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Removal of COD, BOD and Color from Municipal Solid Waste Leachate using Silica and Iron nano particles - A Comparative Study

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Abstract

Application of nano particle in the treatment of municipal solid waste leachate is of recent interest. In this paper, the effectiveness of silica nano particles synthesized from blast furnace slag and iron nano particle synthesized from chemicals was studied for the removal of organic pollutants and color. The synthesized nano particles were characterized using SEM, TEM, EDX and FTIR analysis. Batch experiments were conducted to remove the BOD, COD and color from Aged landfill leachate (ALL) and leachate from the composting yard (CYL). Influencing parameters like pH, contact time, nano particle dosage and Hydrogen peroxide concentration were studied. The maximum removal was achieved at the pH of 6 for both the nano particle, contact time 90 minutes for silica nano particle and 120 minutes for iron nano particle, silica nano particle dose as 0.4g/50 ml, iron nano particle dose as 0.3g/50ml and hydrogen peroxide concentration was found to be 3M and 4M for silica and iron nano particles respectively. The removal efficiency in CYL and ALL using silica nano particle was obtained as 87.15%, 72.72%, 83.15% and 82.5%, 62.5%, 77.34% for color, BOD and COD respectively. Similarly for iron nano particle, the removal efficiency was found to be 60.3%, 65%, 67.43% and 57.06%, 57.27%, 67% for the removal of color, BOD and COD in CYL and ALL, respectively.

Keywords: Blast furnace slag, nano particles, batch experiments, leachate treatment, composting yard leachate

1. Introduction

Disposal of solid waste in the landfill is one of the common and economically viable waste management approach implemented across the world. In general, most of the metropolitan cities normally perform the landfilling techniques. Thus 90% of the solid wastes in developing countries like India are disposed in an unempirical approach. The accessibility of land for discarding the generated waste is decreasing day by day (Mor *et al.*, 2006; Siddiqui *et al.*, 2006; Sharholy *et al.*, 2005). Despite cumulative emphasis on unconventional options, solid waste dumping to landfill holds a substantial role in both developed and developing nations. Even though a lot of sophisticated technologies are available, land filling is a very common mode of clearance due to its economic viability. In course of time the solid waste disposed in landfill produces a slimy liquid called "Leachate". The production of leachate and managing the produced leachate is one of the environmental concerns as the produced leachate causes damage to soil and water (both underground and surface water). Unless precautionary procedures are taken, this will lead to a great menace to environment (Baccini *et al.*, 1989; Lopez 2004). Increased concentration of leachate makes the underground water to contaminate very easily (Petruzzelli *et al.*, 2007; Sarvako 2007; Yang 2006). Even though the policy makers have implemented stern rules and regulation, fostering of non-engineered landfills and inefficient collection and treatment system still persist in developing nations (Yemma *et al.*, 1988).

The treatment method chiefly depends on the leachate characteristics. Treatment of leachate is classified into two types namely, physicochemical and biological. In general the biological method is an effective method for the treatment of leachate generated from composting yard due to less non-biodegradable compounds present in it whereas the presence of non-biodegradable compound is high in aged landfill leachate due to which the biological treatment method proves to be ineffective. Thus the physicochemical methods triumph over all this difficulty. For large-scale treatment of wastewater, such as leachate from municipal solid waste landfills, the adsorbents used must be inexpensive and also, should be a waste material obtained from industries or agricultural residue which always be supposed to least processing and it must be plentiful in nature (Esfahani *et al.*, 2012). Quite a good number of reports are presented on the treatment of wastewater and also the pollutant present in the environment using nanotechnology (Sarvako *et al.*, 2008).

Nano particles were preferred for wastewater treatment because of its size, unique network, its structure, higher catalytic ability and also its reactivity (Zang *et al.*, 2007;

Bailey *et al.*, 1999). In addition, the reason behind the use of nano particle is due to the fact that the reduced surface to volume ratio will increase the absorptive capacity of the nano particles. One of the economical and cost effective treatment techniques is the Fenton process in which the hydrogen peroxide along with ferrous sulphate produces strong hydroxyl radicals. In this paper, the hydrogen peroxide is allowed to react along with the nano particles in order to produce strong hydroxyl radicals. The review of literature reveals that the elements like Cu (Dukkanci *et al.*, 2010), Ce (Xu *et al.*, 2012) Bi (Luo *et al.*, 2010), Mo (Tian *et al.*, 2011), Al (Patra *et al.*, 2013) were incorporated into the nano particles to increase the catalytic ability which results in the decomposition of hydrogen peroxide which gives even more powerful hydroxyl ions. However, Silica nano particle has not been studied for its potential in removing color, BOD and COD. Hence, in this study, an attempt has been made to utilize the waste from foundry unit, Blast furnace slag (BFS) for synthesis of Silica nano particle (SNP). The synthesized SNP was analyzed for its ability to remove color, BOD and COD from municipal solid waste leachate. In addition, the iron nano particle(INP) synthesized from chemical processes has also been studied for its potential in removing color, BOD and COD present in the municipal solid waste leachate.

2. Materials and methods

2.1. Study area and leachate sampling

Erode, located in the north western part of Tamil Nadu State in India has the city population of 157101(as per 2011 census). In addition to this the floating population was found to be 20,000. The leachate samples used in this study were collected from a municipal solid waste dumpsite located at Vendipalayam in Erode district (11° 34′N and 77° 72′E) of Tamil Nadu, India. The dumpsite receives approximately 300 metric tons of waste every day, which excludes hospital waste. Presently, no proper treatment facility is available to treat the leachate in this dump yard. Leachate samples were collected using pre-cleaned polythene bottles and stored in the refrigerator at 4 °C for further experimentation purpose by following the standard procedure.

2.2. Synthesis of Silica nano particles

Raw material used for the synthesis of SNP was granulated blast furnace slag, a waste obtained from a foundry unit. The slag was well grained using a domestic mixer and sieved using 150 microns size sieve. The particles passing through the sieve were collected and stored. The slag and nitric acid was taken in a beaker in the ratio of 1:4 and kept for one hour to ensure the completion of reaction. The precipitated silica was washed five times using double distilled water. The same amount of double distilled water as that of nitric acid was added and the mixture was filtered. The extracted silica was oven dried at 200°C for one hour. The oven dried silica was stored in an air tight container for further use (Jae Kim *et al.*, 2010).

2.3. Synthesis of Iron nano particles

150 ml of distilled water was mixed with 4.1703 g of 0.1M $FeSO_4$ and 150 ml of distilled water was mixed with 3.7224 g of 0.05M EDTA. Both the solutions were mixed together. 2.837 g of 0.75M $NaBH_4$ was mixed in 100 ml distilled water and added drop wise into the mixture solution. The appearance of black colored particles indicated the formation of nano particles. The black colored particles were washed thrice with ethanol, vacuum filtered, oven dried, and pulverized (Allabaksh *et al.*, 2010).

2.4. Characterization of leachate

In this study two different types of leachate were used. The one which was collected from the actual dump site, referred as Aged landfill leachate (ALL). Table 1 represents the composition of solid waste from which the ALL gets generated. In addition to this, the dumpsite also operates an inbuilt composting yard. The leachate generated from this composting yard was referred as Composting yard leachate (CYL). Table 2 represents the composition of solid waste from which the CYL gets generated. All the chemicals used in the study were of analytical grade. The parameters such as pH, BOD, COD, EC,TDS and color were determined using the standard methods prescribed by Bureau of Indian Standards (IS: 3025-2009), and heavy metals were determined using the standard methods prescribed by American public health association (APHA, 2012). Table 3 represents the chemical characteristics of ALL and CYL.

Table 1. Composition of solid waste from which the ALL generated (Ramesh *et al.*, 2009)

Components	Percentage
Paper	3.841
Plastics	5.833
Organic material	4.826
Garden trimmings	60.68
Organic fraction	1.89
Jute	2.8
Wood pieces	5.43
Construction debris	1.05
Glass	1.21
Metal	5.01
Cloth	1.93
Rubber	5.56

Table 2. Composition of solid waste from which CYL generated

Components	Percentage
Wood & wood products	4.76
Pulp, paper &cardboard	4.48
Food, food waste, beverages & tobacco	74.14
Textiles	6.82
Garden, yard & park waste	4.39
Glass, plastic, metal & other inert	5.42

2.5. Characterization of nano particles

Fourier Transform Infrared (FT-IR) Spectroscopy (Thermo Nicolet, AVATAR 330) and Scanning Electron Microscopy (SEM) (ZEISS HR-SEM), Transmission electron microscope

(TEM)(JEOL/ JEM 2100), EDX (Oxford Inca software) were used to characterize the nano particles. FTIR was used to find the presence of functional group in the nano particle, SEM and TEM analysis was used to find the size, shape, structure and internal morphology of the nano particles and in order to find the elemental composition of nano particle EDX analysis was also performed.

Table 3. Chemical characteristics of ALL and CFL

Parameter	ALL	CYL
Age	>10	<5
pH	7.6	7.4
Color	20000	14000
EC	23.420	9.141
TDS	26830	19020
COD	19691	22148
BOD	24000	16000
Na	483	352
K	1657	1118
Cd	Nil	Nil
Cr	0.039	0.007
Cu	Nil	Nil
Zn	0.065	0.004
Fe	Nil	Nil
Pb	Nil	Nil
No_3^-	477.500	452.100

**All in mg l^{-1} except pH, EC (µS cm^{-1}), color (pt.co)*

2.6. Experimental procedure

Batch studies were carried out to perform the experiments using two different nano particles. The nano particle dose was varied from 0.1 to 0.5 g/50 ml. pH was varied between 2 to 10 and the contact time as 30 to 150minutes and the hydrogen peroxide concentration as 1 to 5M. The supernatant was filtered using Whattman 42 filter paper and analyzed for the percentage removal of color, BOD and COD. The color, BOD and COD were analyzed by following the standard methods provided in IS: 3025 (2009). The percentage removal was calculated using Eq. (1).

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

C_i-Initial concentration (mg l^{-1})

C_f- Final concentration (mg l^{-1})

3. Results and discussion

3.1. SEM and EDX Analysis

The surface structure and elemental composition of SNP and INP were studied using SEM and EDX analysis and presented in Figure 1. It can be noted from Fig 1b, the amount of silica content in the raw blast furnace slag was found to be 9.74%. After the extraction of silica from the blast furnace slag the amount of silica present in it was found to be 26.05% (Fig. 1d). The raw blast furnace slag was spherical in shape whereas the SNP was irregular in shape. The SEM images of INP were found to be grain shaped. The presence of larger particles was mainly due to agglomeration of particles. The surfaces of the nano particle seem to be quite uneven and aggregated in appearance.

3.2. TEM Studies

TEM analysis was carried out to examine the microstructure of the nano particle. The TEM images of SNP and INP are presented in the Figure 2(a) and 2(b) respectively. It can be observed from the figure that the particles were irregular in shape. Further, it can be noted that color contrast was observed in INP (Fig. 2b) in which the dark portion indicates the presence of metallic iron and the lighter portion indicates the presence of iron oxide (Yuvakumar *et al.*, 2011). In SNP (Fig. 2a), the particles are irregular in shape.

3.3. FTIR analysis

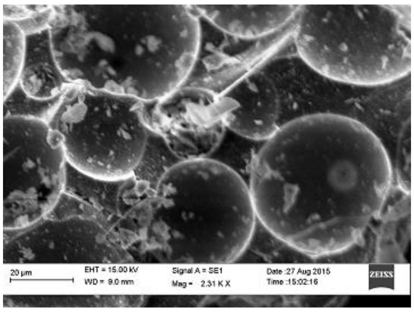
FTIR analysis was carried out to identify the occurrence of functional group present in the sample and the results are presented in Fig. 3a and Fig. 3b for SNP and INP, respectively. It can be noted from Fig. 3a, the peak at 455 cm^{-1} corresponded to Si-O rocking vibration (Singho, 2012). The peak at 800 cm^{-1} was due to the Si-O bending vibration. The strong peak observed at 3400 cm^{-1} indicates the stretching of O-H bond which was mainly due to the presence of H_2O in the sample (Aguiar *et al.*, 2009). Further, it was reported that the SNP have the affinity to absorb water from the atmosphere (Yazdimamaghani *et al.*, 2010) and hence, it can be stated that the obtained SNP were hygroscopic in character. Other than the strong peaks, many medium peaks were also observed which indicates the formation of amorphous phase of SiO_2. From the FTIR spectra of INP (Fig. 3b), it can be observed that the peak at 500 cm^{-1} represents the presence of Iron oxide. The wide band observed in the region between 3200-3400cm^{-1} corresponded to the stretching of -OH bond.

3.4. Batch experiments

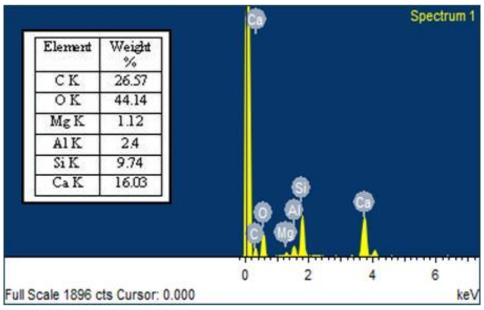
Batch experiments were conducted to study the influence of various operating parameters such as pH, dosage of nano particles, concentration of hydrogen peroxide and contact time.

3.4.1. Effect of pH

The effect of pH on the removal of color, BOD and COD was studied by varying the pH range from 2 to 10. The experiments were carried out for both aged leachate (ALL) and also the leachate from the composting yard (CYL) using SNP and INP by keeping the contact time as 30 minutes with the hydrogen peroxide concentration of 1.5M and nano particle dose of 0.1 g/50 ml and the results are presented in Fig. 4.



(a)



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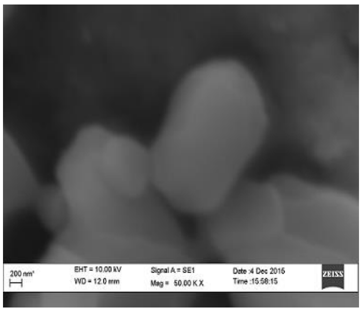
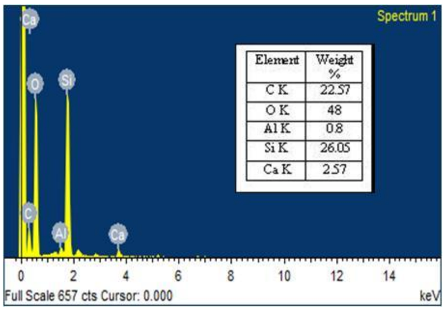
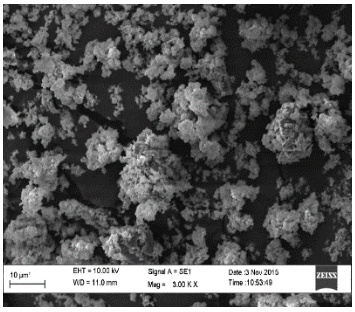


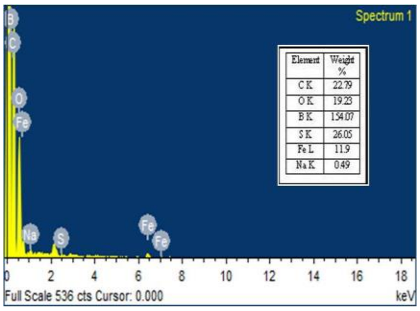
(c)



(d)

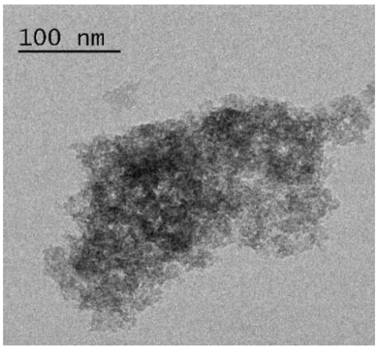


(e)

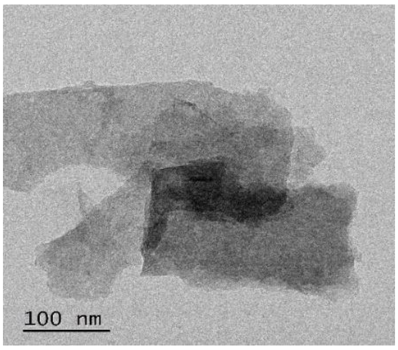


(f)

Figure 1. SEM and EDX of Raw BFS(a,b), SNP(c, d) and INP(e,f)



(a)



(b)

Figure 2. TEM image of (a) SNP and (b) INP

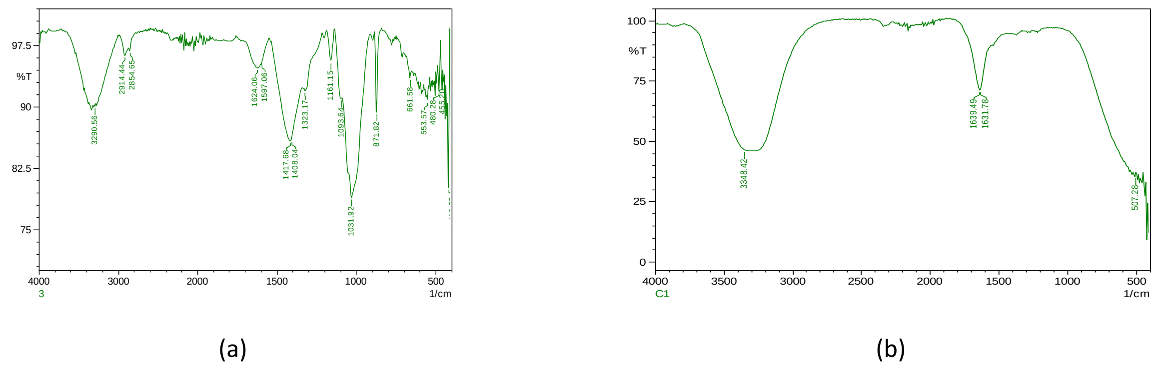


Figure 3. FTIR image of (a) SNP and (b) INP

It can be noted from the figure that the maximum removal of color, BOD and COD for both ALL and CYL were obtained at pH 6, for both SNP and INP, respectively. This is due to the fact that in alkaline condition, the nanoparticles along with hydroxyl radical lead to the production of hydroxide precipitate (Shu *et al.*, 2007). This precipitate occupied the active sites in the nano particles and prevented the further activity. Hence, beyond pH 6 the removal efficiency was found to be constant. The oxidation and reduction potential was more from the pH 2 to 6 and at higher pH no more oxidation was observed. Thus the nano particles were more effective in acidic pH than at alkaline pH. The obtained results were consistent with the results reported in literature (Farrokhi *et al.*, 2009; Jashni *et al.*, 2009).

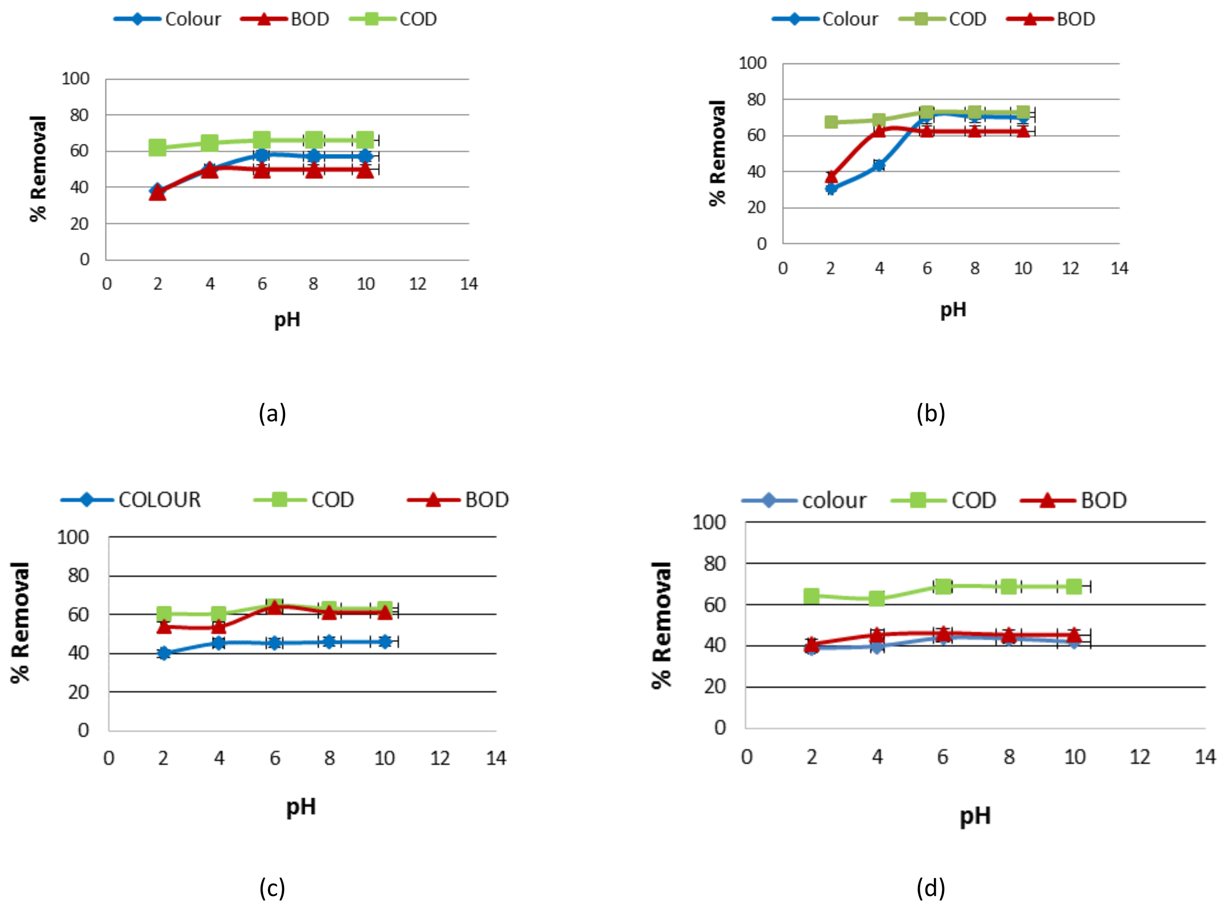


Figure 4. Effect of pH on removal of color, BOD and COD (a) ALL (SNP), (b)CYL(SNP), (c)ALL(INP) and (d) CYL(INP)

Further, it can be noted that for SNP, the removal efficiency of color, BOD and COD for CYL (Fig. 4b) was found to be 70.37%, 62.50% and 73.09% respectively and for ALL (Fig. 4a) it was found to be 57.82%, 50.16% and 66.01% respectively. Similarly for INP application in CYL, the removal efficiency of color, BOD and COD was found to be 49.78%, 63.75%, 66.01% respectively (Fig. 4d) and for ALL it was found to be 59.39%, 55.45, and 77.34% respectively (Fig. 4c). The difference in removal efficiency was due to the presence of large amount of organic compound in CYL than that of ALL.

3.4.2. Effect of nano particles dosage

The effect of SNP and INP dosage on the removal efficiency of colour, BOD, COD are presented in the Figure 5. By keeping the other parameters constant (pH 6, contact time 30 minutes and hydrogen peroxide concentration 1.5M), the experiments were carried out for ALL and CYL by varying the SNP dosage from 0.1 to 0.5 g/50 ml. The maximum removal efficiency for SNP was observed at the dosage of 0.4g/50 ml. At this dosage the removal efficiency of color, BOD, COD was found to be 50.35%, 62.59%, 67.42% and 72.28%, 65.94%, 68.81%, for ALL and CYL respectively. Similarly, using INP, the maximum percentage removal was observed at the dosage of 0.3g/50 ml, at which the maximum removal was found to be 54.50%, 54.56% and 64.73% for ALL and 56.49%, 63.98% and 68.37% for CYL respectively. By increasing the active surface area there was more possibility of interaction within nano particle and organic, inorganic substances present in the leachate. Petrisevski *et al.,* (2007) reported that increasing the oxidation and reduction reaction is also the possible reason for the removal efficiency. Further, it can be noted from the figure that the removal efficiency found to be almost constant beyond 0.4 g/50ml for SNP and 0.3g/50 ml for INP. This is due to the fact that the excess amount of nano particles causes turbidity and reduces the oxidation potential and interference in leachate treatment (Shu *et al.*, 2010). Shu, (2007) reported similar trend in azo dye removal using nanoparticles and the removal efficiency was found to be 95%. Shao–Feng, (2005) reported that the removal of hexavalent chromium with nano particle was found to be 100% at 0.4 g l^{-1} and the removal was found to be 26% in 0.1 g l^{-1}.

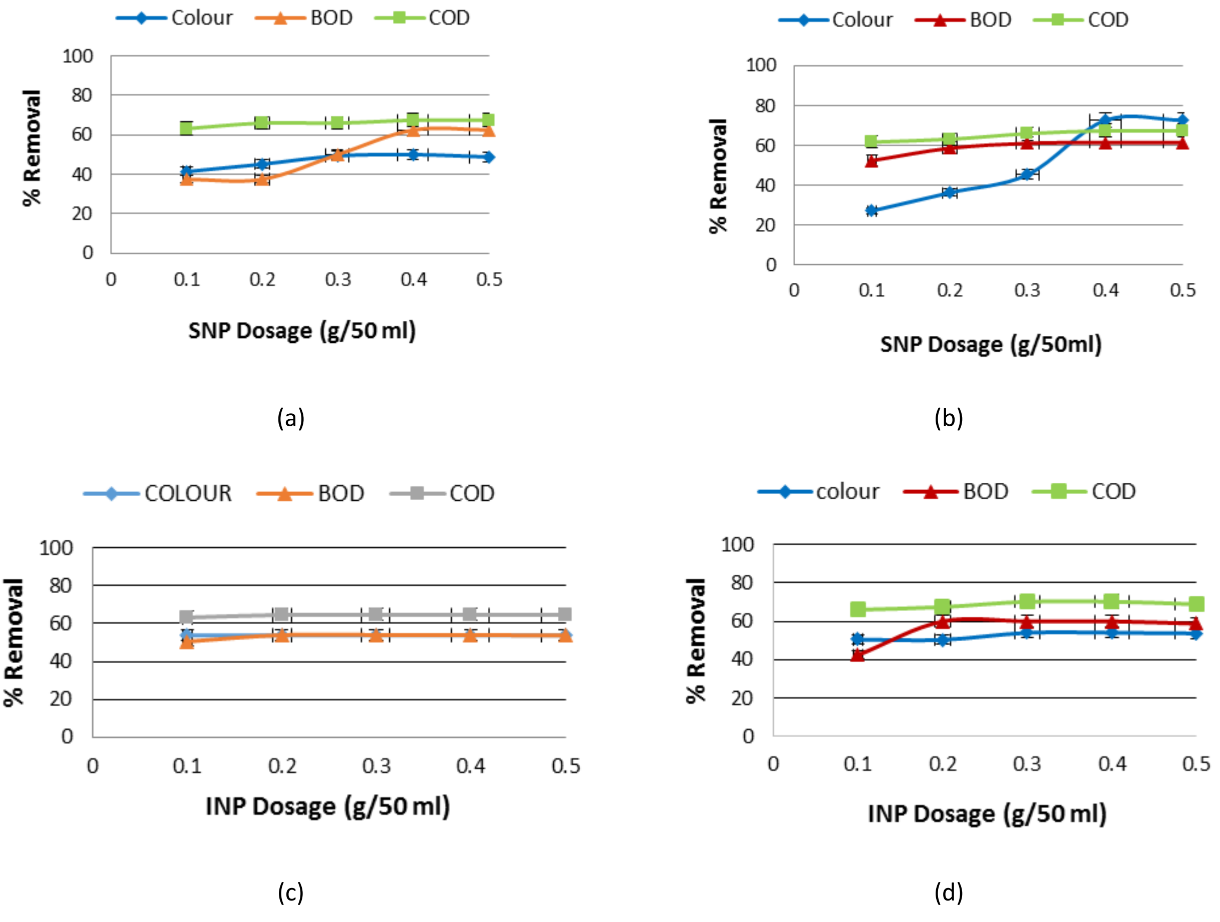


Figure 5. Effect of nano particle dosage on the removal of Color, BOD and COD
(a) ALL(SNP),(b) CYL(SNP), (c) ALL(INP) and (d) CYL(INP)

3.4.3. Effect of contact time

The effect of contact time was studied by keeping other parameters as constant, i.e., pH 6, adsorbent dosage 0.4& 0.3 g/50 ml, Hydrogen peroxide 1.5M and varying the contact time in the range of 30 - 150 minutes (Figure 6). The optimum contact time was found to be 90 minutes at which the maximum removal of color, BOD, COD was achieved for CYL and ALL. The maximum percentage removal of color, BOD,COD was obtained as 67.90%, 75.83%, 81.95% and 74.87%, 72.41% and 84.84% for ALL (Fig. 6a) and CYL (Fig. 6b) respectively, for SNP. It can be noted from the figures that increase in contact time increases the removal efficiency. It signifies that the reaction between the silica nano particle and hydrogen peroxide was almost completed between the reaction time of 30 to 90 minutes. Similar trend was observed for INP, with pH 6, nano particle dosage 0.3g/50 ml, the maximum removal efficiency of ALL (Fig. 6c) and CYL (Fig. 6d) was found to be 64.73%, 56.36%, 67.43% and 65.05%, 55.37%, 68.94% respectively, at 120 minutes. The obtained results are consistent with the results reported in literature for leachate treatment (Farrokhi *et al.*, 2009, Jashni *et al.*, 2008).

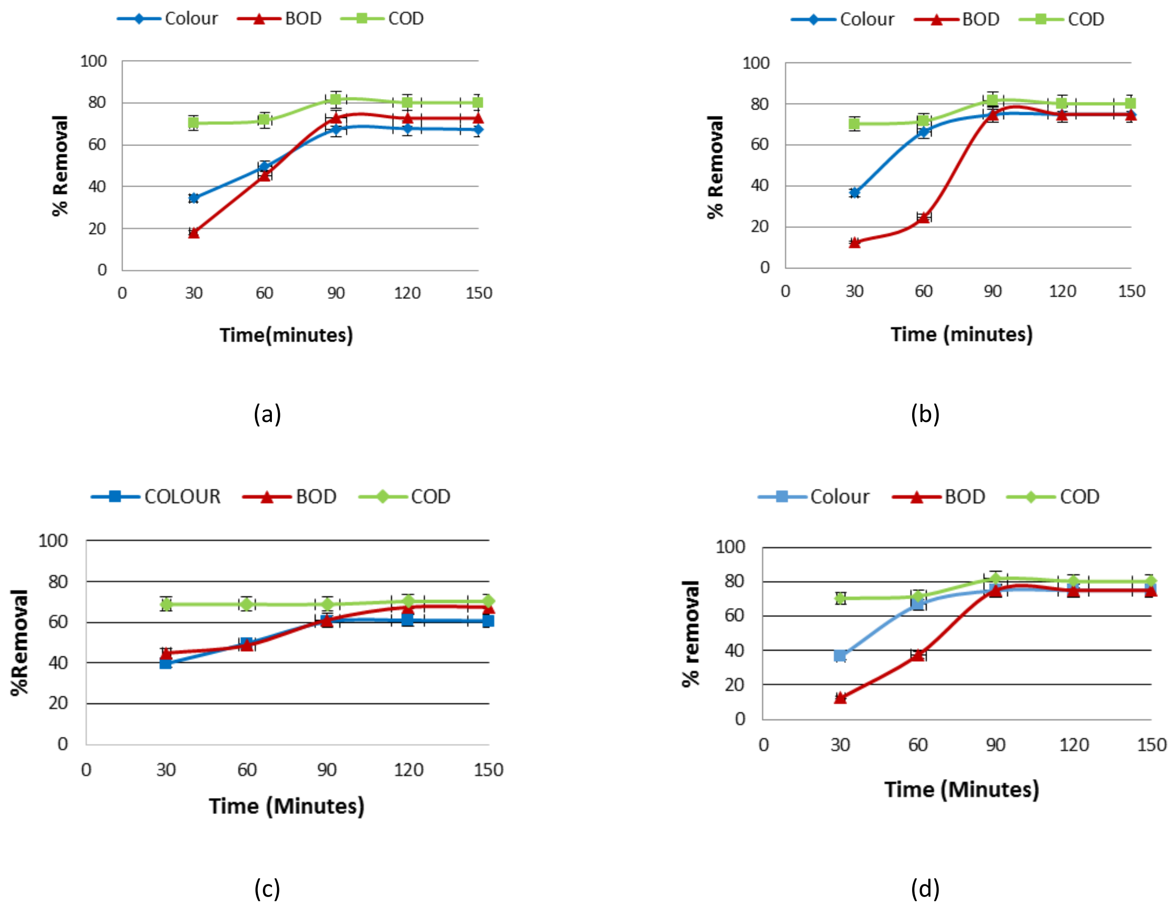


Figure 6. Effect of contact time on the removal of color, BOD and COD (a)ALL (SNP),(b)CYL(SNP), (c)ALL(INP) and (d) CYL(INP)

3.4.4. Effect of Hydrogen peroxide concentration

With optimized pH as 6 and nano particle dosage as 0.4 g/50 ml and 0.3 g/50 ml (for SNP and INP respectively) and contact time as 90 and 120 minutes (for SNP and INP respectively) the hydrogen peroxide concentration was varied from 1.5M to 3.5M and 1M to 5M for SNP and INP respectively. The removal efficiency of color, BOD and COD was found to be 77.15%, 62.72%, 87.15% and 82.56%, 62.51%, 77.34 % for ALL (Fig. 7a) and CYL (Fig. 7b) respectively, for SNP. It can be noted from the figure that the maximum removal efficiency was achieved at 3M. Similar trend was observed by using INP and maximum removal efficiency was found to be 57.06%, 57.27%, 67.43% for ALL (Fig. 7c) and 60.36%, 65.09%, 67.43% for CYL (Fig. 7d) respectively, for the Hydrogen peroxide concentration of 4M.

4. Conclusions

The potential of SNP synthezed from blast furnace slag, a waste obtained from the foundary unit and INP synthesized using chemicals, for the removal of color, BOD and COD was studied. The effect of pH, contact time, nano particle dosage and hydrogen peroxide concentration were investigated and the optimum condition for maximum removal using SNP was found to be pH 6, contact time 90 minutes, nano particle dose 0.4g/50mL and hydrogen peroxide concentration 3M. Similary, for INP it was found to be pH 6, contact time 120 minutes, nano particle dosage 0.3g/50 mL and hydrogen peroxide concentration 4M. Based on the obtained results, it can be concluded that the removal efficiency using both SNP and INP was comparitively high for CYL than the ALL due to the nature of leachate characteristics.

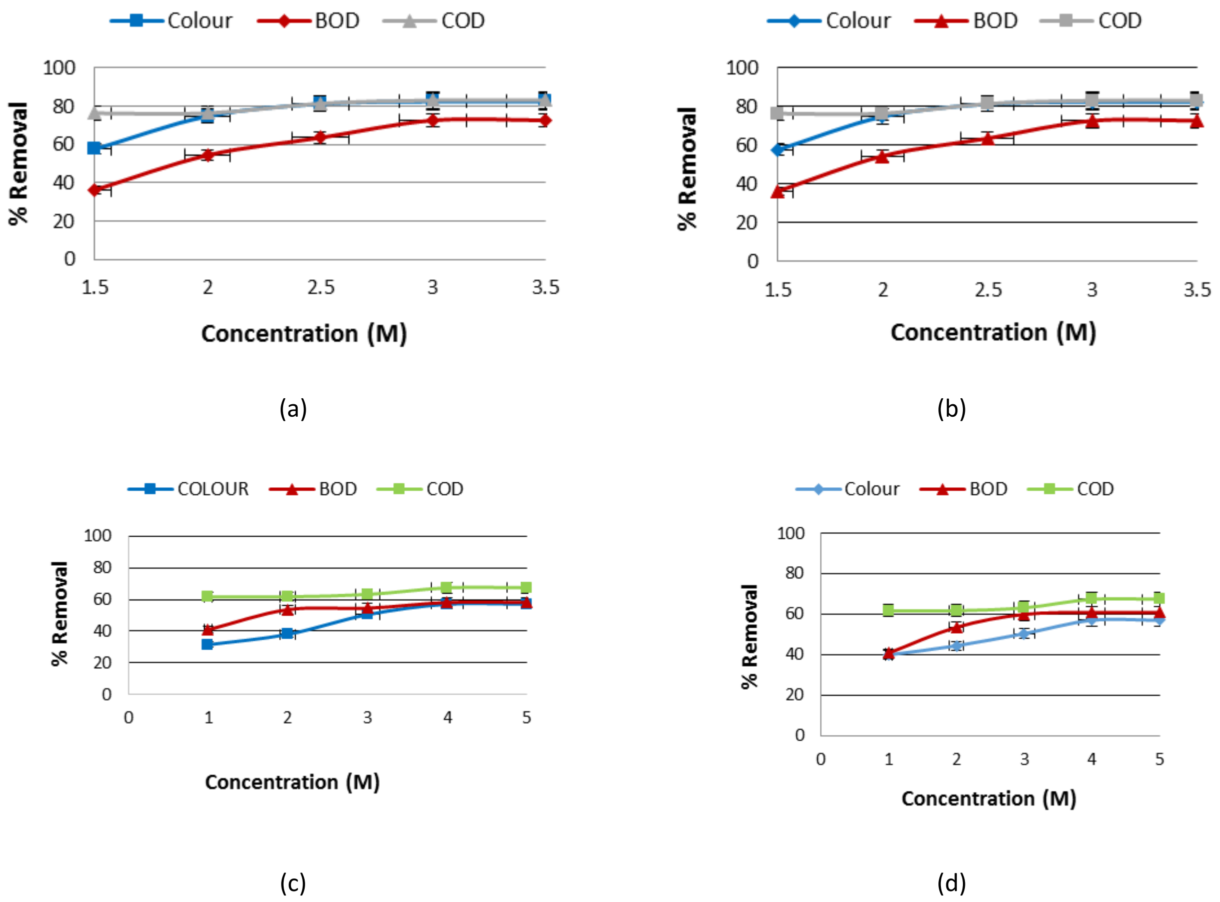


Figure 7. Effect of hydrogen peroxide on the removal of color, BOD and COD (a)ALL (SNP),(b) CYL(SNP), (c) ALL(INP) and (d) CYL (INP)

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