## Simultaneous Removal of Lead and Nickel Ions from Aqueous Synthetic Solutions by Chitosan Coated Cobalt Ferrite

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This research study deals with lead and nickel simultaneous removal from aqueous solutions by the use of chitosan coated cobalt ferrite as adsorbent. Batch removal tests were performed in order to establish the main parameters that influence the sorption capacity, removal efficiency and the selectivity of this adsorbent. The values of sorption capacity for lead and nickel experimentally determined are: 56.23 mg/g and respectively 45.11 mg/g. Langmuir and Freundlich adsorption isotherms were used to interpret the sorption experimental data. The kinetic data were explored by pseudo-first order, pseudo-second order and intraparticle diffusion kinetic models. The experimental data were well fitted with the pseudo-second order model for both heavy metals. The main conclusion that can be drawn from this research is that this material can be successfully used for the removal of lead and nickel from binary aqueous solutions and wastewater.

Keywords: lead and nickel removal, chitosan coated cobalt ferrite, lead and nickel sorption

In recent years, a great importance was granted to the removal of heavy metals from wastewater and synthetic solutions using sorption process. This is due to the fact that the sorption process has numerous advantages such as: the possibility of using a large number of natural materials (low-cost) as sorbents [1-5], it can be performed without complex equipments, the process can be simple with a fast retaining rate, it can be used for removal and recovery of a large number of heavy metals, it does not require adding additional substances like precipitation, it does not involve high operation costs, the removal efficiency can reach values of 100%. Another main advantage is that it can be used with high efficiency for removal of metal ions in dissolved state even from dilute solutions [6, 7].

Consequently, many researches were conducted to develop new adsorbents with high surface area, high sorption capacity and sometimes with magnetic properties [8]. Furthermore, research interests are now focused on the development of the novel nano-metal oxides, nanomixed oxides such as ferrite simple or doped with different materials as efficient adsorbents for water remediation [9]. It is well known that some of the magnetic materials have as a main disadvantage the fact that they have low pollutant removal potential [10]. This is the reason for their surface modification by the use of some surfactants or/ and by the use of their coating. In this sense, cobalt ferrite  $(CoFe_{0}O_{1})$  simple or coated with different materials has interesting properties that recommend it for using as adsorbents of heavy metals from solutions and wastewater [11].

Among the heavy metals, lead (II) and nickel (II) ions were selected for this study due to the fact that both of them are considered to be toxic for human beings [12]. Environmental lead pollution is mainly due to industrial units, leaded petrol, smoke emissions from coal and gas power stations and use of paints [12]. Lead has also important negative effects to the ecosystems. Nickel is used for the production of stainless steel, non-ferrous alloys, Ni-based superalloys, industrial machineries, precision electronics and catalysts [13]. Once in the human body, nickel has pathological effects varying from contact dermatitis to lung fibrosis, cardiovascular and kidney diseases, and even cancer [13].

Therefore, the present work investigates the application of chitosan coated cobalt ferrite as a metal sorbent for the remediation of aqueous binary solutions containing different concentrations of lead (II) and nickel (II) ions. The effects of experimental controlling parameters such as *p*H, contact time, and initial metal ion concentration were also studied and optimized. Langmuir and Freundlich adsorption isotherms were applied to fit the experimental data. The kinetics data were examined by the pseudo-first order, pseudo-second order and intraparticle diffusion kinetic models to establish the best correlation coefficient.

#### **Experimental part**

#### Materials and methods

Chitosan coated cobalt ferrite nanoparticles were obtained from the Coordination Chemistry Laboratory from Ilie Murgulescu Institute of Physical Chemistry, Romanian Academy. It was previously characterized. By diluting stock solutions of 1000 mg/L Pb(NO<sub>3</sub>) (Merck), the solution of lead with concentration that varied between 200 mg/L to 10 mg/L were obtained. The same procedure was performed to obtained diluted solutions of nickel with the same concentrations from 1000 mg/L Ni(NO<sub>3</sub>)<sub>2</sub> (Merck). Equal volumes of Pb(II) and Ni(II) with the same concentrations needed to perform sorption experiments. HNO<sub>3</sub> 1N and NaOH 0.1N analytical grade were used to establish the effect of the *p*H on sorption capacity of the adsorbent used. An Agilent 3200P pH Meter was used to determine the values of the solution's *p*H.

Batch systems tests were performed on an orbital shaker type GFL 3015 by contacting synthetic binary aqueous solutions of Pb(II) and Ni(II) with adsorbent at room temperature and 150 rpm. Lead and nickel concentrations in initial and in solution after sorption were determined by the use of contrAA<sup>®</sup> 300 Analytik Jena Atomic Absorption Spectrometer. The influence of *p*H to Pb(II) and Ni(II)

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sorption was conducted by contacting 0.025 g of sorbent with 25 mL of Pb(II) and Ni(II) solution at different concentrations for 24 h to reach the equilibrium. *p*H value varied in the range of 2 to 6.5 at  $21\pm2^{\circ}$ C. Experiments were performed in the contact time ranged between 5 to 600 minutes to establish the effect of contact time.

The metal uptake by chitosan coated ferrite (Q), and the removal efficiency ( $\eta$ ) were determined by the use of the eqs. (1) and (2), respectively:

$$Q = \frac{(C_i - C_f) \cdot V}{m}$$
 (1)

where:

Q – lead/nickel uptake (mg/g);

 $C_i$  - the concentration of lead/nickel ions in the initial solution (mg/L);

C<sub>f</sub> - the concentration of lead/nickel ions remaining in solution at different times (mg/L);

V - the solution volume (L);

m – starting sorbent weight (g).

$$\eta = \frac{(C_i - C_f)}{C_i} \cdot 100 \tag{2}$$

where: η is removal efficiency (%);

 $C_i$  - the concentration of lead/nickel ions in the initial solution (mg/L);

C<sub>f</sub> - the concentration of lead/nickel ions remaining in solution at various times (mg/L).

#### **Results and discussions**

*pH value influence on sorption capacity of chitosan coated cobalt ferrite nanoparticles* 

The first parameter that was studied is *p*H. This is due to the fact that it is well known that *p*H has an important effect to the ionic state of functional groups from the surface of adsorbent, and also to the ionic state of the heavy metals in the solution [14].

Consequently, we had chosen to perform the experiments in the *p*H range 2-6.5 at  $21\pm2^{\circ}$ C. The *p*H above 6.5 were not evaluated because, at this *p*H values, lead will precipitate as Pb(OH)<sub>2</sub> or Pb(OH)<sub>3</sub> and nickel as Ni(OH)<sub>2</sub> [14]. Consequently, at higher *p*H value precipitation and adsorption will compete in the process of the heavy metals removal.

Figure 1 demonstrates the sorption of lead and nickel from aqueous binary solutions is strongly dependent on the solution's *p*H value.

It is obvious that the amount of heavy metals retained by chitosan coated cobalt ferrite nanoparticles increases with the increase of *p*H value. It was also observed that the quantity of lead retained by chitosan coated cobalt



Fig.1. Effect of *p*H on retaining capacity of chitosan coated ferrite at initial lead concentration 99.3 mg/L and nickel concentration 101.75 mg/L

ferrite nanoparticles is higher than the quantity of nickel. Thus, chitosan coated cobalt ferrite has a higher selectivity for lead.

Another conclusion that can be drawn is the fact that the optimum pH value for both metal ions removal by chitosan coated cobalt ferrite nanoparticles is equal with 5. This value of pH corresponds to the initial solution. Consequently, the next tests will performed at solution's pH without change of its.

The observation that the retaining capacity of chitosan coated cobalt ferrite increases with the increase of the pH value is attributed to the fact that chitosan has various functional groups such as  $-NH_2$ , -OH that can be protonated and presented in positively charged form at low *p*H value [15]. At low *p*H value, the highest concentration of H<sup>+</sup> and H<sub>3</sub>O<sup>+</sup> in the aqueous solution will determine a competition between these cations and heavy metals ions for free sorption sites on the chitosan coated cobalt ferrite's surface. Furthermore, at lower *p*H value an electrostatic repulsion can occur between heavy metals ions and positively charged sorbent surface.

A decrease of retaining capacity of chitosan coated cobalt ferrite was observed at higher *p*H value than 5, which can be attributed to the fact that all the available sorption sites of the adsorbent are bonded by the heavy metals. A further heavy metals sorption might not occur.

The results obtained are in agreement with other research studies related to lead and nickel removal by sorption [11-16].

# The contact time influence on sorption capacity of chitosan coated cobalt ferrite

It is well known that adsorption is a time-dependent process. Consequently, this research step has as main objective to analyze the effect of contact time on adsorption efficiency and removal efficiency. This will help us to better comprehend the adsorption kinetics of lead and nickel from binary solutions.

The adsorption kinetic curves of Pb(II) and Ni(II) onto chitosan coated cobalt ferrite and the variation of removal percentage versus contact time are shown in figures 2 and 3.

As shown in figure 2, heavy metals sorption onto chitosan coated cobalt ferrite can be divided in two stages: an adsorption with higher rate at the beginning (the sorption increases rapidly during the first 60 min) and an adsorption with a much lower rate (the sorption is nearly constant for both heavy metals after 360 min). The amount of sorbed metals ions did not significantly change with time after the 360 min considered equilibrium time. The initial stage



Fig. 2. The effect of the contact time on retaining capacity of chitosan coated cobalt ferrite



**Time (minute)** Fig. 3. The effect of the contact time on removal efficiency of chitosan coated cobalt ferrite

with high rate is attributed to the large number of available binding sites, resulting in a concentration gradient [17].

The available active sites will be gradually occupied by heavy metals ions with the increase of contact time, and accordingly the sorption rate will decrease. The same trend of variation of removal efficiency in function of the contact time was noticed (fig. 3).

The tests were performed also with the single heavy metal ion solutions to determine the influence of the competitor ions on sorption capacity. In case of solution loaded with Pb(II) ions, the sorption capacity of chitosan coated cobalt ferrite has a value equal to 58.13 mg/g. For the solution loaded only with Ni(II) sorption capacity experimental determined 57.04 mg/g.

The following conclusions have been distinguished from this part of the research study:

-the sorption capacity has value equal to 56.23 mg/g for lead and 45.11 mg/g for nickel for sorption from binary solutions;

-the maximum removal efficiency is 56.63 % for lead and 44.33% for nickel for removal from binary solutions;

-the presence of other heavy metal ions (competitors) determines the decrease of the sorption capacity, and removal efficiency for both heavy metals tested;

-the shaking time 6 h is enough to reach the equilibrium.

#### Adsorption isotherms

Adsorption isotherms are generally used to describe the relationship between the amount of metal adsorbed and the metal ion concentration remaining in solution [18]. Many equations have been used to analyze the experimental adsorption equilibrium data. The adsorption mechanism, the surface properties and affinity of the sorbent can be determined based on the equation parameters and the underlying thermodynamic assumptions [18]. Various isotherm equations like Freundlich, Langmuir, Temkin and Dubinin–Radushkevich isotherm have been involved in describing the equilibrium characteristics of adsorption [18]. Among these the Freundlich and Langmuir equilibrium isotherm are the most widely used.

In this stage of the study, the equilibrium data obtained for Pb(II) and Ni(II) removal by chitosan coated cobalt ferrite were examined to establish the equilibrium model that characterize better sorption process. For this purpose, binary solutions with lead concentration varied between 99.3 to 4.87 and nickel concentration in the range 101.75 to 4.95 were involved. We fitted experimental data with Langmuir and Freundlich isotherms. According to Langmuir model, metal ions are chemically adsorbed at a fixed number of sorption sites, each site is occupied with only one ion and all sorption sites are energetically equivalent [18].

The equation (3) formulates the linear form of the Langmuir equation [18]:

$$\frac{C_{e}}{Q_{e}} = \frac{1}{Q_{max}} \cdot C_{e} + \frac{1}{Q_{max}K_{L}}$$
(3)

where :

 $Q_{\rm c}$  is the amount of heavy metal ions retained at equilibrium (mg/g); C\_ is the equilibrium concentration of heavy metals (mg/L);  $K_{\rm r}$  is the Langmuir model parameter related to energy of adsorption (L/mg) and  $Q_{\rm max}$  defines the maximum sorption capacity (mg/g).

Freundlich model assumes that the concentration of adsorbate on the adsorbent surface increases with the adsorbate concentration and an infinite amount of adsorption can take place [18]. This model is widely used to describe a sorption process on heterogeneous surfaces.

The equation (4) describes Freundlich model [18]:

$$Q = K_F \cdot C_e^{1/n}$$
(4)

where :

 $K_{\rm F}$  is Freundlich constant which represents sorption capacity and n is Freundlich constant that show sorption intensity.

The linear form of Freundlich equation is as follows:

 $\ln Q = \ln K_F + \frac{1}{n} \ln C_e$ 

Figure 4 has been used to appreciate the values of Langmuir parameters ( $K_L$  and a) for lead and nickel sorption by chitosan coated cobalt ferrite.

Figure 5 depicts the Freundlich linearized isotherm for lead and nickel sorption on chitosan coated cobalt ferrite.

The data presented in figures 4 and 5 have been used to calculate the Langmir and Freundlich parameters were







Fig. 5. Freundlich linearized isotherm for lead and nickel sorption on chitosan coated cobalt ferrite

LANGMUIR AND FREUNDLICH SORPTION PARAMETERS									
Heavy metal ions	Langmuir parameters			Freundlich parameters					
	Q <sub>max</sub> (mg/g)	K <sub>L</sub> (L/mg)	R <sup>2</sup>	K <sub>F</sub> (mg/g)	n	$\mathbb{R}^2$			
Pb(II)	66.67	0.1630	0.987	10.2	2.0576	0.976			
Ni(II)	50	0.1639	0.982	7.05	1.9920	0.955			

Table 1

calculated by the use of data presented in figures 4 and 5. The values obtained are presented in table 1.

Table 1 and figures 4 and 5 revealed that the Langmuir equation (correlation coefficient  $R^2 = 0.987$  and  $R^2 = 0.982$ , respectively, supporting information table 1), better describes the sorption isothermal behaviors for both heavy metals sorbed.

#### Adsorption kinetic

Kinetic models, such as the pseudo-first-order kinetic, pseudo-second-order kinetic, and intraparticle, were applied to the experimental data to elucidate the adsorption mechanism. For this purpose, tests were performed by contacting samples containing chitosan coated cobalt ferrite and binary solutions at time from 5 min and 480 min.

The pseudo-first-order model has been proposed for sorption processes that the rate of adsorption on sorbent is proportional to the number of active sites available onto adsorbent.

The pseudo-first-order model [19] is expressed by Lagergren equation:

$$\frac{dQ_t}{dt} = k_1 (Q_e - Q_t)$$
(6)

where:  $Q_e$ ,  $Q_1$  are the sorption capacities at equilibrium and at time t (mg/g), and  $k_1$  is the rate constant of pseudofirst order sorption (min<sup>-1</sup>).

The linear form of the Lagergren model (eq. 7) has been used to predict the applicability of the pseudo-first model to heavy metals sorption process onto chitosan coated cobalt ferrite:

$$\log(Q_{e} - Q_{t}) = \log Q_{e} - \frac{k_{1}}{2.303} \cdot t$$
 (7)

where  $Q_e$  and  $Q_t$  represent the amount of heavy metals sorbed on sorbent (mg/g) at equilibrium and at time t, respectively and  $k_1$  is the rate constant of first-order sorption (min<sup>-1</sup>). The rate constant ( $k_1$ ) and correlation coefficient  $R^2$  have been determined from the plot of log( $Q_e$ - $Q_t$ ) versus f(t) (fig. 6).





The second-order kinetic model is expressed by the equation [19]:

$$\frac{\mathbf{t}}{\mathbf{Q}_{\mathsf{t}}} = \frac{1}{\mathbf{k}_2 \mathbf{Q}_{\mathsf{e}}^2} + \frac{\mathbf{t}}{\mathbf{Q}_{\mathsf{e}}} \tag{8}$$

where:  $k_2$  is the rate constant of second-order adsorption (g/mg·min).

Linear plots  $t/Q_1$  against t (fig. 7) have been utilized to establish the kinetic parameters. The  $k_2$  and  $Q_2$  values were calculated from the slope and intercept of the plot (fig. 7).



Fig. 7. Pseudo-second order sorption kinetics of Pb(II) and Ni(II) onto chitosan coated cobalt ferrite

There are numerous studies suggesting that at initial stages of adsorption diffusion happens by external mass transfer which is then followed by intraparticle diffusion [10, 20-23]. This can be due to the fact that, at the beginning of the sorption process, a large concentration gradient is between the aqueous phase and the adsorbent surface that will lead to a quicker movement of the solute onto the sorbent surface [10]. Even though the largely intraparticle diffusion takes place, this is a slow process [22]. The model that explains the sorption process as an intraparticle diffusion process was introduced by Weber – Morris being namely intraparticle diffusion kinetic model. Weber-Morris stated that in some sorption processes the solute uptake is proportional to  $t^{0.5}$  instead of the contact time [23].

Intraparticle diffusion rate [21] is mathematical expressed by the following equation:

$$\mathbf{Q}_{t} = \mathbf{k}_{i} \mathbf{t}^{0.5} \tag{9}$$

where  $k_i$  is intraparticle diffusion rate (mg/mg·min<sup>0.5</sup>).

Plots of  $Q_t$  versus  $t^{0.5}$  have been involved in determination of the value of intraparticle diffusion rate constant  $k_i$  (mg/ mg·min<sup>0.5</sup>). If the intraparticle diffusion is the rate-limiting step, the plot of  $Q_t$  versus  $t^{0.5}$  is a straight line with the slope  $k_i$  [10].

<sup>b</sup> By examining the data presented in figure 8 it can be concluded that the plot is not linear in shape and trend line does not pass through the origin and consequently the intraparticle diffusion is not the rate-limiting step in case of lead and nickel sorption onto chitosan coated cobalt ferrite. Regarding the plots from the figure 8, they are multi linear with three distinct regions. Thus, three different kinetic mechanisms can be involved. According to other research studies, it can be assumed that the initial curved region

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Fig. 8. Intraparticle diffusion sorption kinetics of Pb(II) and Ni(II) onto chitosan coated cobalt ferrite

will correspond to the external surface uptake, the second stage represents a gradual uptake reflecting intraparticle diffusion as the rate limiting step and the final plateau region show equilibrium uptake [24].

Thus, we calculated values of kinetic parameters and correlation coefficient ( $\mathbb{R}^2$ ) for the pseudo-first order and pseudo-second order kinetic models that are shown in table 2.

According to the data presented in table 2, the pseudosecond-order model is in better agreement with experimental data regarding the kinetics of Pb(II) and Ni(II) ions sorption by chitosan coated cobalt ferrite (correlation coefficient  $R^2 = 0.999$  and 0.998) compared to pseudo-first-order and intraparticle diffusion. In this case, the rate determining step is the chemical adsorption and not the physical sorption and the mass transfer.

#### Conclusions

Thus, by this work, batch experiments were performed in order to test sorption capacity of chitosan coated cobalt ferrite. Binary metal solutions (lead and nickel loaded) were used for this purpose. Comparative tests were accomplished to determine the influence of the competitive metal ions in the solution. From these tests, it can be concluded that for both heavy metals, sorption capacity of chitosan coated cobalt ferrite decreases slowly when they were in a binary solution comparatively with the single solution. The optimum conditions for sorption process are pH equal with 5 and a contact time of 6 h. The maximum adsorption capacities of chitosan coated cobalt ferrite used in this study are 56.23 mg/g for lead and 45.11 mg/g for nickel. Langmuir adsorption model is in better agreement with the experimental data. The sorption process showed a pseudo-second-order kinetics, which suggests chemical reactions between heavy metal ions and functional groups on the adsorbent surface.

Findings from this research study indicated that chitosan coated cobalt ferrite can be successfully applied to remove lead and nickel for aqueous solutions and wastewater.

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### Table 2 THE RATE CONSTANT K, AND R<sup>2</sup> CORRELATION COEFFICIENT VALUES FOR THE PSEUDO-FIRST ORDER AND PSEUDO-SECOND ORDER KINETIC

Heavy	Pseudo-first order		Pseudo-second order sorption			
metals	sorption kinetics		kinetics			
	k1 (min-1)	$\mathbb{R}^2$	k₂ (g/mg∙min)	$\mathbb{R}^2$		
Pb(II)	9.212.10-3	0.929	2.7524.10-3	0.999		
Ni(II)	9.212.10-3	0.897	1.5921-10-3	0.998		

#### **References**

1. KAMAR, F.H., NECHIFOR, A.C., NECHIFOR, G., SALLOMI, M.H., D. JASEM, A.D., Rev. Chim. (Bucharest), **67**, no. 1, 2016, p. 1

2. DELEANU, C., SIMONESCU, C.M., CONSTANTINESCU, I., Rev. Chim. (Bucharest), **59**, no. 6, 2008, p. 639

3.KAMAR, F.H., NECHIFOR, A.C., MOHAMMED, A.A., ALBU, P.C., CRACIUN, M.E., Rev. Chim. (Bucharest), **66**, no. 5, 2015, p. 615

4. STEFAN, D.S., BELCU, M., STEFAN, M., MARINESCU, I.A., Rev. Chim. (Bucharest), **61**, no. 1, 2010, p. 31

5. SIMONESCU, C.M., MARIN, I., TARDEI, C., DRAGNE, M., CAPATINA, C., Rev. Chim. (Bucharest), **65**, no. 7, 2014, p. 750

6.SIMONESCU, C.M., FERDES, M., Polish Journal of Environmental Studies, **21**(6), 2012, p 1831

7. TOADER, G., STANESCU, P.O., ZECHERU, T., ROTARIU, T., EL-GHAYOURY, A., TEODORESCU, M., Arabian Journal of Chemistry, DOI: 10.1016 / J. Arabjc. 2016, 03.009

8. LIU, J.-F., ZHAO, Z.-S., JIANG, G.-B., Environ. Sci. Technol. **42**(18), 2008, p. 6949, doi:http://dx.doi.org/10.1021/es800924c.18853814.

9. MAHMOUD, M.E., ABDELWAHAB, M.S., FATHALLAH, E.M., Chem. Eng. J. 223, 2013, p. 318, doi:http://dx.doi.org/10.1016/j.cej.2013.02.097. 10. SINGH, M., DOSANJH, H.S., SINGH, H., Journal of Water Process Engineering **11**, 2016, p. 152

11. CULITA, D.C., SIMONESCU, C.M., DRAGNE, M., STANICA, N., MUNTEANU, C., PREDA, S., OPREA, O., Ceramics International **41**, 2015, p. 13553, http://dx.doi.org/10.1016/j. ceramint.2015.07.150

12. MASOUMI, F., KHADIVINIA, E., ALIDOUST, L., MANSOURINEJAD, Z., SHAHRYARI, S., SAFAEI, M., MOUSAVI, A., SALMANIAN, A.-H., ZAHIRI, H.S., VALI, H., NOGHABI, K.A., Journal of Environmental Chemical Engineering **4**, 2016, p. 950

13. COMAN, V., ROBOTIN, B., ILEA, P., Resources, Conservation and Recycling **73**, 2013, p. 229

14. TAN, Y., CHEN, M., HAO, Y., Chem. Eng. J. 191, 2012, p. 104

15. SIMONESCU, C.M., DELEANU, C., STANCU, M., CAPATINA, C., Journal of Environmental Protection and Ecology 13(2), 2012, p. 462 16.SIMONESCU, C.M., TATARUS, A., TARDEI, C., PATROI, D., DRAGNE, M., CULITA, D.C., PATESCU, R.-E., BUSUIOC, L.T., MELINTE, I., Rev. Chim. (Bucharest), **66**, no. 5, 2015, p. 732

17.AWUAL, MD. R., Chemical Engineering Journal **266**, 2015, p. 368 18.RASHID, M., LUTFULLAH, F. K., Journal of Water Process Engineering **3**, 2014, p. 53

19. NGAH, W.S.W., ENDUD, C.S., MAYANAR, R., React. Funct. Polym. 50, 2002, p. 181

20. ZHANG, X., BAI, R.B., J. Colloid. Interf. Sci., 264, 2003, p. 30

21.AZZAM, A.M., EL-WAKEEL, S.T., MOSTAFA, B.B., EL-SHAHAT, M.F., Journal of Environmental Chemical Engineering **4**, 2016, p. 2196

22.BABU, B.V., GUPTA, S., Adsorption 14, 2008, p. 85

23.QIU, H., LV, L., PAN, B.-C., ZHANG, Q.-J., ZHANG, W.-M., ZHANG, Q.-X., J. Zhejiang Univ. Sci. A 10, 2009, 716

24. .POPURI, R.S., VIJAYA, Y., BODDU, V.M., ABBURI, K., Bioresource Technology **100**, 2009, p.194

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