional treatment system can be used for drinking water. In the second day on October 15, 2009, the stream water quality was monitored hourly during 7 h. The aim is to explain in detail the potential sources of pollutants. Even if

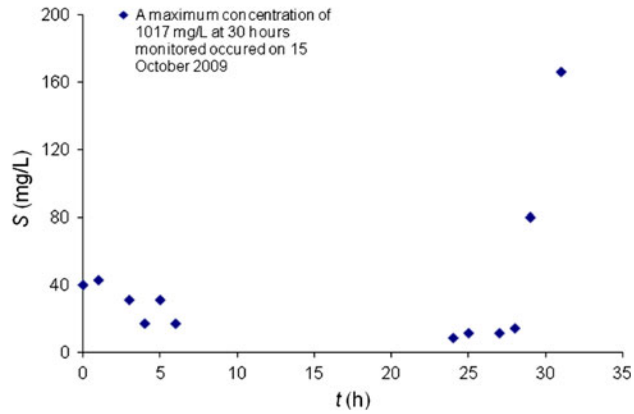

Fig. 4 Results of Ulu Pontian River water monitoring

a concentration of 8.57 mg/L monitored at 08:00 am is the lowest O&G value contained in the water, the results (see Fig. 4) present a very high concentration of 1017 mg/L monitored at 02:00 pm. This is due to the factories which were closed during the night and therefore not released any wastes into the environment. The O&G concentration monitored in the morning was very low, because of its residue has only detected in the water sample; the major contaminant has flowed downstream the monitoring location. In the afternoon, the pollutant loads from the factories has reached the monitoring location and thus monitored huge amount of O&G in the water sample. Excessive amount of O&G in raw water is very difficult to remove in a conventional treatment plant; it is the reason that SAJ has decided to close the operational BBWTP from 15 to 16 October 2009 to prevent contamination of drinking water in the storage tanks.

Monitoring the performance of the GACBF

The GACBF installed at mouth of the WIC is the only way to proactively monitor and insure the quality and safety of raw water for drinking water production. Using GAC as adsorbent is convenient and also offers a performance benefit. As the filter needs to be evaluated only once, there is a computation required to predict the efficiency and lifetime of adsorbent. For example, in a previous study by Sidhu (1983) has proposed to assess the collection efficiency of the environmental contaminants onto AC. This study verified that: (1) before passing the GACBF, the O&G concentrations in stream water range from 5.7 to 101.2 mg/L; the water quality does not meet the MINWQS, (2) after passing the GACBF, the O&G concentrations in the water range from 0.0 to 0.1 mg/L; the water quality is within the MINWQS and the performance of the GACBF to remove O&G contaminant is excellent, and (3) due to residual O&G attached on the canal banks before

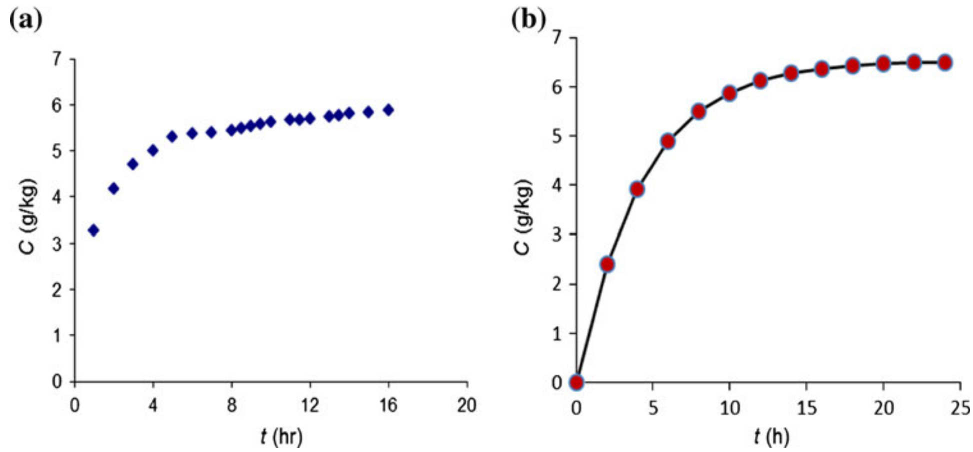

Fig. 5 Curve of O&G accumulated in the GACBF: **a** experiment and **b** calculation model

installation of the GACBF and thus released automatically with turbulent; a concentration of 2.1 mg/L O&G was detected for the first time sampling at outlet of the filter.

Accumulation rate and adsorption capacity of the GACBF

Hypothesis

A previous study by Mwinyihija et al. (2006) has proposed the ecotoxicological approach to assess the impact of tanning industry effluent on river health. Schmotzer et al. (2002) used a combination of experimental and process modeling methods to determine the fundamental equilibrium and kinetic parameters for multi-component adsorption of organic impurities on AC. In this study, a systematic approach for developing conceptual models was based upon the assumptions made that: (1) since the specific gravity of O&G is lower than water, it floats on the top surface; (2) concentrations of O&G measured are not the actual concentrations homogenously dispersed in bulk water; and (3) progressive increase of O&G accumulated in the GACBF shifts downward pursuant to *t*.

The actual O&G concentrations in the stream water were lower than the measured concentrations. This is due to the fact that water flows below the top surface were absent from O&G. To develop the mathematical models, the following hypothesis were made: (1) $c = v \times A \times S \times t$ is defined as accumulative quantity of O&G traversed through the GACBF, where v is water velocity (in m/h), A is the area traversed (in m^2) and S is concentration of O&G measured in the water sample (in g/m^3), and (2) $C = LN(c)$ is defined as quantity of O&G accumulated in the GACBF pursuant to t. Figure 5 shows the resulting plots a curve of C versus t is adjacent between the experiment (Fig. 5a) and calculation (Fig. 5b) and is analogous to curve theory as shown in Fig. 2.

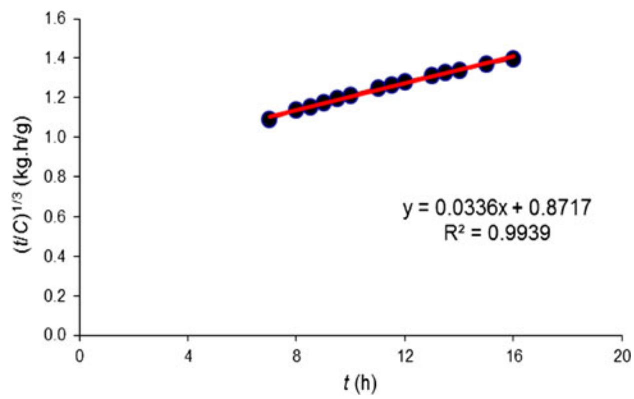

Fig. 6 Results of the linear regression analysis for the calculations of k_1 and L

Calculation

The curves of theory, experiment, and calculation (see Figs. 2, 5) of C versus t show that the increasing trends are comparable. Better knowledge of empirical models gives new insight for the calculations of accumulation rate and adsorption capacity of the GACBF. A plot (Fig. 6) of $(t/C)^{1/3}$ versus t as modeled in Eq. 7 gives a straight line with a equals 0.0336 and b equals 0.8717. Correlation for all the parameters in equation is good ($R^2 = 0.9939$, see Fig. 6). Using Eq. 8 permits us to calculate k_1 which is equal to 0.23 g/kg h^2 and using Eq. 9 to calculate L for the GACBF to remove O&G from Ulu Pontian River water. This study finds that L equals to 6.53 g/kg. The results interpret that the capacity of the GACBF is able to remove about 6.53 g O&G per kilogram GAC from stream water since r equals 0.23 g/kg h. The lifetime for the GACBF was calculated to be about 59 days. The calculation model (Fig. 5b) is sufficiently accurate to permit the control of adsorption of O&G onto GAC from stream water.

Conclusion

This study developed the mathematical models for adsorption of O&G onto GAC from stream water. The models tested the data of monitoring the efficacy of the GACBF installed at mouth of the WIC during the 16-day-period. Experimental results show that the performance of the GACBF to remove O&G from stream water is excellent. Functional adsorption equations accounting for accumulation rate, water quality, flow rate, and accumulation time were presented. All the parameters in equations have physical interpretation. Experimental data validation showed that the equations are sufficiently accurate. A new methodology for calculating the accumulation rate and adsorption capacity was proposed to contribute to clean technology and environmental contamination investigation and assessment. The models to calculate accumulation rate coefficient and adsorption capacity for the GACBF to remove O&G from Ulu Pontian River water were presented. The lifetime of the GACBF installed to remove O&G from the river water was determined.

Acknowledgment This article used the data monitored by Central Laboratory of the SAJ (local water supply company) from October 14, 2009 to November 2, 2009. Data and information provided by the company were greatly appreciated.

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