The use of lightweight expanded clay aggregate (LECA) as sorbent for PAHs removal from water

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ABSTRACT

Lightweight expanded clay aggregate (LECA) has been explored as a sorbent for the removal of PAHs (phenanthrene, fluoranthene and pyrene) from water. The efficacy of LECA as a sorbent for PAHs was assessed using contact time, mass of sorbent and sorption isotherms in a series of batch experiments. Maximum (optimum) sorption was reached at 21 h after which the amount of PAHs sorbed remained almost constant. Batch experiments were conducted by shaking a 100 ml solution mixture of individual PAHs (containing 0.02 mg/L) with LECA. The maximum sorption was 70.70, 70.82 and 72.12%, respectively for phenanthrene, fluoranthene and pyrene when a mass of 0.2 g of sorbent was used. There was an increase in sorption as a result of an increase in mass of sorbent until a maximum was reached at a mass of 4.0 g LECA with 92.61, 93.91 and 94.15% sorption of phenanthrene, fluoranthene and pyrene respectively. Sorption data were fitted to the linearised forms of the Freundlich and Langmuir isotherm models to determine the water-LECA partitioning coefficient. Physical sorption caused by the aromatic nature of the compounds was the main mechanism that governed the removal process while the hydrophobicity of the PAHs also influenced the sorption capacity. LECA can be used as an alternative method for aqueous PAHs removal.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of diverse organic compounds containing two or more fused aromatic rings of carbon and hydrogen atoms [1]. They are ubiquitous pollutants formed from the combustion and pyrolysis of organic matter such as coal, oil, wood and fossil fuels like diesel and petrol [2]. PAHs are also released during oil spills where their types and quantities depend on the nature of the petroleum involved. Lighter oils have small but detectable amounts of PAHs while the heavier petroleum products can have elevated levels [3].

PAHs exposure usually occurs in the form of mixtures rather than individual compounds [4]. PAHs are compounds of environmental importance because of their carcinogenicity and mutagenicity [5]. There is a recent global concern about the increase in PAHs contamination of water systems [6]. The PAHs detected in highest concentration in drinking water are fluoranthene, phenanthrene, pyrene and anthracene [7]. Their presence in surface water or groundwater is an indication of a source of pollution [8]. Due to their low solubility in water and high affinity for particulate matter, PAHs are not usually found in water in high concentrations. The concentrations of total PAHs in water vary between 2.5–9.0 ng/L in tap water, 1.0–10 ng/L in groundwater, 2.7–7.3 ng/L rainwater and about 10–830 ng/L in surface water [9]. The European Union, EEC Directive 98/83/EC has set a limit of 0.1 ng/L for total PAHs in drinking water [10].

PAHs are generally resistant to biodegradation and are not easily removed by traditional physicochemical methods like flocculation, coagulation, sedimentation, filtration or ozonation [11]. Methods like chemical or biological oxidation, ion exchange, electrolysis and solvent extraction can be complicated to operate and sometimes prove inefficient for removing trace levels of pollutants [12]. Sorption however has been proved to be effective for the removal of persistent organic pollutants, and activated carbon is extensively used for this purpose [6,13]. When compared to other methods of PAH removal, sorption is simple, convenient, easy to design and can be operated with very little technical know-how [14].

In the last decade, studies on the use of sorption methods for removing PAHs (and other organics) from water include the use of oil shale ash and mineral particles [15], commercially available fibrous sorbents like Zylon [16], immature clay, leonardite [6], kapok and cattail fibres [17], agricultural waste such as sugar cane baggase, green coconut shells, etc. [18], silica gel [19] and petroleum coke-derived porous carbon [20].

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Isotherms are generally used to predict and evaluate the sorption capacity of sorbents [21]. The Freundlich isotherm is the most widely used model for quantifying the sorption of dilute organic chemicals from aqueous solutions. This non-linear model is based on the assumption that the sorption system involves heterogeneous surfaces, limited sorption sites and potential energy interactions [22]. The Langmuir isotherm has also gained popularity due to its ability to fit a wide variety of sorption data quite as well as the Freundlich isotherm [23]. The Langmuir isotherm is based on the assumptions that sorption is limited to one monolayer where all surface sites are equivalent [24].

Lightweight expanded clay aggregate (LECA) is a special type of clay that has been pelletised at high temperature. As it is fired, the organic compounds in the clay burn off forcing the pellets to expand and become honeycombed while the outside surface of each granule melts and is sintered. The resulting ceramic pellets are lightweight, porous, chemically inert, have a neutral pH value and non-biodegradable. They are also non-combustible, have excellent sound and thermal insulation properties, contain no harmful substances, are resistant to frost and chemicals and will not break down in water. LECA filters have uniform pore distribution and are not significantly lost when backwashed. LECA is used in gardening to retain moisture in the soil, in concrete for building purposes and in water treatment facilities for the filtration and purification of municipal wastewater and drinking water [25–28]. There are no known disadvantages of LECA as a material for water treatment. Though LECA has successfully been used for removal of heavy metals in water [29], its use as a sorbent for aqueous PAH removal has not been reported.

This study investigates the sorption properties of LECA for the removal of PAHs like phenanthrene, fluoranthene and pyrene from water.

2. Materials and methods

2.1. Standards, solvents, reagents and materials

Phenanthrene (98%), fluoranthene (98%) and pyrene (98%) were obtained from Aldrich, St. Louis, MO 63103-USA. Analytical grade chemicals were used as received in all other cases. Analytical HPLC grade dichloromethane was used for extraction. The LECA used in this study was obtained from the Saint-Gobain Weber Company, Filtralite Department in Oslo, Norway.

The physicochemical properties of LECA and PAHs are presented in Tables 1 and 2 respectively.

2.2. Experimental procedure

2.2.1. Properties of LECA

The surface image (Fig. 1) and elemental composition of LECA were determined with a Zeiss Supra 55 VP FEG scanning electron microscope (SEM).

2.2.2. Treatment of LECA

In order to remove any contamination that might have occurred during the handling and transport of the LECA particles and to ensure the elimination of a possible complication in the interpretation of analytical results, LECA pellets were washed twice in methanol and rinsed with deionised water. They were air dried at room temperature and then baked in a pre-heated oven at 320 °C for 3 h. The pellets were then stored in a dry air-tight container.

2.2.3. Preparation of PAHs spike solution

Approximately 2.5 mg each of phenanthrene, fluoranthene and pyrene were dissolved in dichloromethane (DCM) to the mark of a 25 ml volumetric flask. The resultant standard solution had a

Table 1
Typical characteristics/properties of LECA.

Parameter	Value
Particle size (mm)	1.5–2.5
Surface area (m^2/m^3)	~2200
Specific surface area (m^2/kg)	~2.93
Bulk density (kg/m^3)	750
Particle density	1600
Fire resistance	High
Thermal conductivity coefficient (W/m K)	0.07–0.10
Resistance to acids	High
Resistance to alkali	High
pH resistance	High
SiO_2 (%)	62
Al_2O_3 (%)	18
FeO_3 (%)	7
CaO (%)	3
MgO (%)	3
K_2O (%)	4
Na_2O (%)	2
C (total) (%)	0.02
Loss on ignition	1.36

Source: Saint-Gobain Weber Company, Filtralite Department in Oslo, Norway and Dansk Leca A/S, Randers, 8900-Denmark [30,31].

concentration of ~0.1 mg/ml of each of the three compounds. An aliquot of 0.2 ml was transferred into a 1000 ml volumetric flask and made to the mark with distilled water to give a final concentration of 0.02 mg/L (DCM is highly volatile and 0.2:1000 ml of water is negligible and does not contribute to solvent effect). The resultant spike solution was transferred into a stoppered borosilicate bottle and shaken at 300 rpm for 2 h to ensure a homogeneous distribution of the PAHs. Aliquots of 100 ml each of the resultant aqueous solution were used for the LECA sorption experiments.

2.2.4. Extraction and analyses of PAHs

After each batch experiment, the spiked water was decanted and centrifuged. Determination of levels of PAHs in water before and after treatment with LECA was performed by liquid–liquid extraction. PAHs were extracted by shaking with dichloromethane, first with 3 ml at 300 rpm in a borosilicate glass for 30 min, then 4 ml and 3 ml respectively in a separating funnel. The dichloromethane extracts were combined and dried with anhydrous Na_2SO_4 before GC–MS analysis.

GC–MS analysis was carried out using Agilent Technologies 7820B GC/5975 MSD system with an Agilent 190915-433 capillary column (30 m × 250 μm × 0.25 μm). Samples were injected at
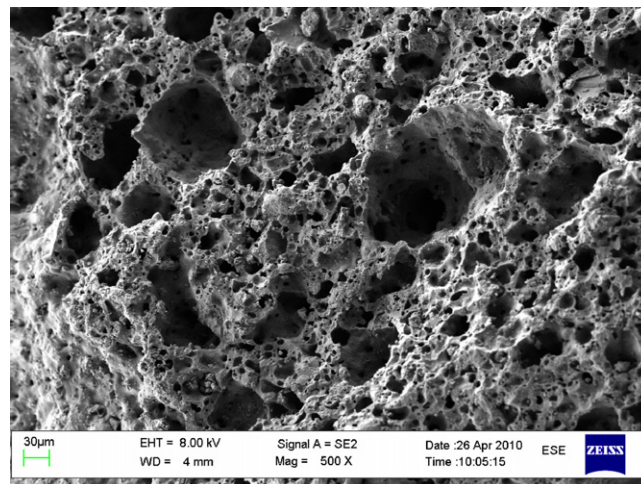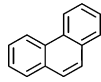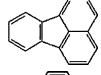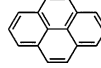

Fig. 1. Surface image of a LECA particle under a scanning electron microscope (SEM) showing pores.

Table 2
Physicochemical properties of PAHs used in the study.

Compound	MF	MW	Mp °C	CP	log K_{ow}	Vp, torr	S_{H_2O} (mg/L)	Chemical structure
Phenatherene	$C_{14}H_{10}$	178.23	110	3	4.57	6.8×10^{-4} at 20 °C	1.6 at 25 °C	
Fluoranthene	$C_{16}H_{10}$	202.25	111	3	5.22	10^{-6} to 10^{-4} at 20 °C	0.265 at 25 °C	
Pyrene	$C_{16}H_{10}$	202.25	149–156	3	5.18	6.9×10^{-9} at 20 °C	0.135 at 25 °C	

Adopted from, IARC: International Agency for Research on Cancer; US EPA: US Environmental Protection Agency.
MF: molecular formula, MW: molecular weight, Mp: melting point, Vp: vapour pressure, CPS: solubility.

a rate of $1 \,\mu l \,min^{-1}$ by pulsed spiltless injection and helium was used as the carrier gas at a constant flow rate of $1.2 \,ml \,min^{-1}$. The oven program was started at 50 °C and held for 1 min, increased at a rate of $15 °C \,min^{-1}$ up to 300 °C and held for 2.5 min. Mass detection was operated in a SIM mode with MS source temperature at 230 °C. Quantification was done using calibration curves of standard phenanthrene, fluoranthene and pyrene which gave R^2 values of 1.000, 0.999 and 1.000, respectively.

A recovery of 90.1% was obtained for the extraction method as judged by the results of spiking.

2.2.5. Effect of contact time on sorption

Batch experiments were performed with 100 ml aliquots of 0.02 mg/L solution. Aliquots were transferred into borosilicate glass bottles and 0.2 g aliquots of LECA were added. The bottles were covered with aluminium foil, screw capped and shaken for durations between 1 and 21 h at 300 rpm. For each batch, a blank containing only distilled water and LECA was performed. Extraction of PAHs remaining in the solution after each LECA interaction was done as described in Section 2.2.4.

2.2.6. Effect of mass of sorbent on sorption

Different masses of LECA between 0.2 and 4.0 g were shaken with 100 ml of 0.02 mg/L of PAHs spiked water for 21 h in each case and the residual aqueous PAHs were quantified.

2.2.7. Sorption isotherms

In order to determine the sorption capacity of LECA for removing PAHs from untreated water, solutions of PAHs (phenanthrene, fluoranthene and pyrene) of concentrations 0.0005, 0.0025, 0.005, 0.01 and 0.02 mg/L were shaken with 0.2 g each of LECA for 6 h (the time period during which the most rapid sorption occurs, see Section 2.2.5). These concentrations are lower than the solubility of the individual PAHs in water (Table 2). The amount of PAHs before and after LECA interaction was determined as in Section 2.2.4.
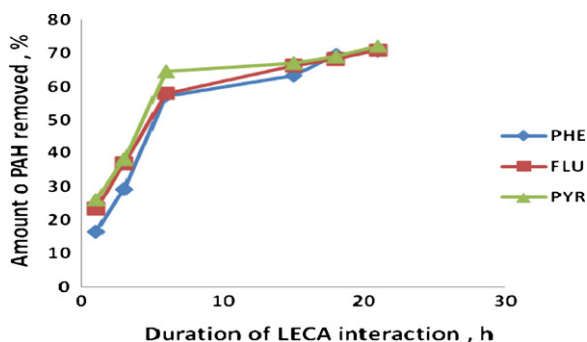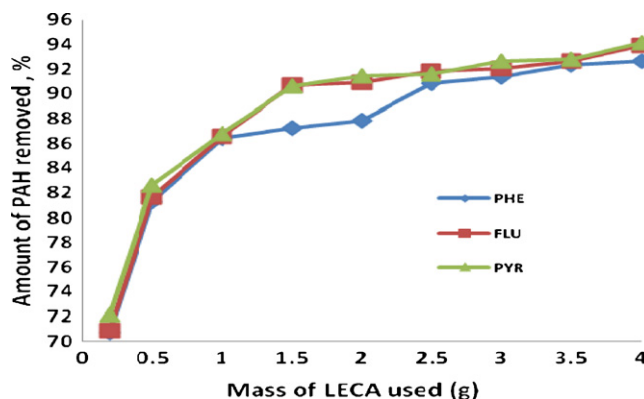

Fig. 3. Amount of PAH sorbed versus mass of LECA at 21 h.

3. Results and discussion

The size of the LECA used was in the range of 1.5–2.5 mm. Elemental analysis with Zeiss Supra 55 VP FEG SEM showed the presence of C, O, Na, Mg, Al, Si, K, Ca, Ti and Fe. The SEM surface image shows a rough surface with many pores (Fig. 1).

The outcome of the sorption studies with LECA are presented in Fig. 2 (PAHs sorbed versus contact time), Fig. 3 (amount of PAHs sorbed versus mass of LECA at 21 h), Fig. 4 (Maximum amount (%) of PAHs removed at maximum time of 21 h) and Fig. 5 (the linearised form of the Freundlich and Langmuir sorption isotherms). Table 3 shows the Freundlich and Langmuir parameters for the PAHs studied.
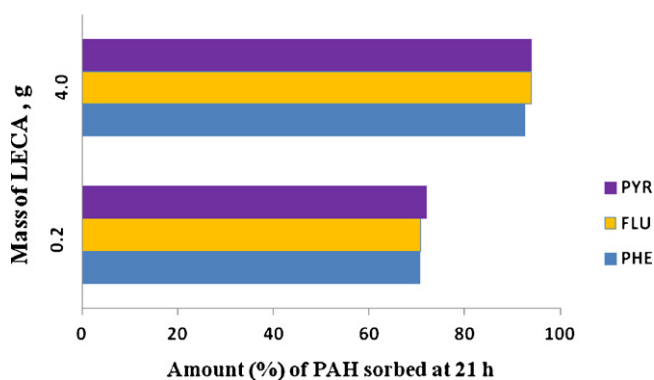

Fig. 2. PAH sorbed versus contact time.

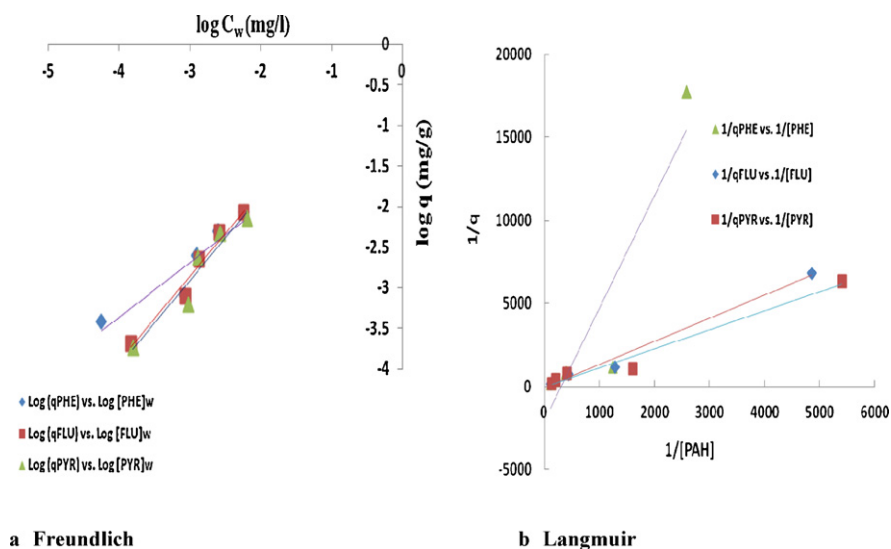**Fig. 4.** Maximum amount (%) of PAHs removed at 21 h.

Fig. 5. Sorption isotherms of phenanthrene, fluoranthene and pyrene on LECA.

3.1. Effect of contact time on sorption

When batch experiments were conducted to determine the rate of uptake of PAH by LECA at room temperature (25 °C), it was observed that the rate of sorption increased with contact time (Fig. 2). The results show that all PAHs were removed at a rapid rate during the first 1–6 h, by which time approximately 60% of all PAHs had been removed (57.3, 57.8 and 64.6% of phenathrene, fluranthene and pyrene respectively). This was the basis for the selection of a 6 h duration for the isotherm experiments. After 6 h, the uptake efficiency decreased gradually as sorption increased until it reached a maximum at 21 h. At this time, 70.7, 70.83 and 72.12% of phenathrene, fluoranthene and pyrene respectively had been removed. Considering the similarity of the sorption curves, it is very clear that molecular weight is not a determinant for the rate of sorption. Sorption occurred in two stages regardless of molecular weight, a fast interaction in the first 6 h and a slower one for the remaining period. The main sorption mechanism seem to be based on a fast attraction of the PAH with high hydrophobicity or low polarity towards the hydrophobic sites of the LECA surface immediately on exposure. This mechanism was enhanced with contact time. Hence PHE was less removed at the maximum sorption time than FLU and PYR which had similar sorption patterns with contact time. This similarity could be attributed to their being structural isomers. Only 70.7% of PHE had been removed at 21 h, while 70.83 and 72.12% of FLU and PYR had been removed (Figs. 2 and 4). This trend is consistent with sorption mechanisms that are driven by PAH hydrophobicity. Similar trends for sorption processes based on hydrophobicity of PAHs have been reported in a case where leonardite was used as sorbent [6] and also when lignin-based sorbents were used [18].

3.2. Effect of sorbent mass on sorption

The sorbent mass was varied for the maximum sorption time of 21 h for a 100 ml solution containing 0.02 mg/L of PAHs, and the removal of PAHs was improved from masses of 0.2 g to 4.0 g. In the time series for a mass of 0.2 g, there was no futher sorption beyond 21 h, although a very rapid sorption was observed for 0.5 g of sorbent at 81.24, 81.58 and 82.57% (PHE, FLU, PYR) to 2.5 g at 90.88, 91.82 and 91.59% (PHE, FLU, PYR) (Fig. 3). The removal process proceeded gradually from this point until a maximum was reached at 92.61, 93.91 and 94.15% for 4.0 g of sorbent. Further removal was impossible beyond this point. Several results which support the increase in sorption with mass of sorbent have been reported for different systems including the use of 1-amino-2-naphthol-4-sulfonate as a selective solid-phase sorbent for the extraction of gold [32] and the use of green coconut shells as sorbent for the removal of toxic metal ions [33]. However, these are inorganic materials and may have different sorption mechanisms.

3.3. Sorption isotherms

In a solid–liquid system where effective interaction occurs, the uptake of PAHs consists of the following steps (i) diffusion transport of molecules from the solution to the exterior surface of sorbent through a boundary layer, (ii) intra-particle diffusion of molecules through the interior pores of sorbent and (iii) sorption of molecules onto the active sites on the interior surface [20]. In this study, the linear forms of the Freundlich and Langmuir isotherms were used in plotting the graphs in Fig. 5. Regarding the Freundlich isotherm, reasonable correlation coefficients of 0.86, 0.97 and 0.92 were obtained for phenanthrene, fluoranthene and pyrene respectively (Table 3). The equation is given as $\log q = \log K_f + 1/n \log C_w$; where q is mass of sorbed pollutant per unit of sorbent (mg/g), C_w (mg/L) is the dissolved PAH in water, n is the measure of linearity and K_f is Freundlich constant. The values of K_f and $1/n$ are obtained from the intercept and slope of the plot of $\log q$ versus $\log C_w$. K_f is related to the sorption capacity while n is related to the sorption intensity. The Freundlich isotherms in Fig. 5a can be considered as barely nonlinear for Flu and Pyr and explicitly nonlinear for Phe

Table 3
Freundlich and Langmuir parameters for phenanthrene, fluoranthene and pyrene.

PAH	Freundlich			Langmuir			
	K_f (mg/g)	n	R^2	b (L/mg)	Q_0 (mg/g)	R^2	R_L
Phenanthrene (Phe)	0.22	1.48	0.86	−312.50	$−4.75 \times 10^{-4}$	0.85	−0.19
Fluoranthene (Flu)	2.02	0.95	0.97	−53.19	$−1.35 \times 10^{-2}$	0.99	−15.67
Pyrene (Pyr)	1.70	0.95	0.92	−41.67	$−2.10 \times 10^{-2}$	0.97	6.00

with n values (Table 3) outside the range of $0.95 < n < 1.05$, as within these boundaries, the isotherms are considered linear [34].

When $n = 1$, a similarity is indicated to the Langmuir isotherm, while $n \geq 10$ indicates that heterogeneous sorption is favoured [19]. This is also confirmed by the heterogeneous surfaces of LECA particles (Fig. 1). Nonlinear isotherms have also been obtained for several PAHs using sorbents such as aspen wood, leonardite, black, carbon-free biopolymers, coconut shell, sugar cane, etc. [18,35]. Sorption coefficients of 0.22, 2.02 and 1.70 were obtained for PHE, FLU and PYR respectively.

In the case of the Langmuir isotherm (Fig. 5b), correlation coefficients of 0.85, 0.99 and 0.97 were obtained for phenanthrene, fluoranthene and pyrene respectively. These figures are very similar to those obtained for the Freundlich isotherm (Table 3). The linear equation is given as $1/q = 1/Q_0 b \times 1/C_w + 1Q_0$, where q and C_w have the same meanings as above; Q_0 is the Langmuir constant analogous to maximum amount of sorption corresponding to complete monolayer coverage (mg/g) and b is the Langmuir constant which is related to the rate of sorption intensity [36]. The values of b and Q_0 (Table 3) are obtained from the intercept and slope of the plot of $1/q$ versus $1/C_w$. The Langmuir isotherm is usually interpreted with an equilibrium parameter R_L, given as $R_L = 1/(1 + bC_0)$ where C_0 is the highest PAH concentration (mg/L), in this case 0.02 mg/L. The R_L values obtained for PHE, FLU and PYR are -0.19, -15.67 and 6.00, respectively. These R_L values do not favour Langmuir predictions since when $R_L > 1$ (unfavourable), $R_L = 1$ (linear), $R_L = 0$ (irreversible) and $0 < R_L < 1$ is favourable [19]. Therefore results of our study indicate that the sorption of PHE, FLU and PYR by LECA are more similar to the Freundlich than the Langmuir model. Regarding the lack of correspondence of the isotherms, it has been reported that the sorption of organic solutes in solution is generally dependent on various solute/sorbent interactions like Van der Waals and dipole interactions as well as weak intermolecular forces [37,38].

3.4. Comparison of the performance of LECA with other sorbents

When the fibrous synthetic sorbent p-phenylene-2,6-benzobisoxazole sold under the trade names Zylon and PBO was used for aqueous PAH (including phenanthrene and pyrene) sorption studies, an average of 95% removal was achieved in 24 h when an average of 351.95 mg of fibre was shaken in batch experiments with 20 ml aliquots of 50 μg/L of solution. The mass of 10 cm of PBO fibre (containing 322 filaments) was equated to 0.01 g. The maximum sorption of 95% can be presented as 2.7 μg/g removal [16]. Also a study involving the use of natural coal leonardite as sorbent (0.5 g of sorbent to 0.5 L of solution) for the removal of 5 PAHs including pyrene and benzo[k]fluoranthene resulted in 82% removal of a 100 μg/L solution in 24 h equilibrium time. This figure can be presented as 82 μg/g removal [6]. In the case of LECA, the 92.61, 93.91 and 94.15% (Section 3.2) removal of PHE, FLU and PYR from 100 ml solution of 0.02 mg/L concentration by 4.0 g sorbent can be presented as 0.4631, 0.4696 and 0.4708 μg/g removal respectively at 21 h. However when 0.20 g of LECA was used, the 70.7, 70.83 and 72.12% removal of phenathrene, fluoranthene and pyrene respectively obtained can be presented as 7.070, 7.083 and 7.212 μg/g. The removal efficiency of PBO is lower while that of Leonardite is higher when compared to that of LECA. Among these three sorbents PBO is a speciality fibre with a generally high production and sale cost [39]. The price of leonardite is around US$5/Kg [40] and that of LECA is around 0.15 US$/Kg [41]. Though it may not be reasonable to compare prices from two different companies selling different types of sorbents, it appears that LECA is a much cheaper alternative. The low cost of LECA also affords the use of several folds of the particles to increase the sorption of

PAHs in field applications. Furthermore, LECA is heat resistant and therefore can be regenerated by incineration at high temperatures.

4. Conclusion

The study has revealed that phenanthene, fluoranthene and pyrene can be effectively removed from water by batch sorption experiments using LECA as sorbent. The extent of removal was enhanced with an increase in the mass of LECA from 70.7, 70.83 and 72.12% of PHE, FLU and PYR for a mass of 0.2 g of LECA to 92.61, 93.91 and 94.15% of PHE, FLU and PYR for a mass of 4.0 g of LECA for 100 ml aliquots of 0.02 mg/L solution. The extent of sorption also increased with contact time. The sorption of the PAHs tested followed the order PYR > FLU > PHE, which is consistent with the sorption mechanism driven by PAH hydrophobicity. The sorption isotherms followed the Freundlich model with correlation coefficients of 1.48, 0.95 and 0.95 for PHE, FLU and PYR respectively. The precision and accuracy of the methods used in the study were satisfactory with a recovery of 90.1%. The price of LECA compares favourably with other natural sorbents. LECA can thus be used as an effective alternative material for PAH removal from contaminated water.

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