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Evaluating Algibon adsorbent and adsorption kinetics for launderette water treatment: towards sustainable water management

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Abstract:

Grey water from commercial laundry facility was used for treatment to substitute the conventional water sources. Algibon, A800 derived from mesoporous alginic acid and Starbon S300, carbonaceous mesoporous polysaccharide-derived materials, silica gel (SG) and activated carbon (AC) were used for treatment of that wastewater. The optimum adsorbent dosing and pH value for each adsorption system are defined. The adsorption efficiency are in the order of A800 > SG > AC > S300 and the removal rate reached to 91% when A800 was used. Furthermore, the reaction followed the second-order kinetic model and the rate constant is high when A800 adsorbent is applied.

Key words: adsorption, COD, Kinetics, laundrette wastewater, Nano-material, Starbon

Introduction

Water is becoming a threatened resource in the world because of the gap between supply and demand. A study from the United Nations shows that water consumption is increasing at twice the rate of population growth and predicts that by 2025 an estimated 1.8 billion people will live in water scarce areas (Auffarth and Ledin 2002; Friedler and Hadari 2006; Friedler and Gilboa 2010). Population growth and increase urbanization are key factors that create this water-stress. Therefore, there is an increased interest in the reuse of

wastewater in many parts of the world, including both industrialized and developing countries.

Not solely the environmental issues are the driving force for reusing of wastewater, but there are also economic considerations. Reuse of water will lower the total cost of wastewater handling, as it will reduce the load of water sent to treatment plants. Now is an opportune time to focus on optimising water use by substitution of fresh water with alternative water resources.

One alternative source of water is grey water. Grey water is defined as: non-industrial wastewater generated from domestic processes such as washing dishes, laundry and bathing without any sewage input (Christova-Boa and Eden 1996; Emerson 1998).

In-house water demand represents 30 to 60 % of the urban water. However, approximately 15 to 23 % of the total water consumption in the household is utilized in cloths washing. After clothes washing detergents and bleaches, plus on occasion oils, paints and solvents are contaminants found in grey water (Friedler et al. 2005).

Various grey water treatment processes are suggested in the literature, but using green adsorption techniques is a relatively new practice. Most treatment units reported in the literature are based on physical processes such as filtration and disinfection, whilst newer technologies incorporate biological treatments as well (Diaper et al. 2001; Ogoshi et al. 2001; Kanawade 2015; Taskin et al. 2016). However, these conventional methods have several disadvantages, namely, high capital and operational costs (Sostar-Turk et al. 2005).

Adsorption using solid adsorbents is one of the most efficient methods for removal of organic contaminants during wastewater treatment. Its main advantages are simplicity of design and cheap start up and running costs. There are a wide variety of natural materials that can be utilised as adsorbents and in recent years, the search for low-cost materials that have pollutant-binding capacities has intensified.

Recently, novel bio-derived mesoporous materials, Starbon have been developed (Budarin 2007). They have demonstrated extraordinary properties in the adsorption of organic molecules (i.e. dyes and phenols (Parker et al. 2012; Parker et al. 2013). Recently published work also showed that S300 (starch derived Starbon prepared at 300ºC) was effective for the treatment of laundry wastewater (Tony et al. 2016). However, Starbon can also be prepared from alginic acid, resulting in materials with slightly altered textual properties. These materials have not yet been tested in the treatment of 'real-life' wastewater, i.e. laundry wastewater.

This work will focus on the treatment of launderette wastewater using different range of alginic acid derived Starbon adsorbents. To determine the efficiency of these materials they will be compared against commercially available activated carbon (AC) and Silica gel (SG). The effects of adsorption parameters, such as, adsorbent dose and pH, are also investigated.

Materials

Grey Water

Grey water was collected from the wastewater outlet of a commercial laundrette service in the city of York, United Kingdom. This laundry system is operated with a standard washing machine programme using a powder detergent that includes three main ingredients: pH control/salts, water softeners and surfactant cleansers. Samples were collected containing high organic matter content obtained from the first wash cycle and analyzed before treatment. The main characteristics of wastewater are pH 7.4, suspended solids 34 mg/L and COD (704 mg/L).

Adsorbents

Alginic acid derived adsorbents: A300, A450 & A800

Starbon can also be prepared from alginic acid. Full preparation of this material can be found in Parker et al. (2013). Synthesis is similar to starch-derived Starbon, although no *p*-toluenesulfonic acid is required. Alginic acid derived adsorbents used in this study were prepared at a range of temperatures: 300 °C (A300), 450 °C (A450) and 800 °C (A800).

Further adsorbent:

Other commercially available porous materials were used for comparison: activated carbon (AC) Norit® purchased from Fluka and Silica gel (SG), high purity grade with a pore size of 60A and particle size 35-75 μm, purchased from Sigma-Aldrich.

Methodology

Adsorption experiments were carried out in batch mode by pouring 20 mL of grey water into a vessel, adsorption added and the mixture allowed to stir for the desired amount of time (Figure 1). The treated wastewater was sampled at regular time intervals to determine COD removal efficiency. In the case of the experiments where the effect of the pH was examined, after adsorbent addition the pH was adjusted using H_2SO_4 or NaOH solution, and the solution was allowed to stir for the required time.

Analytical Determinations

The wastewater substrate concentration was measured by its Chemical Oxygen Demand (COD) using the standard methods (APHA, 1998). A Jenway pH meter (3505) Japan was used for pH measurements of the wastewater. Suspended solid is determined according to the standard methods (APHA, 1998).

Results and Discussion

A300, A450 & A800 Adsorbent Testing

COD removal efficiency of a range of alginic acid derived Starbon was determined. Materials prepared at three different temperatures 300 °C (A300), 450 °C (A450) and 800 °C (A800) were used. Results of COD removal with time revealed that the high temperature prepared material was the most effective at removing COD compared to the lower temperature materials. As shown in Fig. 2 (A) A800 gave the highest percentage COD removal from the launderette wastewater at 90.9% and in the fastest time, followed by A450 (77.2%), whilst A300 gave the least adsorption (72.7%). It has to be mention that the increase in the preparation temperature of the porous material, Algibon, results in a change in its textural properties. Therefore, an increase in S_{BET} (Brunauer–Emmett–Teller surface area) from 216 to 349 m^2/g between A300 and A800 is observed. Furthermore, the BJH (Barrett, Joyner, Halenda) pore volume also increased from 0.9 to 1.0 $(Cm^3/g)_{BJH}$ for A300 and A800. Thus, A800 recorded the highest removal percentage as it having the highest pore volume giving higher capacity and large pore diameter leading to quick and easy diffusion into available pores (White et al., 2010). It could also be due to the surface functionality, A800 has lower surface oxygen content and as a result is more hydrophobic than A300 and A450. The hydrophilic nature of the lower temperature adsorbent can enhance water molecule adsorption onto the adsorbent surface, thus creating competition for adsorption sites with the

organic pollutants.

The effect of solution pH on COD removal was investigated. The pH of the wastewater solutions was adjusted over the range of pH 2.0 to 8.0 including the natural pH of the grey water (pH 7.4). A300 and A450 showed big variability in COD removal rate with changes in pH of the wastewater. At pH 2.0 the removal rate was as low as 4.5%, this increased to 59.0% at pH 6.0 and then again to 72.7% at pH 7.4. Interestingly, A800 only exhibited a slight reduction in COD removal at acidic pH, with the optimum pH was also shown to be the natural pH of the laundry wastewater, pH 7.4. These differences in adsorbent behaviour are likely due to different interactions of the acidified water with the adsorbent surface.

According to the previous studies (Alatalo et al. 2016), zeta potential (as a measure of surface charge) of the carbonaceous mesoporous adsorbent material, Algibon, has been observed to become increasingly more negative as the pH value is increased. Since, in the acidic pH, excess protons (H^+) is increased on the adsorbent surface (A800) suggests a competition of the H^+ ions with the organics molecules in the grey water. Consequently, repulsive forces between the Algibon adsorbent and adsorbate are occurred. Therefore, the adsorption is reduced. By contrast, pH increase results in de-protonation of the active sites of the adsorbent, thus the negatively charged sites. Hence, the adsorption is increased (Bellona et al. 2004; Bandini 2005). The increase in the adsorption capacity by increasing the pH could be due to either electrostatic interaction and/or chemical reaction between the organics molecules in wastewater and the adsorbent A800. To illustrate, the large reduction in adsorption at basic conditions compared to acidic ones can be attributed to both hydrophobic interactions and charge interactions. At pH 7.4 a large electrostatic attraction exists between the positively charged adsorbent surface and the anionic wastewater. As the pH of the system increases, the number and strength of negatively charged sites increases and the number of positively charged sites decline which results in an increase in adsorption capacity of Algibon. Therefore, lowering the pH has the opposite effect leading to electrostatic repulsion occurring (Namasivayam and Kavitha 2002; Karim et al. 2009).

In order to determine maximum adsorption capacity tests were done with various adsorbent doses, whilst other parameters remained fixed. It is apparent from (Fig. 2 C) that by increasing the dose of the Algibon, the number of adsorption sites available for interaction is increased, resulting in the increased percentage of grey water adsorption capacity which is reached to 768, 898 and 920 mg-COD/g-A for A300,, A450 and A800, respectively at Algibon dose of 10 mg. However, the further increase of adsorbent dose more than 20 mg/20

mL does not get an enhancement of sorption capacity. This may be attributed to: (a) increased adsorbent dose relative to pollutant concentration and volume leading to not all adsorption sites becoming saturated (Shukla et al. 2002); (b) particulate interaction (i.e. aggregation) due to high adsorbent dose. Such aggregation would result in a decrease in available surface area and an increase in diffusional path length (Shukla et al. 2002). Those results of increasing the adsorption capacity with increasing the adsorbent amount are in agreement with the previous findings of Mittal et al. (2014).

Activated Carbon, Norit®, Adsorbent Testing

The feasibility of activated carbon (AC), Norit®, for the removal rate of launderette wastewater was assessed. For such studies, a 20 mg/20 mL AC was added to the launderette wastewater and the solution stirred for 120 minutes before sampling. Results showed that equilibrium was reached within 60 minutes (Fig. 3A), and COD removal reached 72.7%.

The effect of pH on the removal rate was also tested. Compared to the alginic acid derived materials, AC was seen to be more strongly affected by the acidic pH range, with no COD removal apparent at pH 2 or 3 (Fig. 3B). This loss in adsorption performance may be due to loss of active sites on the adsorbent surface at these strongly acidic pH (Cotea et al. 2012).

Under the selected condition for contact time (60 minutes), the amount of activated carbon was varied from 5 to 25 mg. Adsorption capacity reached a maximum of 512 mg-COD/g-AC. Raising the catalyst does above 20 mg/20 mL of wastewater, shows no further increase in the adsorption capacity. Using 20 mg of activated carbon gave the highest COD removal, (72.7%) and therefore, 20 mg of activated carbon was used in further studies.

Silica Gel Adsorbent Testing

Silica gel, SG, was also tested for comparison with the novel alginic acid derived materials. Firstly, the effect of contact time was examined. Results were similar to those of AC; adsorption equilibrium was reached within 60 minutes (Fig. 4 (A)) and uptake was inhibited at acidic pH (Fig. 4(B)). This pH affect is not in accordance with previous work by Cotea et al. (2012). Here it was observed that the adsorption of polyphenols by porous silica was favored in acidic medium. This could illustrate pH control of adsorption is related to the nature of the pollutants being removed (Wojnicki et al. 2013).

Further test for the SG adsorption was done to check the amount of SG needed for the treatment. Figure 4C illustrated that the increase in the SG from 5 to 20 decreases the adsorption capacity efficiency, which is best at 5 mg of SG. However, the removal rate increased with increasing the doses of SG from 5 to 20 mg/20 mL wastewater to reach to 81.8% COD removal; however, further increase does not get further removal. This means the tendency of adsorption decreased because of the saturation of the active pores is reached (Fu et al. 2008).

Discussion of Different Adsorbent Performance

It is noted from Fig. 5 that the efficiency of the adsorption of organics in the launderette wastewater are in the order of A800> SG>AC>S300. A800 is more efficient than S300, this is likely due to the increased mesoporosity of the material enabling good interactions with the polluting organic molecules. S300 has smaller pore diameters than A800 which could result in pore blockage due to aggregation of bulky molecules around pore entrances (Valix et al. 2006). As a result the full surface area of the adsorbent cannot be utilized, reducing the effectiveness of adsorption.

For the adsorbents A800, SG and AC original pH of the wastewater results in the highest % COD removal. However, for S300 acidic pH gives maximum removal of 81.8% at pH 2 and 3. This could be due to the difference in the nature of the adsorbent surface that results from the use of different starting material and preparation temperature (Parker et al. 2012; Parker et al. 2013).

Overall these results demonstrate that the effective adsorption organics in launderette wastewater is dependent on both the pore structure and the surface functionality of the adsorbent (Parker et al. 2012).

Table 1 shows the comparison of adsorption capacities and removal efficiencies of launderette wastewater using Algibon (A800) adsorbent with other forms of adsorbents. The comparison provides some suggestion as to the use of the prospective adsorbent in real launderette wastewater treatment. However, the maximum adsorption capacity also depends on the initial pollutant concentration load. Comparison of A800 with the other range of adsorbents listed in Table 2 provides superior adsorption capacity (920 mg/g) and a removal efficiency (90.9%) to the other adsorbents. Though, several types of adsorbents exhibit higher removal efficiencies, which are reached to 98, 97 and 93% for Zeolite A4, natural Zeolite and GAC, respectively; it should be considered that those materials remove lower loads of

organics (1 mg/L for Zeolite A4 and natural Zeolite and 208 mg/L for GAC adsorbent) in launderette wastewater according to those references (Sostar-Turka et al. 2005; Fang et al. 2016). The removal efficiencies range is attributable to the variety of the organic loads and types that present in the different launderette wastewater. Those organic loads are contributed by the increase in availability of effective surface sites resulting from the increased adsorbent/adsorbate ratio (Gupta 2010; Park et al. 2010; Patil et al. 2011). Based on the high adsorption capacity, the present A800 adsorbent is favourable for green remediation.

Kinetic Studies for Grey Water Treatment

The kinetic study is considered an important index for organic matter removal from grey water valued by COD. Sorption kinetics of this grey water on the studied adsorbents is presented in Fig. 6. In order to investigate the mechanism on sorption, three common kinetic models are used to study the kinetics of sorption processes; the zero, first and second order reaction kinetics. The highest values of the regression coefficient (R^2) are used as an indication to interpret the most suitable model to describe the adsorption kinetics. Based on R^2 values (given from Fig. 6 and illustrated in Table 2), ranging from 0.35 to 0.72, from 0.53 to 0.78 and from 0.76 to 0.81 for zero, first and second orders, respectively, confirm the adsorption reaction obeying the second order nature since the correlation coefficient for the second order is the highest one for the four different adsorbent used. This may be illustrated by the electrons exchange between the sorbent and sorbate (Ho 2006). It is worth noting that similar investigation was described in the literature fitting the second order kinetic reaction for different adsorption techniques (Ho et al. 1999; Kula et al. 2008; Ben Hamissa et al. 2010; Ashour et al. 2014; Wang et al. 2015; Stoia et al. 2015).

Values of the kinetic constants, K_0, K_1 and K_2 for the three models, zero, first and second kinetic models are estimated from the plots (A), (B) and (C), respectively, in Fig. 6. All data found are presented in Table 2. The highest K_2 value is recorded 0.0001 $mgL^{-1}min^{-1}$ for A800 adsorbent. However, direct comparison of the K_2 values of the different adsorbent used in this work is not possible since there is a difference in the nature of those adsorbent. Additionally, half adsorption time, $t_{1/5}$, which is defined as the essential time for adsorption to take up to half of its equilibrium for organics removal from launderette wastewater; is calculated. This time is usually used as a measure for adsorption rate (Alzaydien and Manasreh 2009). The calculations of the half time reveal that the adsorption speed is very high in the zero order kinetic models and is lower than those values for the first order reaction model. However, it is

much smaller for the second order reaction models ranging only from 14 to 71 min. The type of adsorbent affects half adsorption time; A800 is demonstrating the lowest half time, which is 14 min for the second order model fit. Thus, the tendency of adsorption is at its highest rate when A800 is used for laundrette wastewater.

Conclusion

The present study demonstrated the adsorption of organics from laundrette wastewater using different adsorbent materials including commercial activated carbon and Sicilia gel besides three novel alginic acid derived materials, A300, A450 and A800. A800 is the most efficient adsorbent and is not affected significantly by changes such as pH of the water. The adsorption kinetics was studied and the reaction is well fitted with the second order reaction model. This material shows excellent promise for the simple, fast and effective treatment of grey water for reuse.

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Figure Caption:

Figure 1. Schematic diagram of a lab-scale adsorption test

Figure 2. Effect of (A) reaction time, (B) pH and (C) adsorbent dose on COD removal by A300, A450 and A800 from laundry wastewater.

Figure 3. Effect of (A) reaction time, (B) pH and (C) adsorbent dose on COD removal by AC from laundry wastewater.

Figure 4. Effect of (A) reaction time, (B) pH and (C) adsorbent dose on COD removal by SG from laundry wastewater.

Figure 5. Comparison of adsorption of grey water onto different adsorbent

Figure 6. (A) Zero-, (B) first- (C) second- order reaction kinetics for grey water adsorption using various adsorbents

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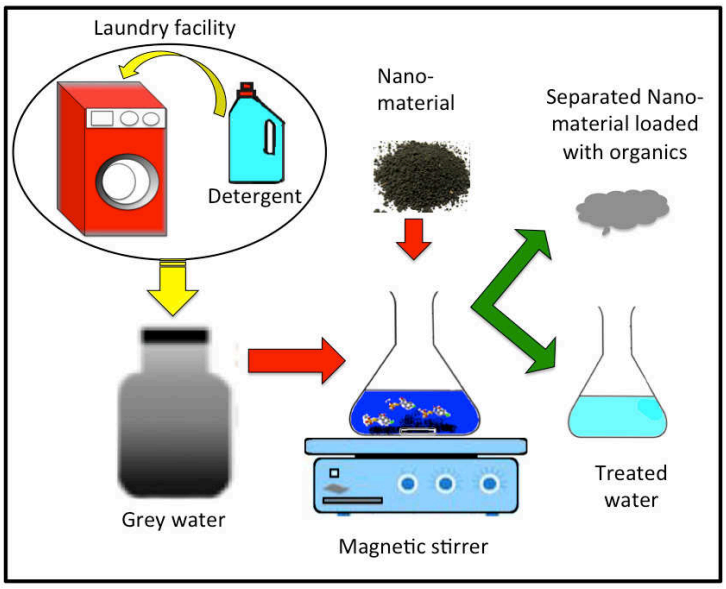


Fig.1

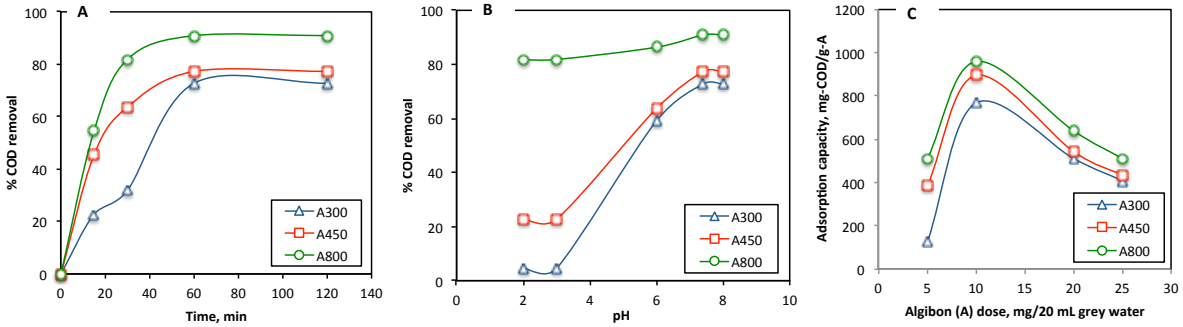
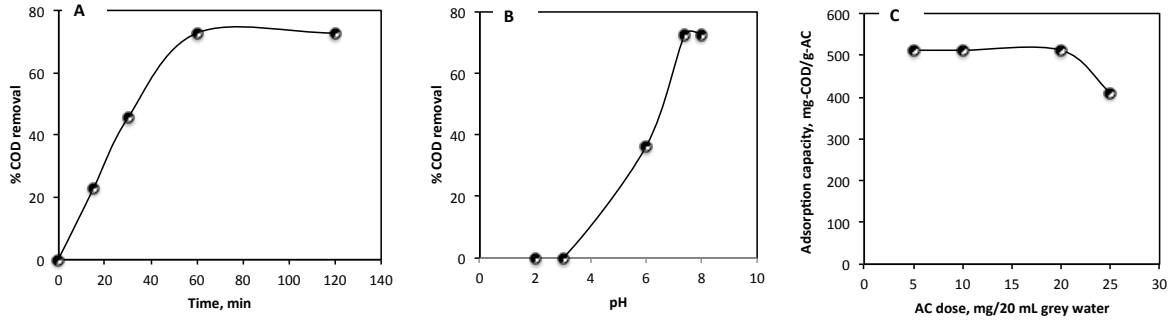


Fig. 2



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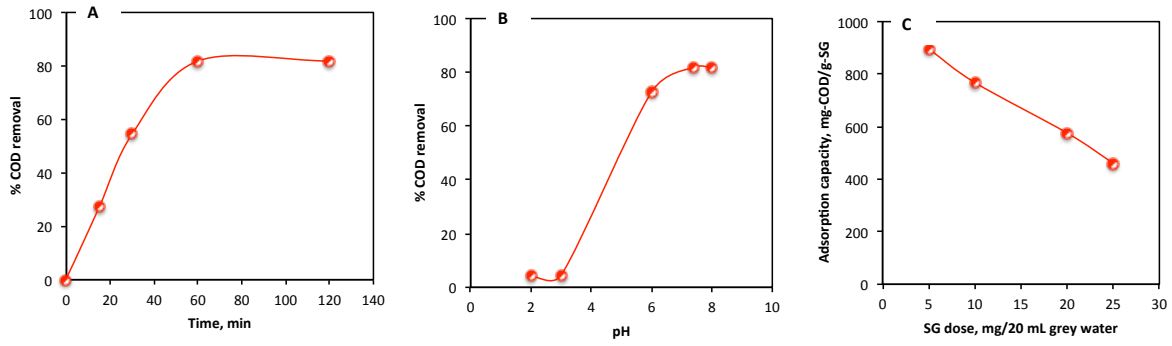

Fig. 4

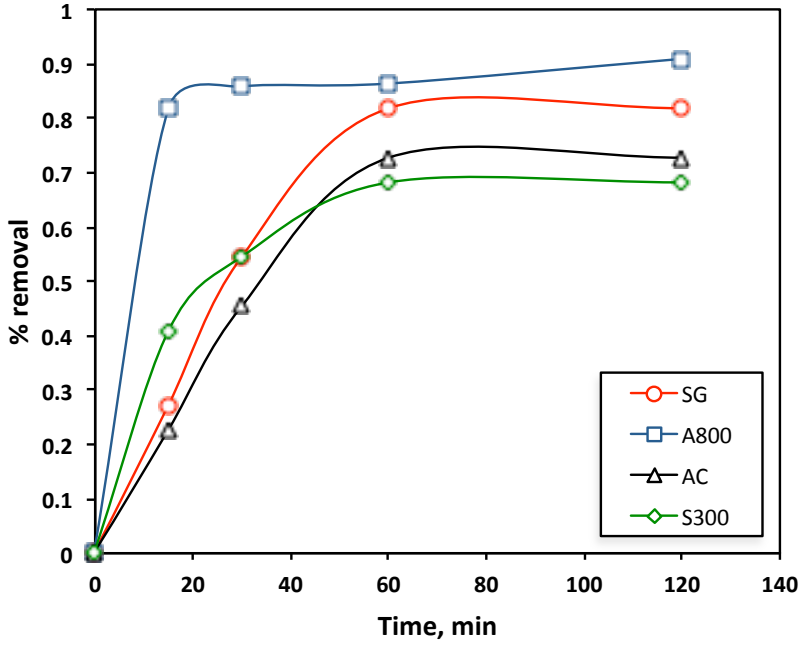

Fig. 5

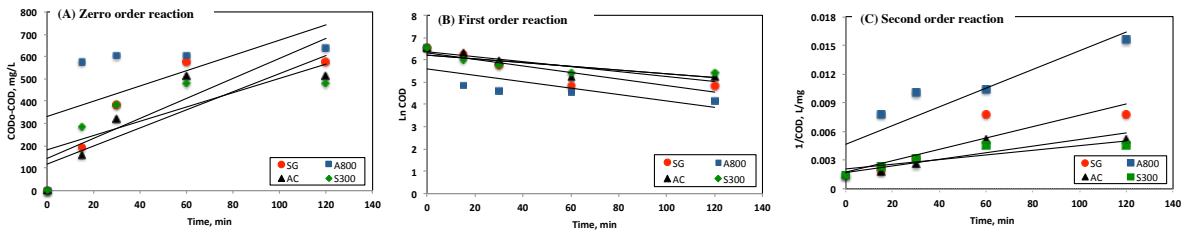

Fig. 6

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Table 1. Comparison of launderette wastewater adsorption capacities with different forms of adsorbents

Adsorbent	Surface area, m^2/g	Launderette wastewater			Adsorption capacity, mg/g	% Removal	Ref.
		Source	Temperature, K	Initial concentration			
A800	265	Commercial	298	704 mg-COD/L	920	90.9	Current work
SG	299	Commercial	298	704 mg-COD/L	896	81.8	Current work
AC (Norit)	798	Commercial	298	704 mg-COD/L	512	72.7	Current work
S300	332	Commercial	298	704 mg-COD/L	448	63.6	(Tony et al., 2016)
Natural Zeolite	48	Synthetic containing PO_4	-	2 mg-PO_4/L	-	90.0	(Agustina et al., 2014)
GAC	-	Hospital	335	208 mg-COD/L	-	93.0	(Sostar-Turka et al., 2005)
Vermiculite	-	Synthetic containing organics & clay		1 mg/L of Sr^{2+}, Cs^{+}, Co2+	18	75.0	(Fang et al., 2016)
Natural Zeolite	-	Synthetic containing Sr^{2+}, Cs^{+}, Co^{2+}, clay & sodium oleate	298	1 mg/L of Sr^{2+}, Cs^{+}, Co2+	23	97.0	(Fang et al., 2016)
Zeolite A4	-	Synthetic containing Sr^{2+}, Cs^{+}, Co^{2+}, clay & sodium oleate	298	1 mg/L of Sr^{2+}, Cs^{+}, Co2+	25	98.0	(Fang et al., 2016)

AMP-PAN*	32.69	Power plants	298	1329 mg-Cs/L	81	81.3	(Park et al., 2010)

*AMP-PAN: ammonium molybdophosphate-polyacrylonitrile

Table 2. Kinetic data of launderette wastewater adsorption with various adsorbents based on different kinetic models

Adsorbent	Zero order kinetics			First order kinetics			Second order kinetics		
	K_0, min^{-1}	$t_{½}$, min	R^2	K_1, L $Mg^{-1}min^{-1}$	$t_{½}$, min	R^2	K_2, mg L^{-1} min^{-1}	$t_{½}$ min	R^2
SG	4.48	1580	0.72	0.0147	47.10	0.78	0.00006	23.70	0.79
A800	3.42	1200	0.35	0.0144	48.10	0.53	0.00010	14.20	0.81
AC	4.05	1430	0.73	0.0111	62.40	0.78	0.00003	47.30	0.79
S300	3.20	1130	0.58	0.0008	845.0	0.68	0.00002	71.00	0.76