

Environment Friendly and Low-Cost Method for Removal of NO₂ from Aqueous Media using Paper Pulp

S. R. Arthisree¹, G. Sowmyadevi², M. Sujatha², D. Sirisha¹

¹Department of Biotechnology, PSG College of Technology, Coimbatore, TN, India.

²St. Anns College for Women, Mehdiapatnam, Hyderabad, Telangana, 500028 India.

Corresponding author: sujatha.stanns09@gmail.com. Ph# 9866049923

Abstract:

An efficient and economic method to remove NO₂ from aqueous media using paper pulp has been developed. In this context, work has been carried out on analysis of NO₂ in laboratory and developing a control technology for its removal using news paper pulp as an adsorbent. Newspapers were collected from the scrap stalls and were used as an adsorbent. The tests are performed in terms of contact time, concentration, adsorbent dosage and temperature. FTIR spectral studies were carried out before and after adsorption of NO₂ aqueous solution on the paper pulp. Results revealed that NO₂ molecules are completely adsorbed. The developed method can also be implemented to remove NO₂ from air pollution.

Keywords: *Economic, Paper pulp, Adsorption, Contact time, Dosage, Aqueous solution, FTIR studies, Concentration, Air pollution*

Introduction

Air pollution is a major concern in India. It can damage ecosystems, threaten human health, and reduce well-being. The earth's climate system is affected by air pollution. Large-scale industrialisation and urbanisation leads to the significant increase in the number of industries, vehicles and traffic flow [1]. Air pollution causes eye irritation, lung cancer, asthma, bronchitis etc. Epidemiological studies have shown that there is an important relationship between air pollution and negative health effects.

The NO_x represent a family of seven compounds of nitrous oxides. Regulations are passed for NO_x from all the seven compounds as it generated by androgenic activities. NO₂ is pollutant itself and it initiates the reactions to form ozone and acid rain [1, 2]. The Nitrogen oxides are precursors for photochemical smog, acid rain and ozone accumulation. The oxides of Nitrogen–NO_x, are highly corrosive and hazardous to health. Due to the environmental concerns caused by air pollution, extensive research is being done to control NO_x pollution. Global weather patterns will shift and the Nitrogen dioxides trap heat in upper atmosphere due to its positive effects. It is becoming difficult to calculate nitrogen effects in climate models.

NO₂ abatement and control technology is relatively complex issue. This complex issue and situation has to be solved at global, national and at international levels by adapting to a

natural solution which will not bring any change in the environment [3]. Taking that unpleasant situation into the hands of the environmentalist a solution has to be framed. Various processes are investigated and proposed for controlling SO_x and NO_x . Among these adsorption process is simple, cost effective and highly reliable process. In this context work has been carried out on analysis of NO_2 in laboratory by developing a control technology using paper pulp as an adsorbent [4].

Methods and Materials

Selection of Adsorbent:

Paper is actually a network of cellulose fibres held together by hydrogen bonds (Fig.1). Cellulose is a polymer and there is variation in the nature and number of cellulose units depending upon the type of cellulose present in the paper (Fig.2). SEM images of different papers showed the fibrous nature of celluloses [5, 6].

Table- 1: Chemical composition of news paper pulp

	Cellulose	Hemi cellulose	Lignin	minerals
Newspaper	32%	21%	26%	21%

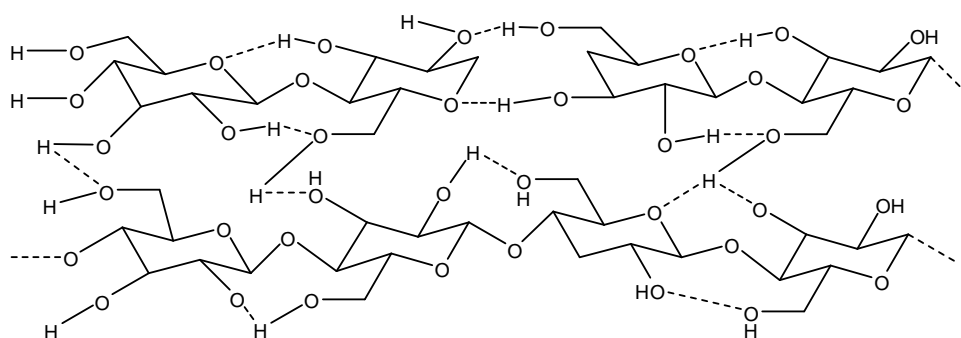


Fig.1: Network of cellulose fibres held together by hydrogen bonds

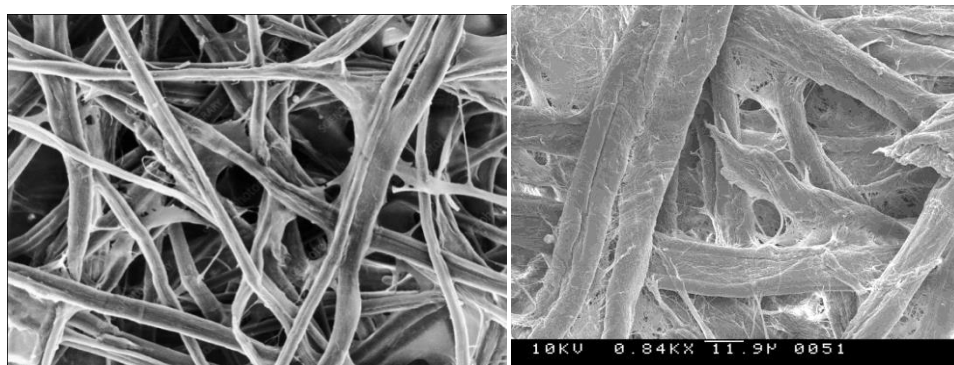


Fig.2: SEM images of different papers showing the fibrous nature of cellulose

Cellulose and other carbon-containing compounds present in paper pulp act as an excellent adsorbent to remove pollutants from aqueous solution. Considering all of these things is rational and can be used as an adsorbent to remove NO_2 .

Experimental procedure:

Newspapers, waste material are collected from the scrap stalls and were used as adsorbent. These newspapers are washed with millipore water to remove grease other dust particles and

were made into the paste using mortar and pestle. The paste was dried for 15 days until they become crisp. Then this dried paste was grinded to powder by mortar and pestle and they were packed in air tight container for further use.

An aqueous solution of 100ml of NO_2 of various concentrations (ranging $26\mu\text{g}/\text{m}^3$ – $260\mu\text{g}/\text{m}^3$) is taken in 100ml Stoppard bottle and 1gram of adsorbent is added to the solution. Batch adsorption experiments are carried out at room temperatures. Optimum contact time is maintained. The UV spectrophotometer is used to determine the initial and final concentrations. NEDA reagent is used as absorbing media for analysis of NO_x . The experiments on the adsorption of aqueous NO_2 solution were studied in terms of contact time, concentration, rate and temperature [10, 11].

Communication time result

Adsorption tests are performed at room temperature ($+ 25^\circ\text{C}$). The contact time effect was studied for the initial concentration of $104\mu\text{g} / \text{m}^3$ of NaNO_2 solution and the adsorbent size of 1 gram. The final filtering of NO_2 solution was determined at various intervals i.e. 0, 15, 20, 30, 45, 60 minutes [7 - 12].

Effect of concentration

The experiments were performed by varying the concentration of NO_2 between $26\mu\text{g}/\text{m}^3$ – $260\mu\text{g}/\text{m}^3$. 60minutes of contact time was maintained and 1gm of adsorbent was added [13-15].

Effect of dosages

The amount of adsorbent dosage will affect the adsorption capacity. Taking that factor into consideration, in the present study different amounts of adsorbents ranging from 0.1 gm - 1gm were added [16-19]. The Final concentration of the NaNO_2 solution is determined. The removal capacity will give direction to determine the optimum dosage.

Results and Discussions

Effect of contact time

A fixed concentration $104\mu\text{g}/\text{m}^3$ of aqueous NO_2 solution is made to pass through the catalytic tube at a constant temperature and filled with 1gm of adsorbent with a surface area of 10.99sq.cms . The experiment was performed with different contact time and variation in the concentration was determined by NEDA method. The results were given in Fig.3. The volume of NO_2 emissions by paper pulp has been found to increase with increasing contact time. Uptake of NO_2 molecules by paper pulp occurs rapidly at initial stages and gets elapsed till it reaches to equilibrium. This behaviour can be attributed to the diminishing active sites of paper pulp. The optimal contact time is 45 min beyond when the removal efficiency is unsatisfactory [18, 19]. Fig.5 shows a smooth curve with no kinks showing the monolayer coating of the adsorbent.

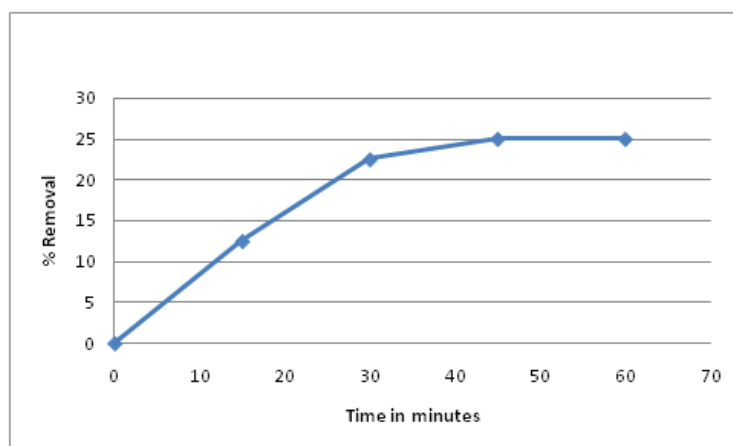


Fig.3: Variation of Contact Time between Paper Pulp and Aq. Solution of NO₂

Effect of NO₂ concentration

The experiments were performed by varying concentration of NaNO₂ solution at room temperature with 1gm of adsorbent. The results were given in Fig.4. Adsorption of NO₂ by paper pulp decreases with the increase in concentration which is observed in the Fig.4. At lower concentration the percentage removal of NO₂ is very high because the ratio of initial concentration of NO₂ to the available surface area is low and so the percentage removal is high [21-23]. At higher concentrations the availability of active sites is less so adsorption becomes less.

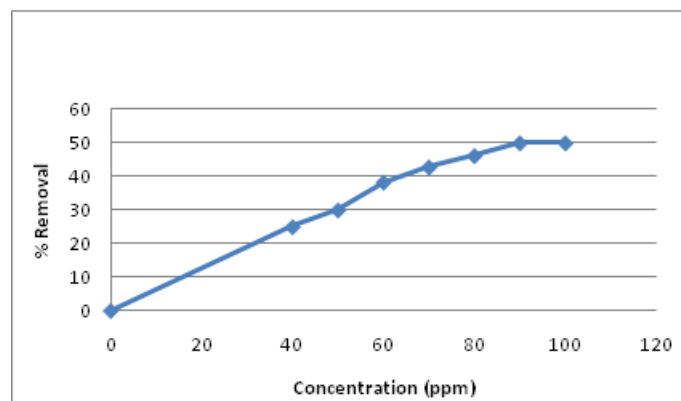


Fig.4: Effect of initial concentration of NO₂ on Adsorption

Effect of adsorbent dosage

NaNO₂ aqueous solution is passed through the various amounts of adsorbent in order to infer the dosage. A fixed concentration (40ppm) of NaNO₂ and contact time (45 min) was maintained at room temperature. The results were given in the Fig.5. From the figure it reflects that NO₂ removal increases with the increasing the amount of paper pulp due to the availability of more active sites on surface area. Beyond 6gms removal efficiency is not striking. Very slight change or no removal of NO₂ beyond an optimal dose may be attributed to the achievement of equilibrium between paper pulp and NO₂ at the operating conditions [22, 23].

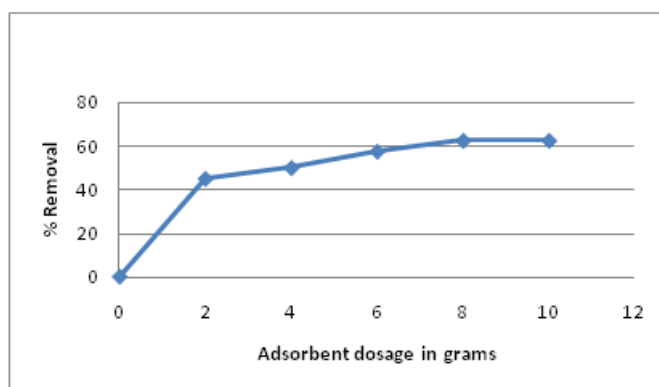


Fig.5: Variation of paper pulp dosages

FTIR Analysis

FTIR spectral studies were carried out, Figure-6, 7 represents before and after adsorption of NO_2 on the paper pulp. The results reveals that the prominent peaks at 2 regions after adsorption are the characteristics of $-\text{OH}$ groups and $\text{C}=\text{O}$ groups which proves that NO_2 molecules are completely adsorbed. The FTIR spectra of NaNO_2 showed strong peaks at 1250 cm^{-1} and at 2150 cm^{-1} and medium peaks at 1400 cm^{-1} and 1325 cm^{-1} . which represents Nitrate and Sodium groups Fig.6. These peaks were not observed after adsorption (Fig:7).

The mechanism of Adsorption is suggested as adsorption of NO_2 by paper pulp may be due to ion-dipole interactions chemical and structure of surface of paper pulp. Lignin, Cellulose, hemicelluloses functional groups are binding to those materials

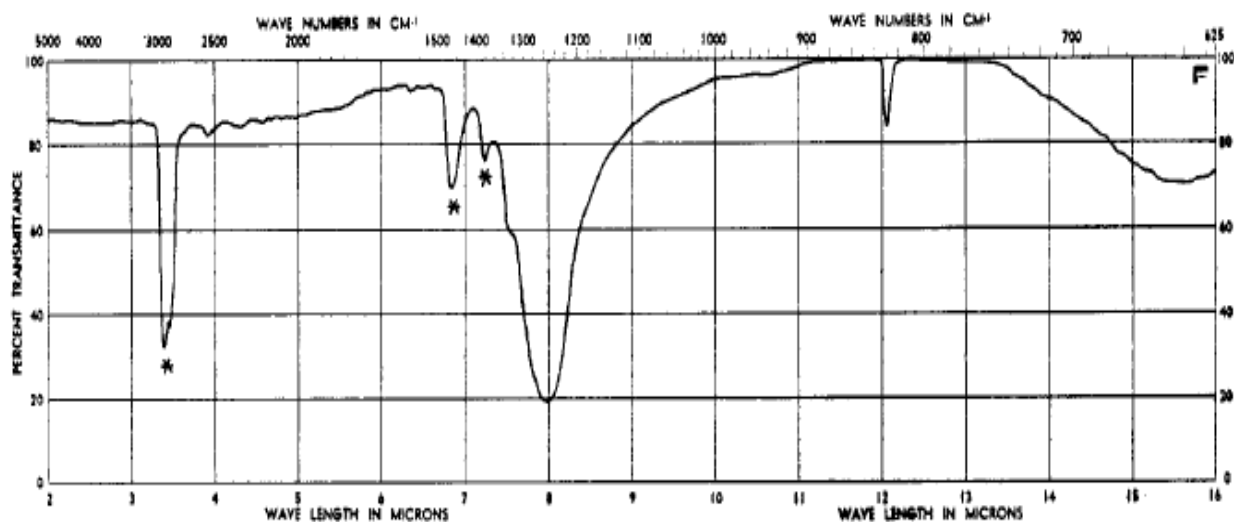


Fig.6: FTIR spectra of NaNO_2 before Adsorption

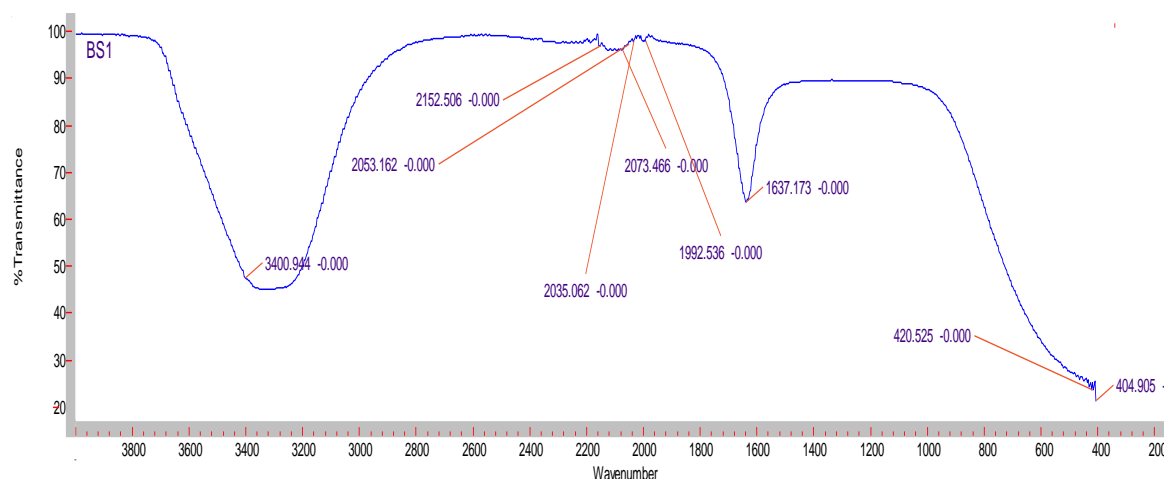


Fig.7: FTIR spectra of NaNO₂ after Adsorption treatment

Adsorption Isotherm studies

Studies of equilibrium on adsorption reveal the information about the capacity of the adsorbent. An adsorption isotherm is characterized by certain constant values which express the surface properties and affinity of the adsorbent and also be used to compare the adsorptive capacities of the adsorbent for different adsorbate equilibrium data can be analyzed using commonly known adsorption systems. Several mathematical models can be used to describe experimental data of adsorption isotherm. The Freundlich and Langmuir models were employed for the analysis of adsorption took place in the adsorption [19-23].

Freundlich Adsorption isotherm

In the Freundlich adsorption isotherm intercept (the reciprocal of slope (n)) represents the adsorption intensity and capacity the results were shown in the figure 8 The intercepts are decreasing with increase in temperature, adsorption capacity is decreasing up to certain temperature indicating lower temperature is favourable for adsorption and is becoming constant from 40-60°C indicating further increase in temperature will not influence adsorption capacity. It is slightly enhanced at higher temperature which will not influence adsorption capacity.

The adsorption intensity is also decreasing with increasing in temperature indicating lower temperature is favourable for adsorption process.

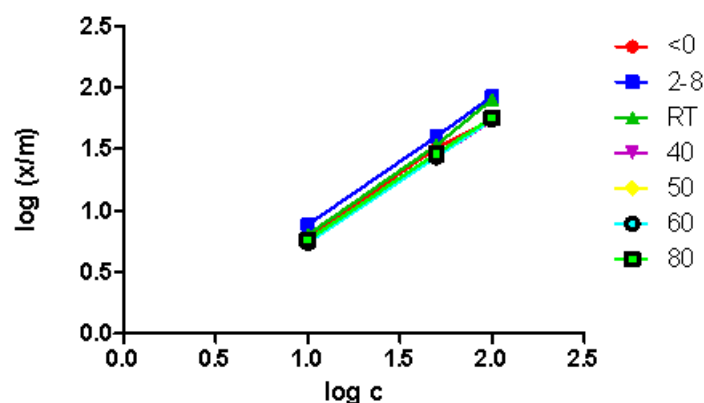


Fig.8: Freundlich Adsorption isotherm

Langmuir adsorption Isotherm: According to Langmuir adsorption isotherm the Q_e/C_e versus Q_e should give a linear relation (20-25). The present experiment was plotted with the same graph and shown in the fig.9. From the figure it was observed that the adsorption capacity is increasing with temperature initially as the temperature in increasing there is a deviation in the linear relation. From the figure it is concluded that adsorptive removal of NaNO_2 by paper pulp at 0°C , $2-8^\circ\text{C}$ temp can also described by Langmuir adsorption Isotherm.

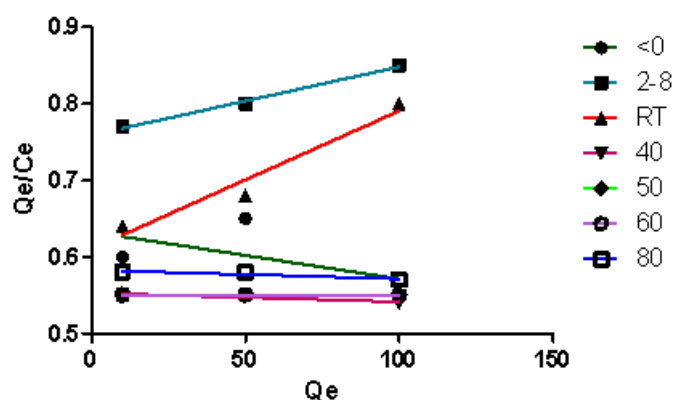


Fig.9: Langmuir adsorption Isotherm

Conclusions:

- The extent of NO_2 removal by the paper pulp was found to increase with the increase in contact time. The optimal contact time is 45 min beyond which the removal efficiency is not appreciable.
- The percentage removal of aqueous solution of NO_2 increased with increase in concentrations, the maximum percentage removal of NO_2 is observed at the higher concentrations compared to lower concentrations.
- The transport of NO_2 increases with increase in paper pulp dosage (2, 4, 6, 8 and 10 grams). Percentage removal was found to increase rapidly at the beginning which declined as paper pulp dosage is increased. Optimum dosage was 25 minutes and 6gms respectively.
- The usage of waste plant materials is a vital development in sustainable environment technology. The quality of air /water can be improvised for underdeveloped countries as plant material is widely accepted and is more receptive by the environmentalists as it solves the problem of solid waste.
- Our research promotes low cost-ecofriendly technology and further work can be extended to column studies and controlling air pollution caused by NO_x .

References

- [1] E. Fattore, V. Paiano, A. Borgini, A. Tittarelli, M. Bertoldi, P. Crosignani, R. Fanelli, *Environ Res.* 111:1321-1327 (2011). <https://doi.org/10.1016/j.envres.2011.06.012>
- [2] S. Kent Hoekman, Curtis Robbins, *Fuel process technol.* 96: 237-149 (2012). <https://doi.org/10.1016/j.fuproc.2011.12.036>
- [3] Hartmut Spliethoff, Ulrich Greul, Helmut Rüdiger, Klaus R.G. Hein, *Fuel.* 75:560-564 (1996). [https://doi.org/10.1016/0016-2361\(95\)00281-2](https://doi.org/10.1016/0016-2361(95)00281-2)
- [4] G. J. Frost, S. A. McKeen, M. Trainer, T. B. Ryerson, J. A. Neuman, J. M. Roberts, A. Swanson, J. S. Holloway, D. T. Sueper, T. Fortin, D. D. Parrish, F. C. Fehsenfeld, F. Flocke, S. E. Peckham, G. A. Grell, D. Kowal, J. Cartwright, N. Auerbach, T. Habermann, *Journal Of Geophysical Research*, 111:1-19 (2006). <https://doi.org/10.1029/2005JD006354>
- [5] H. Takeuchi, M. Ando, N. Kizawa, *Industrial and Engineering Chemistry Research.* 16: 303–308 (1977). <https://doi.org/10.1021/i260063a010>.
- [6] <https://www.sciencephoto.com/media/874491/view>.
- [7] https://upload.wikimedia.org/wikipedia/commons/d/df/Filter_paper_840_3x3_copy.jpg.
- [8] Asheesh Kumar Yadav, et al., *Journal of Hazardous materials.* 128: 289-293 (2006). <https://doi.org/10.1016/j.jhazmat.2005.08.006>
- [9] Krishna G Bhattacharyya, Arunima Sharma, *Journal of Hazardous Materials* 113: 97–109 (2004). <https://doi.org/10.1016/j.jhazmat.2004.05.034>
- [10] A.K. Bhattacharya, T.K. Naiya, S.N. Mandal, and S.K. Das, *Chem.Eng.J.*, 137: 529 – 554 (2008). <https://doi.org/10.1016/j.cej.2007.05.021>
- [11] M.Srimurali, A. Pragathi, J. Karthikeyan, *Environmental pollution* 99:285-289 (1998). [https://doi.org/10.1016/S0269-7491\(97\)00129-2](https://doi.org/10.1016/S0269-7491(97)00129-2)
- [12] P. Brown, I. Jetcoat, D. Parrisha, S. Gilla, E. Grahams, *Adv. Environ. Res.*, 4 (i): 19-29 (2000).
- [13] M. Dakiky, M. Khamis, A. Manassra, M. Mereb, *Adv. Environ. Res.* 6: 533-540 (2002). DOI: 10.4197/ Sci. 25-1.2
- [14] H. Demiral, I. Demiral, F. Tumsek, and B. Karabacakoglu, *Chem.Eng.J.*, 144: 188 –196 (2008). <https://doi.org/10.1016/j.cej.2008.01.020>
- [15] Shashi Prabha Dubey, Krishna Gopal, J.L. Bersillon, *Journal of Environmental Biology*, 30: 327-332 (2009).
- [16] A. H. Mahvi, R. Nabizadeh, F. Gholami, A. Khairi, *Iran. J. Environ. Health. Sci. Eng.*, 4 : 191-196 (2007).
- [17] KN. Tarun, C. Pankaj, KB. Ashim, KD. Sudip, *Chem. Eng. J.*, 148: 68-79 (2008). <https://doi.org/10.1016/j.cej.2008.08.002>
- [18] NingchuanFeng, XueyiGuo, ShaLiang, YanshuZhu, JianpingLiu, *Journal of Hazardous Materials*, 15: 49-54 (2011). <https://doi.org/10.1016/j.jhazmat.2010.08.114>
- [19] G. Annadurai, R.S. Juang, D.J. Lee, *Sci Technol* 47 : 185-190 (2003). <https://doi.org/10.2166/wst.2003.0049>
- [20] G. da C.Cunha, L.P.C.Romao, M.C.Santos, B.R.Araújo, S.Navickiene, V.L.de Pádua, *Bioresource Technology*, 101: 3345-3354 (2010). <https://doi.org/10.1016/j.biortech.2009.11.096>

- [21] J. Monika, V. Garg, Kardirveluk, *J. Hazardous materials*; 162: 365 – 372 (2009).
<https://doi.org/10.1016/j.jhazmat.2008.05.048>
- [22] G. da C.Cunha, L.P.C.Romão, M.C.Santos, B.R.Araújo, S.Navickiene, V.L.de Pádua, *Bioresource Technology*, 101: 3345-3354 (2010).
<https://doi.org/10.1016/j.biortech.2009.11.096>
- [23] N . Gandhi, D. Sirisha, K B. Chandra Sekhar, *Journal of Chemical and Pharmaceutical Research* 8: 127 – 141 (2016).
- [24] D Park, S.R. Lim, Y.S. Yun, J.M. Park, *Chemosphere*, 70: 298 – 305 (2007).
DOI:10.1016/j.chemosphere.2007.06.007.
- [25] S. Liang, X Y. Guo, N C. Feng, Q H Tian, *Application of orange peel xanthate for the adsorption of Pb²⁺ from aqueous solutions. Journal of Hazardous Materials*, 170: 425–429 (2009). DOI:10.1016/j.jhazmat.2009.04.078.