Synthesis of a new resin functionalized with xanthine moiety and its application for separation of nickel and cadmium in natural water

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A new chelating resin has been synthesized by functionalization of polystyrene-divinylbenzene (8%) with xanthine through -N=N- bonding. The resulting resin has been characterized by elemental analysis, thermogravimetric analysis, infrared spectroscopy, hydrogen ion capacity and capacity of metal ions. The separation and mesurement of Ni(II) and Cd(II) were carried out with this resin and the maximum exchange capacity of Cd(II) was found to be 0.73 mmol g^{-1} and that for Ni(II) was 0.57 mmol g^{-1} at pH 1.0 for both. The eluents 10% thiourea in 0.1 M $HClO_4$ and 1.0 M sulphosalicylic acid have been used for separation of Cd(II) and Ni(II), respectively. The detection limits for Ni(II) and Cd(II) were found to be 3.6 and 4.1 ng mL^{-1}, respectively . The optimum flow rate and sample breakthrough volume of column operation for both the metal ions were found to be 1.2 mL min^{-1} and 3000 mL, respectively. The developed method was applied for determination of these two metal ions in synthetic binary mixtures. The developed method was also applied to natural water sample and compared with the results of previously developed method

Nickel and cadmium are metals of toxicological concern. Cd is a cumulative toxicant having elimination half life of 10-30 years[1], the environmental exposure of which is implicated by a number of clinical complications like renal dysfunction and bone diseases along with cancers. Bone disease "Itai-Itai" resulting from prolonged exposure is a mixed osteomalacia and osteoporosis due to accumulation of cadmium in bones as well as kidney damage manifested by severe pain, multiple fracture, distorted long bone and skeleton[1-3].

Ni toxicity is primarily manifested by respiratory disorders like chronic bronchitis, emphysema and impaired lung functions. Symptom of Ni toxicity includes skin rash (nickel dermatitis), nausea, dizziness, diarrhoea, headache, vomiting, chest pain, weakness and coughing.

Due to certain similarities in chemistry of these two metal ions, selective separation and measurement is an important analytical technique, especially when the metal ions are present in trace levels. Thus, methods capable of overcoming the limitation caused by matrix effect in complicated samples like industrial wastewater need to be developed. Separation followed by preconcentration of identified analyte, is one of the best ways to address this problem.

Solvent extractions[4] and ion exchange[5] have been used widely for the separation and preconcentration of trace heavy metals. The disadvantages of solvent extraction includes high solvent requirement which not only increases the cost of the process but also creates environmental pollution due to the disposal and handling of huge amount of solvents. Chelating resins can be applied very easily for such separation because of their extraordinary absorption property, specificity towards metal ions and simplicity in use.

Thus, there is a growing interest for the development of new functionalized polymers capable of offering good stability, higher capacity and flexibility in working conditions for the separation and preconcentration of trace metal ions from complex samples[6-9].

Functionalization of polymer by incorporation of chelating ligand molecule onto polymer surface through covalent bonding to induce selectivity towards metal ions, provides rugged polymer, free from problem of ligand leaching. Various chelating agents like 1-nitroso-2-naphthol[10], pyrocatechol violet[11], thiosalicylic acid[12], xylenol orange[13], pyrocatechol[14], o-amino phenol[15], tiron[16], pyrogallol[17], nitroresorcinol[18], methylthiourea[19], guanyl thiourea[19], dithiocarbamate[19], cyclohexene oxide[20], cyclohexene

sulphide[20], acyloxime[21], 1-hydrazinophthalazine[22], ethylenediamine[23], 8-hydroxoquinoline[24], aminophosphonate[25], aminomethylphosphonate[26], phenylphosphonic acid[27] were incorporated for preconcentration of Ni and Cd. Synthesis of a chelating resin of high exchange capacity, lower detection limit and free from interference of foreign ions for these metal ions are most challenging.

Xanthine is a bioactive molecule having an important role in renal function. It was, therefore, worthwhile to incorporate this ligand onto polymer surface which could be able to bind and separate these metal ions from complex samples.

Experimental Procedure

Atomic absorption spectrometric measurements were carried out with GBC Avanta Spectrometer with the following conditions for Ni: lamp current 4 mA at wavelength 231.1 nm and for Cd: lamp current 3 mA at wavelength 228.8 nm. The pH measurements were carried out with a Systronics 362 pH meter. Infrared spectra were recorded on a 420 JASCO FTIR. Nitrogen content of dried samples of the resin was determined by using a Perkin-Elmer 240C elemental analyzer and the thermogravimetric analysis of the sample of dried resin was carried out by a Shimadzu TG50/DT50.

Stock solutions (1000 ppm) of Cd(II) and Ni(II) were prepared by dissolving appropriate amounts of analytical reagent grade cadmium nitrate and nickel sulphate hexahydrate (both BDH, Mumbai), respectively in double distilled water and the solutions were standardized before use. Polystyrene–DVB (8%) resin (Bird and Company, Mumbai), xanthine (SRL, India) were used as received. All other chemicals were of reagent grade. The glass apparatuses were soaked in 4 M nitric acid overnight and cleaned with double distilled water before use.

Sludge samples were collected from different points in the Durgapur industrial area (West Bengal, India). The river Tamla nalah flows through Durgapur industrial belt and carries effluent from different industries. The stations selected were as follows: Station 1- Durgapur Chemicals Ltd. (DCL) main drain; Station 2: Junction of Tamla nalah and main drain of DCL; Station 3 : Tamla nalah near alloy steel plant, Durgapur.

Air dried styrene DVB copolymer (5 g, 30-60 mesh) was swollen in chloroform, then filtered and nitrated by 25 mL 18 mol L^{-1} H$_2$SO$_4$ and 10 mL 15 mol L^{-1} HNO$_3$ by stirring at 60°C for 1 h. After that, reaction mixture was poured into an ice-water mixture, then the product was filtered and washed thoroughly with water until free from acid. Next nitrated resin was heated with a mixture of 40 g SnCl$_2$, 12 mol L^{-1} HCl in 50 mL ethanol and refluxed for 20 h at 120°C. The precipitated mass was then filtered off and washed with water and then with 2 mol L^{-1} NaOH followed by 4 mol L^{-1} HCl in order to remove the excess SnCl$_2$. The amino resin was then diazotised according to the literature procedure[28]. The diazotised resin was then treated with xanthine (3.5 g) in NaOH at 0-5°C for overnight. The resulting dark brown resin was filtered and washed with double distilled water and dried in air. Finally the resin with 30-60 mesh size was retained for use.

A 0.5 g portion of the final resin was kept immersed in 12 mol L^{-1} HCl or HNO$_3$ and 12 mol L^{-1} NaOH for 7 days then filtered off and washed with double distilled water. After air drying, nitrogen content and exchange capacity for Ni(II) and Cd(II) as well as H$^+$ ion capacity were determined. Thermogravimetric analysis of the resin was done to determine the stability of resin towards various temperatures.

The nitrogen content of nitrated resin, amino resin and the final xanthine resin were determined by elemental analyzer. The amino group content of the amino resin was determined by non-aqeous tritimetry[29].

A 1.0 g portion of the final resin was converted to its acid form by treating with 6 mol L^{-1} HCl. The resin was filtered off, washed with double distilled water until free from acid. The acidic form of the resin was then equilibrated with 20 mL of 0.1 mol L^{-1} NaOH for a period of 12 h and the excess alkali was back-titrated with standard 0.1 mol L^{-1} HCl in order to determine the hydrogen ion capacity.

In order to find the sorption ability at different pH, a batch equilibration technique was used. 25 mL of the sample solution containing 100 μg mL^{-1} of metal ion was taken in a stoppered glass bottle. The pH of the solution was adjusted to 1-7 with the help of HCl or sodium acetate buffer. Then 50 mg of the resin was added to each of the bottles and shaken for 10 h. The resin was filtered, washed with water and sorbed metal ion was eluted by suitable eluting agent and concentration was determined by flame AAS.

A portion of 0.1 g of the resin with maximum sorbed species was shaken for 24 h with various

eluting agents *viz.* 0.1-1.0 M HCl, 0.1-1.0 M HNO$_3$, 0.01-1.0 M sulphosalicylic acid, 1-10% thiourea solution in 0.1 M HClO$_4$. Then in each case, resin was filtered off and eluted metal ion was measured at the specified condition.

To determine the time of equilibrium for Ni(II) and Cd(II) at *p*H 1.0, the metal ion solution (30 mL, 1000 μg mL^{-1} each) was placed in stoppered bottles and shaken. At regular time interval, the solution was filtered and sorbed metal ion was eluted with suitable eluent from the resin bead and concentration of metal ion was measured by flame AAS.

A 120 × 8 mm glass column was used for the present study. A 500 mg portion of the final resin was suspended in double distilled water and the column was packed by passing the suspension of the resin so that heavier particles occupy the lower half of the column to prevent choking. Then column was washed with double distilled water followed by an appropriate buffer solution.

Maximum sample volume upto which quantitative metal sorption takes place was determined by passing sample volume over a range of 500–5000 mL containing 200 μg of metal ion of interest through preconditioned resin column. The sorbed metal ion was eluted by 10 mL of appropriate eluent. The percent sorptions for different sample volume were determined to find sample breakthrough volume.

Detection limits are crucial for any newly developed method as it gives idea about its sensitivity towards analyte extraction. Studies by passing 3000 mL of sample solution through the preconditioned resin bed containing metal ions in the range of 20-150 μg was carried out for this purpose.

To confirm that the active sites for the sorption of metal on resin surface is xanthine moiety, the final resin was boiled with 1.0 mol L^{-1} sodium carbonate for 24 h to replace the diazo group by hydroxyl ion. After filtration the H$^+$ ion capacity as well as capacity for Ni(II) and Cd(II) of the resulting resin was found close to zero.

To optimize the flow rate of analyte for loading, 500 mL of the sample solution of metal ion containing 2 μg mL^{-1} adjusted to proper *p*H was passed through the column at different flow rates in the range of 0.1-2.0 mL min^{-1}. The percent recoveries of the metal ion of interest at different flow rates were determined. Similarly, to optimize the flow rate of elution, appropriate eluent already optimized was passed through the column containing maximum sorbed

metal ion at different flow rates and percent recoveries at different flow rates of eluent were determined.

To study the effect of diverse ions on the sorption and desorption of metal ion on resin, a suitable aliquot containing 2 μg mL^{-1} with 100 times excess of diverse ions were allowed to pass through the preconditioned column at flow rate of 0.8 mL min^{-1}. The stripping of the metal ion was carried out by using already optimized eluent in a 10 mL calibrating flask and determined by flame AAS.

Results and Discussion
Characterization of resin

Starting from styrene divinylbenzene copolymer (8%, 30-60 mesh), the final resin was synthesized according to Scheme 1. The nitrogen content of compound II was 10.62%. After the reduction, the nitrogen content of the amino resin was 10.92%. The amino group estimation by non-aqeous titrimetry of amino resin shows that its content is 2.85 mmol g^{-1}, which accounts for 38% conversion. The nitrogen content of xanthine resin was 10.55 mmol g^{-1} (14.78%). An empirical composition of the final resin was obtained by consideration that diazotisation is complete but subsequent coupling is not. The uncoupled diazonium compound will ultimately decompose leaving the corresponding phenolic compound. The final product, therefore, contains 1.06, 4.71, 1.79 mmol g^{-1} of xanthine, unconverted nitro and phenolic – OH resin, respectively.

The infrared study of the resin shows strechings at 1517 cm^{-1} for -*N*=*N*-, at 1626 cm^{-1} for -C=N-, 1685 cm^{-1} for -C=O strechings. Other vibrations for xanthine skeleton are the indication of incorporation of xanthine moiety onto resin surface (Table 1).
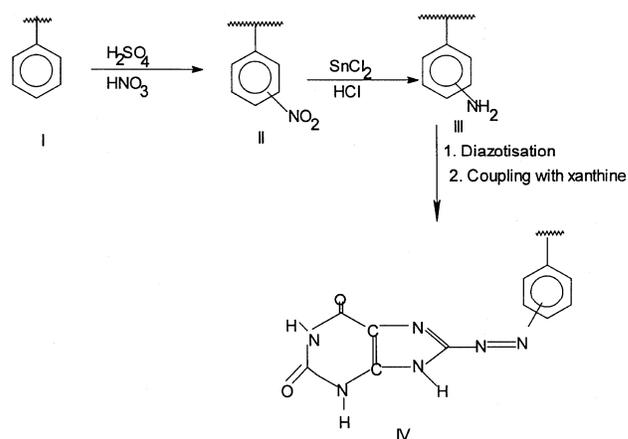
Scheme 1 —Synthesis of xanthine resin

Table 1 — Physical and chemical characteristics of xanthine resin

Colour	: Redish brown
Bead size	: 30-60 mesh
Thermal stability	: 300°C
Hydrogen ion capacity	: 1.17 mmol g^{-1}
Equilibration time($t_{1/2}$)	
for Ni(II)	: 11 min
for Cd(II)	: 14 min
IR data	: 1517 cm^{-1} for - N=N-(m), 1626 cm^{-1} -C=N-(s), 1685 cm^{-1} for -C=O(s), 1408 cm^{-1} for –C–N (m) strechings. Other vibrations like 2353,1341 and 701 cm^{-1} are due to xanthine moiety into polymeric matrix

Table 2 — Desorption of Ni(II) and Cd(II) using different eluents

Eluents	% Recovery	
	Ni(II)	Cd(II)
1.0 M HCl	27.2	38.1
0.1 M HNO$_3$	13.1	22.4
0.5 M HNO$_3$	32.4	46.3
1.0 M HNO$_3$	41.2	53.1
1% Thiourea in 0.1 M HClO$_4$	-	27.1
3% Thiourea in 0.1 M HClO$_4$	-	40.2
5% Thiourea in 0.1 M HClO$_4$	-	63.3
7% Thiourea in 0.1 M HClO$_4$	-	78.4
10% Thiourea in 0.1 M HClO$_4$	-	100.1
0.01 M Sulphosalicylic acid	36.1	-
0.1 M Sulphosalicylic acid	67.3	-
0.5 M Sulphosalicylic acid	83 .2	-
1.0 M Sulphosalicylic acid	100.3	-

Chemical stability of the resin was examined by treating with 12 M HCl or 12 M NaOH. After filtration, the nitrogen content and hydrogen ion capacity as well as capacities for Ni(II) and Cd(II) were measured. No significant change of these parameters were observed. So it may be concluded that the resin is fairly stable in strong acid or alkali medium. Thermogravimetric analysis of the resin shows that the resin is stable upto 300°C.

The time required for uptake of metal ion, 50% of its maximum capacity for Ni(II) and Cd(II) were found to be 11 and 14 min, respectively. Thus, the resin is suitable for column operation under a low flow rate condition. The hydrogen ion capacity measured by previously mentioned method found to be 1.17 mmol g^{-1}

Effect of pH

The sorption behaviour of Ni and Cd onto the resin by batch technique are shown in Fig.1. The maximum exchange capacities were found to be 0.73 and 0.57
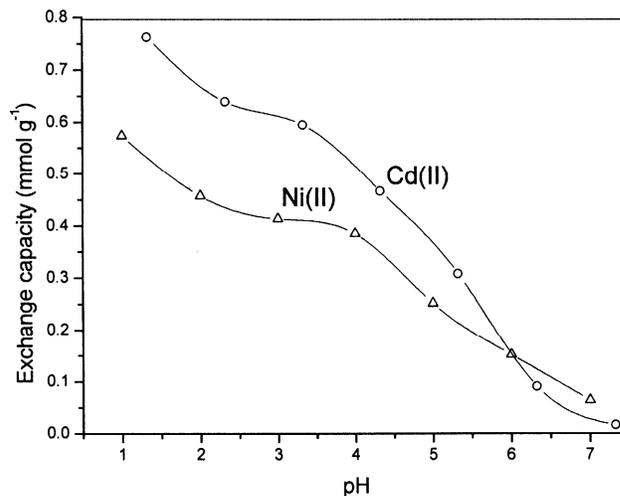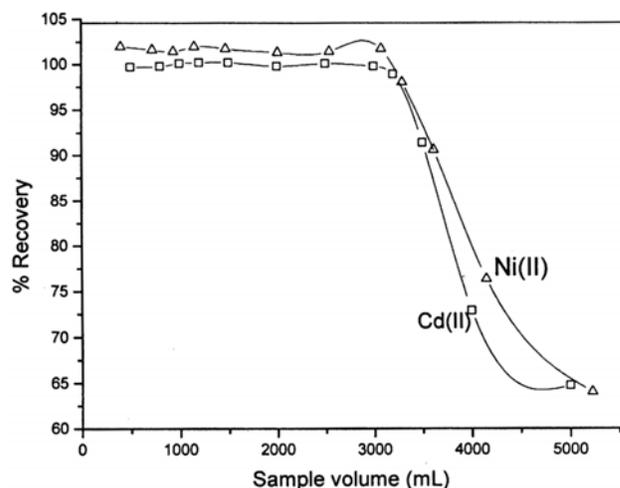

Fig. 1—Exchange capcities (mmol g^{-1}) for Ni(II) and Cd(II) at different pH

Fig. 2—Sample breakthrough volume curve for sorption of Ni(II) and Cd(II)

mmol g^{-1} for Cd(II) and Ni(II) respectively at pH 1.0. The sorption at lower pH may be due to formation of diazo cation that subsequently attacks negative species like CdCl$_4^{2-}$ and NiCl$_4^{2-}$. At pH> 5 there is less possibility of stabilization of diazo cation; also at even higher pH the metals would be precipitated. Thus, the exchange capacity at pH 6-7 is very low. The effects of different eluents on the desorption of metal ions from resin are indicated in Table 2. Complete desorption took place with 25mL 1.0 M sulphosalicylic acid for Ni(II) and 25 mL 10% thiourea in 0.1 M HClO$_4$ for Cd(II).

Sample breakthrough volume and lower limit of detection

The plot of percent recovery against sample volume is shown in Fig. 2. It was found that percent

sorption drops with sample volume after the 3000 mL mark. Upto sample volume of 3000 mL the recovery is greater than 99.5% and it gives a preconcentration factor of 400. This reflects the enhanced accessibility of chelating sites onto the resin.

Effect of diverse ions

The effect of higher concentrations of diverse ions on the sorption of Ni and Cd were examined by batch equilibration experiments and the results are shown in Table 3. The presence of diverse metal ions like alkali, alkaline earth, first transition series including mercury did not interfere in the sorption process. But presence of permanganate and vanadate ion gave some small interference to the sorption. However, the second transition series anions did not offer significant interference. Hence, attempts were made to separate Ni and Cd from each other from industrial sludge samples.

Effect of flow rate of sample solution

The flow rate of the sample affects the retention of both Ni(II) and Cd(II) on the resin. Therefore, the effect of flow rate of sample solution was examined with the column adjusted under optimum condition of pH and eluent. The flow rate higher than 1.2 mL min^{-1} causes a decrease in percentage of sorption. So a flow rate of 1.2 mL min^{-1} was chosen for column operation as optimum flow rate (Fig. 3).

Reuse and stability of resin

In order to test the ability for its reuse, the resin was subjected to several cycles of loading and elution by batch equilibration technique as well as column method. It has been found that the exchange capacity of the resin practically remained unchanged after its repeated use for more than 50 times. Thus, the multiple use of this resin is possible. After one year of storage of resin in a cool dry place, no significant change in exchange capacity was found.

Analytical performance characteristics

The sensitivity of a resin towards metal ion of interest is characterized by lower concentration below which quantitative sorption by chelating matrix is not perceptibly seen. Using the optimum condition of sorption (pH, sample volume and flow rate) detection limit of 3.6 and 4.1 ng mL^{-1} for Ni(II) and Cd(II) were achieved. This lower value of detection limit suggests the higher sensitivity of the final resin. The RSD of determination of concentrations of Ni(II) in industrial waste water was 2.1, 2.3 and 2.4% for Station 1, Station 2 and Station 3, respectively. Again those

Table 3 —Separation of 2 μg mL^{-1} each of Ni(II) and Cd(II) from several binary mixture in sample of 50 mL at pH 1.0

Foreign ions*	% Recovery	
	Ni(II)	Cd(II)
Hg(II)	99.3	96.7
Ag(I)	101.1	99.9
Fe(III)	96.7	98.6
Cu(II)	99.1	100.6
Co(II)	97.8	99.4
Mn(II)	96.8	100.1
Na(I)	99.7	101.2
K(I)	101.3	98.9
Ca(II)	99.6	99.2
Ba(II)	101.2	98.7
Zn(II)	97.2	99.4
MnO$_4^-$	93.2	91.7
PO$_4^{3-}$	98.9	99.6
VO$_3^-$	91.4	89.7
AsO$_4^{3-}$	99.1	98.7
CrO$_4^{2-}$	99.8	98.9
MoO$_4^{2-}$	98.6	100.2
WO$_4^{2-}$	100.1	99.8

*In each case, the amount of foreign ion added was 2000 μg
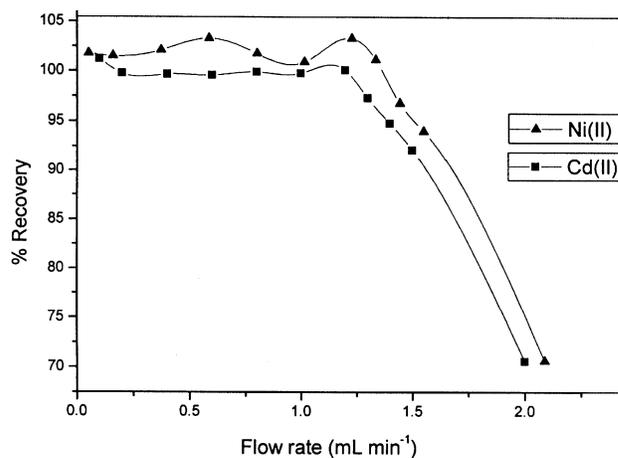

Fig. 3—Flow rate optimization for Ni(II) and Cd(II) absorption on xanthine resin.

values for Cd(II) were respectively 2.3, 2.2 and 2.2% for Station 1, Station 2 and Station 3, respectively.

Applications

Separation of Ni(II) and Cd(II) in binary synthetic mixtures

Different amounts of each of Ni(II) and Cd(II) were mixed having a total volume of 100 mL and each solution was subjected to column operation under optimum conditions. Now Ni(II) was eluted out first using 1.0 M sulphosalicylic acid solution followed by elution of Cd(II) by 10% thiourea solution in 0.1 M HClO$_4$, and the concentrations of the metals were measured as mentioned earlier (Table 4).

Table 4 — Separation of Ni(II) and Cd(II) in binary mixtures

No of observation	Amount taken(µg)	Amount found(µg)*	% Error
1	Cd(II) : 10	Cd (II) : 9.90 ± 0.30	1.0
	Ni(II) : 10	Ni (II) : 10.15 ± 0.20	1.5
2	Cd(II) : 1	Cd (II) : 0.96 ±0.03	4.0
	Ni(II) : 10	Ni (II) : 9.80 ± 0.08	2.0
3	Cd(II) : 1	Cd (II) : 0.97 ± 0.07	3.0
	Ni(II) : 1	Ni (II) : 0.98 ± 0.02	2.0
4	Cd(II) : 10	Cd (II) : 10.12 ± 0.03	1.2
	Ni(II) : 1	Ni (II) : 0.98 ± 0.08	2.0
5	Cd(II) : 100	Cd (II) : 102.12 ± 0.03	2.1
	Ni(II) : 10	Ni (II) : 9.70 ± 0.08	3.0
6	Cd(II) : 10	Cd (II) : 10.11 ± 0.03	1.1
	Ni(II) : 100	Ni (II) : 98.60 ± 0.08	1.4
7	Cd(II) : 100	Cd(II) : 102.30 ± 0.03	2.3
	Ni(II) : 1	Ni(II) : 0.95 ± 0.08	5.0
8	Cd(II) : 1	Cd(II) : 1.04 ± 0.03	4.0
	Ni(II) : 100	Ni(II) : 98.20 ± 0.08	1.87

*Average of five determinations

Table 5—Determination of N(II) and Cd(II) in industrial waste water after separation using developed technique

Sample no.	Ni(II) present method (ng mL^{-1})^a	Cd(II) present method (ng mL^{-1})^a	Ni(II) earlier method[15] (ng mL^{-1})^a	Cd(II) earlier method[15] (ng mL^{-1})^a
1	58.2 ± 0.8	89.3 ± 0.8	59.7 ± 0.5	87.4 ± 0.8
2	32.8 ± 0.4	46.1 ± 0.7	31.6 ± 0.2	46.8 ± 0.6
3	46.9 ± 0.6	65.3 ± 0.4	47.2 ± 0.6	66.1 ± 0.2

^{a}Average of 5 determinations

Analysis of industrial waste water

The samples of industrial waste water collected from Durgapur industrial area were filtered through 0.45 µm Milipore membrane filter. Taking 500 mL of this filtered sample *p*H was adjusted to 1.0 and passed through resin column at a flow rate of 1.2 mL min^{-1}. After elution the concentrations of Ni(II) and Cd(II) were measured in each case. The results of the analysis are shown in Table 5.

The results were compared with the determination of Ni(II) and Cd(II) by an earlier developed method[15] using Tiron chelating resin.

Conclusion

The application of the newly synthesized xanthine resin offers an excellent means for separation and preconcentration of trace Ni and Cd from complex samples like industrial water. The main advantage of the method is that the separation and preconcentration can be achieved by absorption of both the Ni(II) and Cd(II) at same *p*H followed by successive elution of Ni(II) and Cd(II) by different eluting agents. As the eluents are specific for metal ions any sequence of elution can be applied. The high exchange capacity of

the resin and appreciably low detection limit with high sample breakthrough volume makes it capable of preconcentrating trace concentration of metal ions from a large volume of sample and thus attains a high sensitivity of the method. The proposed method is simple, rapid, selective and free from matrix interferences of ions normally present in natural, industrial or environmental samples.

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