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ADSORPTION ISOTHERM STUDIES OF LEAD FROM AQUEOUS SOLUTION USING FLY ASH

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Abstract: Adsorption is one of the most widely applied techniques for environmental remediation. Burning pulverized coal in thermal power plants (TPPs) generates large amounts of fly ash (FA) that must be disposed of or otherwise handled; in an environmentally sound manner. Fly ash could be used as low cost adsorbent material for waste water treatment. Experiments were designed to investigate the removal of lead ions from aqueous solution onto the fly ash have been carried out at room temperature under the batch mode. The effect of various parameters such as agitation time, adsorbent dosage and initial concentration on the extent of removal of lead has been determined. The percentage removal of Pb (II) ions was found to increase with decrease in initial concentration and increase with increase in contact time and dosage of adsorbent. The adsorption process was fast and equilibrium was achieved in about 60 min of contact. The maximum removal of lead was 90.37% obtained at the adsorbent dosage 1.5 g/L. At an optimum agitation time of 60 min, the percentage removal of lead from the aqueous solution is decreased from 91.72% to 83.52% by varying lead concentrations in the aqueous solution from 20mg/l to 140mg/l. Experimental data were analyzed by model equations such as Langmuir and Freundlich isotherms and it was found Freundlich isotherm model best fitted well the adsorption data.

Keywords: Adsorption, Adsorbent, Adsorption isotherms, Lead, Fly ash.

I. INTRODUCTION

Environmental contamination by toxic metals is a serious problem worldwide due to their incremental accumulation in the food chain and continued persistence in the ecosystem. The heavy metals lead, mercury, copper, cadmium, zinc, nickel, chromium is among the most common pollutants found in industrial effluents. Even at low concentrations, these metals can be toxic to organisms, including humans. For instance, lead is a naturally occurring element and a metal which is extremely toxic and it has adverse effects as it accumulates in the body, especially for children and pregnant women. It can lead to behavior disorders, anemia, mental retardation and permanent nerve damage [10, 28]. Lead pollution primarily comes from manmade sources cars used gas with lead, lead smelters, production of storage batteries, metal processing plants and incinerators [13]. Food is the major source of lead intake that who are not occupationally exposed or have high concentration of lead in drinking water.

A characteristic of non-degradability of metals associated with the accumulation capacity in environmental compartments resulted in the development of a range of methods and materials capable of separating or removing species such as lead, copper, cadmium, nickel and other elements from natural waters and industrial effluents[6, 5, 31]. There are many conventional techniques to reduce environmental problems associated with industrial effluents. Some of these techniques are chemical precipitation, chemical reduction or oxidation, filtration, reverse osmosis, filtration, solvent extraction, membrane filtration and adsorption. Every technique has its own advantage and disadvantage. Now a day, adsorption technique remains widely preferred due to its simplicity of design and operation, cost effective, high efficiency and flexibility [14]. Fly ash is an adsorbent that is inexpensive and readily available. The application of fly ash for the treatment of industrial effluents is gaining attention as a cost effective and simple. As an adsorbent, this is a

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surface phenomenon that depends on the higher specific surface area, narrow particle size distribution and the porosity of adsorbent.

The present study deals with the utilization of low cost adsorbents such as fly ash for adsorption of lead metal ions from aqueous solution. The main aim of this research was to determine the effects of different parameters such as agitation time, adsorbent dosage and initial concentration of the solution which were effective at the removal of Pb (II) ions from aqueous solutions [1,2]

II. METHODS AND MATERIALS

2.1 Preparation of adsorbent

The fly ash used in this experiment was collected as a solid waste material of the power plant from “National Thermal power Corporation”, Vizag, Andhra Pradesh, India. The fly ash obtained from burning of coal was dried and sieved into different fractions using Rotap sieve shaker. The fly ash was preserved in glass bottles for use as an adsorbent.

2.2 Preparation of adsorbate

Stock solution of lead (1000 mg/L) was prepared by dissolving 1.6gm of 99% $Pb(NO_3)_2$ in 1L of double distilled water. The concentration range of lead prepared from stock solution varied between 10 to 100 mg/L. The pH of solution was adjusted with 0.1 mol l^{-1} NaOH and HCl. All the chemicals used were of analytical reagent grade[9].

2.3 Analysis

The concentration of lead in the solutions before and after equilibrium were determined by atomic absorption spectrophotometer (perkin elmer aa 110 model). The pH of solution was measured with a Hanna pH meter using a combination glass electrode [15].

2.4 Adsorption Experiment

In this experimentation the effects of various parameters such as agitation time, adsorbent dosage and initial concentration on adsorption were studied by fly ash. The pH of solution was maintained at a desired value by adding 0.1 M NaOH or HCl. The experimental procedure for determining various parameters was discussed [21]

2.5 Effect of agitation time and adsorbent dosage

50 ml of aqueous solution was taken in a 250ml conical flask and add 0.5g of adsorbent having the size of 52 μ m. This sample was shaken in an orbital shaker at room temperature for 1 hr. similarly eight samples were prepared in a conical flask adding 0.5g of adsorbent and exposed to varying agitation times (5min, 10min, 20min, 30min, 40min, 50min, 60min, 120min, 180min, and 240min). The samples were filtered with Whatman filter papers and filtrate was analyzed in Atomic Adsorption Spectrophotometer (AAS) to obtain final concentrations of lead[12,18]. The same experimental procedure was repeated with other adsorbent dosages (1.0 g, 1.5 g). The percentage removal of lead nitrate was calculated as

$$\% \text{ removal} = (C_o - C_i) \times 100 / C_o \quad \text{-----} \quad (1)$$

Graphs are plotted between the contact time and percentage removal of lead nitrate separately to identify the optimum agitation time. The effects of adsorbent dosage on adsorptions were also obtained at this optimum agitation time.

2.6 Effect of the initial concentration of the lead in aqueous solution

50ml of mixed solution containing 50mg/l of lead nitrate was taken in a 250ml conical flask and exposed to 1.5gm of 52 μ m adsorbent size. The flasks were kept in an orbital shaker for optimum contact time (1 hr) at room temperature (300° K). Then the sample was allowed to settle and filtered. The final lead concentration of the filtrate was determined

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by using AAS [7,19]. The same procedure was repeated for other initial concentrations of lead in aqueous solution (20mg/l, 40mg/l, 60mg/l, 80mg/l, 100mg/l, 120mg/l and 140mg/l).

III. RESULTS AND DISCUSSIONS

3.1 Effect of agitation time

Adsorption of lead was measured at a varying contact time for initial concentration of 100 mg/L. from the Fig.1 and table -1, the plot reveals that the lead removal is higher at the beginning; this is probably due to a large surface area of the fly ash being available at the beginning for the adsorption of lead. As the surface adsorption sites become exhausted, the uptake rate is controlled by the rate at which the adsorbate is transported from the exterior to the interior sites of the adsorbent particles [32,35]. Most of the maximum percent lead removal was attained after about 60 min of shaking time. The increasing contact time increased the lead adsorption and it remained constant after equilibrium was reached in 240 min for initial concentrations.

Table -1: Effect of agitation time on removal of Pb (II) using fly ash

S.No	Agitation time, t, min	% removal of lead		
		At 0.5g and 52 μ m	At 1.0g and 52 μ m	At 1.5g and 52 μ m
1	5	82.0761	86.9915	87.2117
2	10	83.1307	88.3334	87.3340
3	20	83.6917	88.9335	87.9631
4	30	84.1091	89.0325	88.1350
5	40	84.8139	89.1963	88.9616
6	50	84.8219	89.8974	89.2761
7	60	85.4217	90.2571	90.3716
8	120	85.4217	90.2571	90.3716
9	180	85.4217	90.2571	90.3716
10	240	85.4217	90.2571	90.3716

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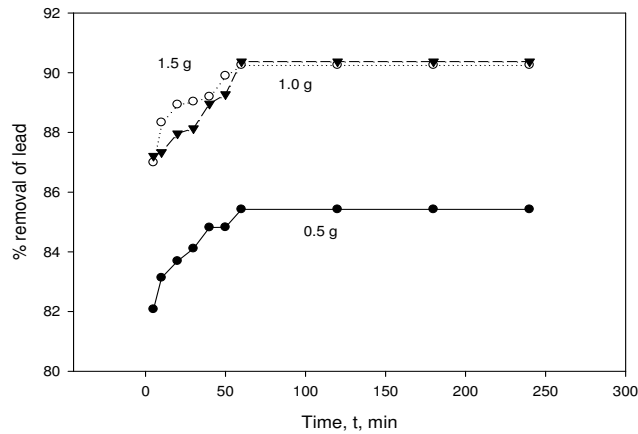


Fig. 1: Effect of agitation time on removal of Pb (II) using fly ash

3.2 Effect of adsorbent dosage

The effect of the adsorbent dosage was studied at room temperature by varying the adsorbent amounts from 0.5 to 1.5 g/L. For all these runs, the initial concentration of lead was fixed at 100 mg/L. Fig. 2 and table-2 shows that the adsorption of lead increases rapidly with an increase in the amount of fly ash due to greater availability of the surface area at higher concentrations of the adsorbent. A significant increase in uptake was observed when the dose was increased from 0.5 to 1.5 g/L. From the results, it is revealed that, within a certain range of initial metal concentration, the percentage of metal adsorption fly ash is determined by the adsorption capacity of the fly ash [22]. The maximum removal of lead was 90.37% obtained at the adsorbent dosage 1.5 g/L.

Table -2: Effect of adsorbent dosage on removal of Pb (II) using fly ash

S.No	Adsorbent dosage, w, gm	Final concentration of lead, C _i , mg/l	% removal of lead
1	0.5	14.5783	85.4217
2	1.0	9.7429	90.2571
3	1.5	9.6284	90.3716

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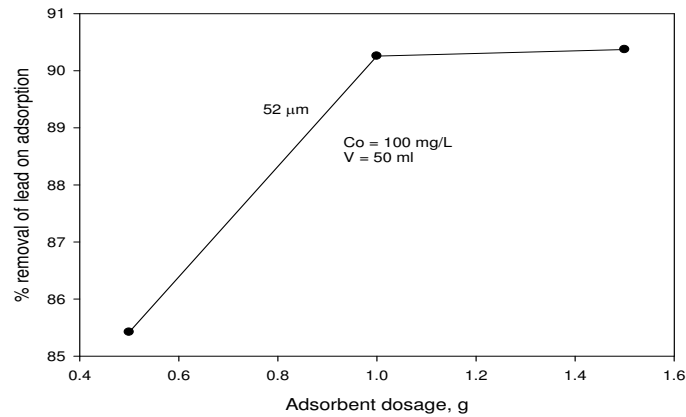


Fig.2: Effect of adsorbent dosage on removal of Pb (II) using fly ash

3.3 Effect of the initial concentration of the lead in aqueous solution

The effect of initial concentration of lead in the aqueous solution on the percentage removal of lead at equilibrium is shown in Fig.3 and table -3. At an optimum agitation time of 60 min, the percentage removal of lead from the aqueous solution is decreased from 91.72% to 83.52% by varying lead concentrations in the aqueous solution from 20mg/l to 140mg/l. It is noticed from the Fig.3 that at lower concentrations of Pb (II), fly ash adsorbs more amounts of Pb (II). Lesser percentage of Pb (II) is removed at higher concentration in the aqueous solution [16]. Such behavior can be attributed to the increase in the amount of adsorbate to the unchanging number of available active sites on the adsorbent (since the amount of adsorbent is kept constant).

Table -3: Effect of initial concentration on removal of Pb (II) using fly ash

S.No	Initial concentration of lead, C_o , mg/l	Equilibrium concentration of lead, C_e , mg/l	% removal of lead
1	20	1.656	91.72
2	40	3.673	90.8175
3	60	6.223	89.6283
4	80	9.2682	88.4147
5	100	11.5877	88.4123
6	120	15.812	86.8233
7	140	23.067	83.5235

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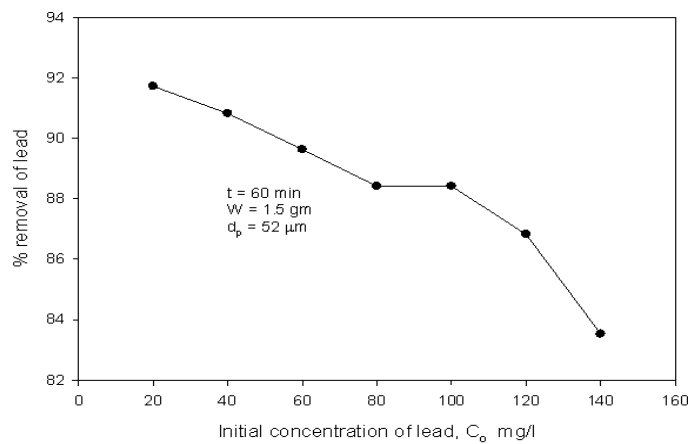


Fig.3: Effect of initial concentration on removal of Pb (II) using fly ash

Adsorption isotherms

3.4.1 Langmuir isotherm

Since chemical forces fall off very rapidly with distance, it is probable that chemisorptions does not extend beyond a single layer of adsorbate on the surface of the solid. It can be anticipated as first pointed out by Langmuir that chemisorptions' adsorbate layers may be only one molecule thick. Langmuir isotherm is the most widely used two-parameter equation. The relationship is a hyperbolic type form:

$$q_e/q_m = bC_e/(1+bC_e) \quad \text{--- (2)}$$

Where C_e is the concentration of the adsorbate at equilibrium, q_e is the amount adsorbed at equilibrium per unit mass of the adsorbent, q_m is the maximum amount adsorbed and b is the coefficient related to affinity. Equation (2) can be rearranged as

$$(C_e/q_e) = 1/bq_m + C_e/q_e \quad \text{--- (3)}$$

From the plots drawn in fig 4 between (C_e/q_e) and C_e , the slope $(1/q_m)$ and the intercept b/q_m are calculated from the table 4 .

Table- 4: Langmuir isotherm for adsorption of Pb (II) with respect to concentration using fly ash

S.No	Initial concentration of lead, C_o , mg/l	Equilibrium concentration of lead, C_e , mg/l	$q_e=(C_o-C_e)/m$ mg/gm	C_e / q_e
1	20	1.656	0.611	2.7103

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2	40	3.673	1.2109	3.0332
3	60	6.223	1.7920	3.4726
4	80	9.2682	2.357	3.9322
5	100	11.5877	2.947	3.933
6	120	15.812	3.473	4.5528
7	140	23.067	3.8978	5.9179

$$C_e/q_e = 0.468C_e + 2.061$$

$$R^2 = 0.899$$

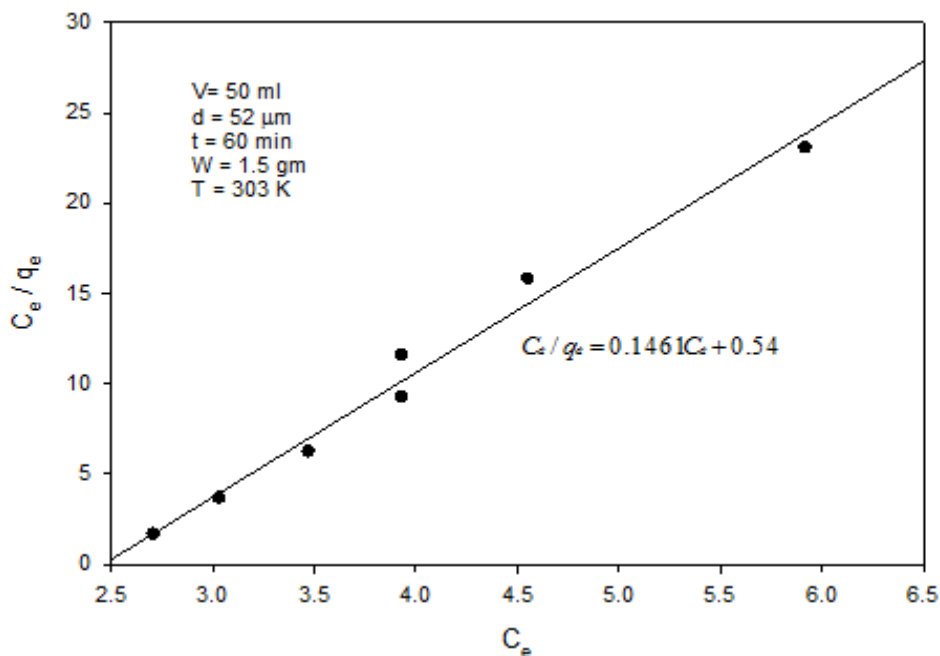


Figure 4: Langmuir isotherm for adsorption of Pb (II) with respect to concentration using fly ash

A further analysis of the Langmuir equation is made on the basis of separation factor, R_L defined as

$$R_L = 1/(1+bC_e) \quad \text{--- (4)}$$

If $0 < R_L < 1$

Favorable adsorption

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$R_L > 1$	Unfavorable adsorption
$R_L = 1$	Linear adsorption
$R_L = 0$	Irreversible adsorption

The equation obtained from the graph is:

$$C_e = 0.468 C_e / q_e + 2.061$$

$$R^2 = 0.899$$

The slope ($1/q_e$) is 0.468 and the intercept $1/bq_e$ is 2.061 are calculated.

The separation factor R_L obtained is 0.2419

In the present study, the Langmuir isotherm shows favorable adsorption indicating strong binding of Pb (II) to the surface of fly ash.

3.4.2. Freundlich isotherm

The Freundlich isotherm is given by

$$q_e = K_f C_e^n \quad \text{--- (5)}$$

Taking logarithms on both sides, we get

$$\text{Log } q_e = \text{log } K_f + n \text{ log } C_e \quad \text{--- (6)}$$

Where K_f and n are known as Freundlich coefficients obtainable from the plots of $\text{log } q_e$ versus $\text{log } C_e$ on the basis of the linear form of the equation (5) Freundlich isotherm is drawn between $\text{log } C_e$ and $\text{log } q_e$ in fig 5 using the data from table 5 (contains adsorption data of effect of initial concentration of Pb (II)).

Table- 5: Langmuir isotherm for adsorption of Pb (II) with respect to concentration using fly ash

S.No	Initial concentration of lead, C_o , mg/l	Equilibrium concentration of lead, C_e , mg/l	$q_e = (C_o - C_e)/m$ mg/gm	$\text{Log } C_e$	$\text{Log } q_e$
1	20	1.656	0.611	0.219	-0.2139
2	40	3.673	1.2109	0.565	0.0831
3	60	6.223	1.7920	0.7939	0.2533
4	80	9.2682	2.357	0.9669	0.3723
5	100	11.5877	2.947	1.0639	0.4693
6	120	15.812	3.473	1.1989	0.5407
7	140	23.067	3.8978	1.3629	0.5908

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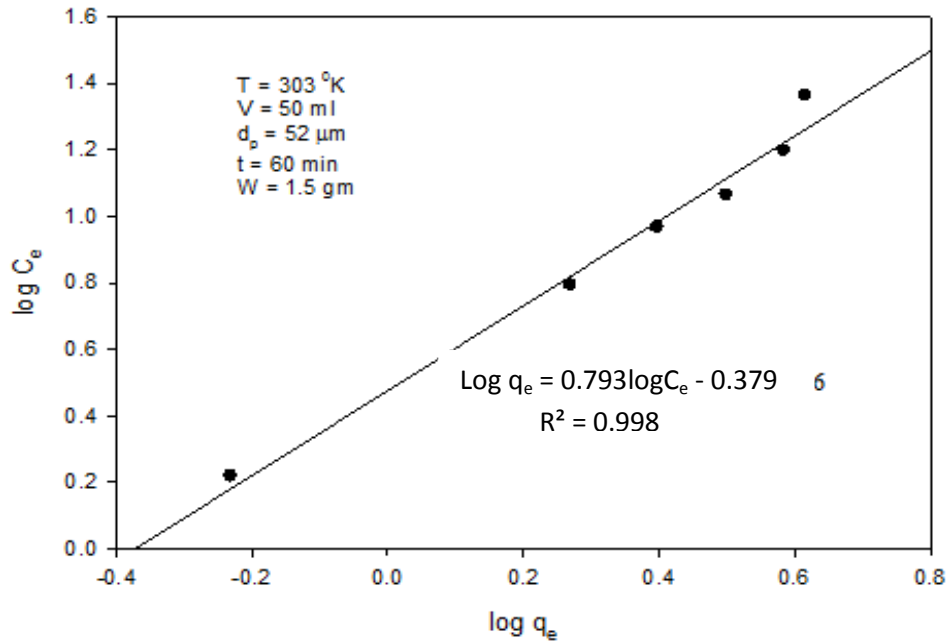


Figure 5: Freundlich isotherm for adsorption of Pb (II) with respect to concentration using fly ash

The following equations are obtained from the graph: $\text{Log } q_e = 0.793 \text{log } C_e - 0.379$

$$R^2 = 0.998$$

For the present data, the slope (n) of the above equation is less than one, fulfilling the Freundlich isotherm condition of $0 < n < 1$. Hence, favorable adsorption.

IV. CONCLUSION

Studies on kinetic parameters for adsorption of heavy metal Pb (II) ions from an aqueous solution using low cost and abundantly available adsorbent – fly ash. The optimum agitation time for the lead metal adsorption is 1hr for all concentrations. The percentage removal of lead from aqueous solution is augmented with increase with the adsorbent. Higher the concentration of lead in the aqueous solution decreases in the percentage removal of lead. In the range of variables studied, percentage adsorption is increased from 68.39% to 91.89%. The experimental data fulfills the Freundlich and Langmuir isotherm conditions indicating favorable adsorption of Pb (II) by fly ash and Freundlich is more favorable than Langmuir isotherm.

REFERENCES

- 1) Adie Gilbert, U., Unuabonah Emmanuel, I., Adeyemo Adebajo, A. and Adeyemi Olalere, G., Biomass and bioenergy. Vol 35, pp 2517-2525, 2011
- 2) Ali Akbar Safari Sinangani., and Hossein Mirahamdi Araki., Environ. Chem. Lett. Vol 8, pp 129–137, 2010
- 3) Antonio Zuorro., and Roberto Lavecchia., Am. J. Appl. Sci. Vol 7 (2), pp 153-159, 2010

International Journal of Innovative Research in Science, Engineering and Technology

(An ISO 3297: 2007 Certified Organization)

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- 4) Aswin Sidhaarth, K.R., Jeyanthi, J., and Suryanarayana, N., Euro. J. of Sci Res. Vol 2, pp 169-184, 2012
- 5) Bulut V.N., Arslan, D., Ozdes, D., Soylak, M., and Tufekci, M., J. Hazd. Mater. Vol 182, 331–336, 2010
- 6) Bulut, V.N., Duran, C., Gundogdu, A., Soylak, M., Yildirim, N., and Elci, L., J. Haz. Mater. Vol 76, 469–474, 2008
- 7) Ece Kok Yetimoglu., Melike Firlaka., Memet Vezir Kahramana., and Sabahattin Deniza., Polym. Adv. Technol. Vol 22, pp 612–619, 2011
- 8) Eligwe, C.A., Okolue, N.B., Nwambu, C.O., and Nwoko, C.I.A., Chem. Eng. Tech. Vol 22, 45-49, 1999
- 9) Gamze Guclu., Ebru Al., Serkan Emik., Tulin B. Iyim., Saadet Ozgumus., Mustafa Ozyurek., Polym. Bull. Vol 65, pp 333–346, 2010
- 10) Gavrilescu, M., Eng Life Sci., 4, pp 219-32, 2004
- 11) Gilberto., and Jorge Speece., J. Hazd. Mat. Vol 120, pp 115-121, 2003
- 12) Guilherme Ferreira., Laercio Caetano., Renata, S. D., Castro, Pedro Magalhaes Padilha., and Gustavo Rocha Castro., Clean Tech. Environ. Policy. Vol 13, pp 397–402, 2011
- 13) Guntae Son., and Seunghwan Lee., Kor. J. Chem. Eng., Vol 28(3), pp 793-799, 2011
- 14) Hengpeng Ye and Zhijuan Yu., Nat. Res., Vol 1, pp 104-109, 2010
- 15) Jyotsna Goel., Krishna Kadirvelu., Chitra Rajagopal., and Vinod Kumar Garg., J. Hazd. Mat. Vol 125, pp 211-220, 2005
- 16) Laxmipriya Panda., Bisweswar Das., and Danda Srinivas Rao., Kor. J. Chem. Eng. Vol 28(10), pp 2024-2032, 2011
- 17) Maria Martinez., Nuria Miralles., Soraya Hidalgo., Nuria Fiol., Isabel Villaescusa., and Jordi Poch., J. Hazd. Mat. Vol 133, pp 203-211, 2006
- 18) Maria Mar Areco., and Maria dos Santos Afonso., Colloids and Surfaces B: Biointerfaces. Vol 81, pp 620–628, 2010
- 19) Md. Salim., and Yukihiko Muneke., Int. J. Chem., Vol 1, 2009
- 20) Meng-Wei Wana, Chi-Chuan Kan, Buenda D. Rogel and Maria Lourdes P. Dalida., Carbohydrate Polymers., 80, 891–899 (2010).
- 21) Meski S., Ziani S., and Khireddine H., J. Chem. Eng. Data., Vol 55, pp 3923–3928, 2010
- 22) Miguel A. Aguilar-Gonzalez., Alexander V., Gorokhovskiy and Alfredo Aguilar-Elguezabala., Mat. Sci. and Eng. Vol 174, 105–113, 2010
- 23) Mobasherpour, I., Salahi, E., and Pazouki, M., Arab. J. Chem. 2011
- 24) Mohsen A. Hashem., Int. J. Phys. Sci. Vol 2(7), pp 178-184, 2007
- 25) Ofomaja, A.E., Unuabonah, E.I., and Oladoja, N.A., Biores. Tech. Vol 101, pp 3844–3852 2010
- 26) Onwu, F. K., and Ogah, S. P. I., Afr. J. Biotech. Vol 9(42) 7086-7093, 2010
- 27) Runping Hana., Zhu Lua., Weihua Zoua., Wang Daotong., Jie Shi., and Yang Jiujun., J. Hazd. Mat. Vol 137, pp 480-488, 2006
- 28) Sheng, P.X., Ting, Y.P., Chen, J.P., and Hong, L., J. Colloid Interface Sci., Vol 275, pp 131-41, 2004
- 29) Simos Malamis., Evina Katsou., and Katherine., J. Haralambous., Wat. Air Soil Pollut., Vol 218, pp 81–92, 2011
- 30) Smadar Elmachliy., Benny Chefetz., Elisha Tel-Or., Lorena Vidal., Antonio Canals., and Aharon Gedanken., Wat. Air Soil Pollut., Vol 218, pp 365–370, 2011
- 31) Soylak, M., Kaya, B., and Tuzen, M., J. Hazd. Mater., Vol 147, pp 832–837, 2007
- 32) Tikhomirova, T. I., Kubyshev, S. S., Sorokina, N. M., and Goloviznin, V. A., J. Anal. Chem. Vol 66(1), pp 2–5, 2011
- 33) Wei Song., Lei Zhang., Lei Shi., Da-Wei Li., Yang Li., and Yi-Tao Long., Microchim Acta., Vol 169, pp 321–326, 2010
- 34) Yunchul Cho., Suyeon Jang., Young Kim., Sridhar Komarneni., and Sungpyo Kim., J. Porous Mater. Vol 18, pp 125–131, 2011
- 35) Yunsong Zhanga., Weiguo Liub., Li Zhanga., Meng wanga., and Maojun Zhaoa., Appl. Surface Sci. Vol 257, pp 9809–9816, 2011
- 36) Yakup Arica, M., Cigdem Arpa., Aysun Ergene., Gulay Bayramoglu., and Omer Genc., Carbohydrate Poly. Vol 52, pp 167–174, 2003
- 37) Zhexian Xuan., Yanru Tang., Xiaomin Li., Yinghui Liu., and Fang Luo., Biochem. Eng. J. Vol 31, pp 160–164, 2006