Removal of Ni (II) By Sol-Gel Silica Functionalized With Efficient Metal Chelating Agents

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Abstract

In this study a comprehensive investigation of doping of sol-gel silica with efficient metal chelating agents is provided at room temperature using infrared spectroscopy, elemental analysis, SEM, EDS and XRD. Sol-gel silica was doped with a series of metal chelating agents to investigate the sorption of Ni (II) ions from aqueous media. It is observed that the chelating agents are encapsulated by the pores of the xerogel from where they interact with the metal ions by ion exchange and chelation mechanism. Nickel complexes of the chelating agents were also synthesized. Sol-gel silica was doped with metal complexes for comparison with sol-gel silica having sorbed metal ions. Same amount of Ni (II) was observed in both samples, showing high efficiency of the method for removal of Ni (II). The batch adsorption experiments were performed at room temperature and neutral pH. The advantage of the method is high sorption capacity at room temperature and neutral pH. The developed method can be applied on large scale removal of toxic heavy metals. The sorption process completed within a minute. 0.1 N HNO₃ resulted in complete desorption of metal ions from the gels. The regenerated sorbents were reused several times with negligible loss of sorption capacity.

Highlights

- Sol-gel silica was functionalized with efficient metal chelating agents.
- Sol-gel silica was also functionalized using metal complexes of the chelating agents for comparison.
- Sol-gel silica functionalized with chelating agents was used for the removal of Ni (II).
- Functionalized sol-gel silica showed high sorption capacity and fast desorption of Ni (II) at room temperature and neutral pH.
- The sorption process completed within one minute. 0.1 N HNO₃ resulted in complete desorption of metal ions from the xerogels.
- The developed method can be applied on large scale removal of toxic heavy metals.

Introduction

Water pollution is the major challenge that human beings are facing since long. Naturally, weathering and erosion of rocks is the main source of toxic heavy metals. Along with natural sources there are anthropogenic activities such as mining, welding, metallurgical operations, alloys manufacturing, fertilizers, pesticides and some other industries [1].

Among heavy metals, nickel is highly toxic. Volcanic rocks and soil naturally contain nickel. Nickel is used in aircrafts, automobiles, nickel–cadmium batteries, cosmetics and coins. Mineral leaching by weathering of rocks to water reservoirs is the cause of water pollution [2]. Water soluble nickel salts pollute water systems [3]. Enamel and paint industries also release nickel containing waste matter in water bodies [4]. Nickel is highly carcinogenic. Although it plays a vital role in production of red blood cells but it becomes toxic when its concentration exceeds the permissible limits. Previously, water contaminated by toxic

heavy metals has been purified by coagulation [5], ion exchange [6], reverse osmosis [7] and adsorption [8]. Adsorption is superior to all other methods.

Morgan in 1920 introduced the term chelate as the groups that coordinate to central metal atom in such a way that a heterocyclic ring is formed [9]. Ethylenediamine (EDA) is a bidentate ligand having two nitrogen donor atoms so it has two donor sites for the attachment of metal ion [10]. Dimethylglyoxime (DMG) is a bidentate ligand. It has been used as selective reagent for the analysis of nickel. Composition of the complexes has been reported as $M(DMG)_2$ [11]. DMG is a bidentate ligand used for the gravimetric estimation of nickel (II) ions. 1, 10-phenanthroline (1,10-Phen) is a bidentate heterocyclic ligand with two nitrogen atoms as donor sites. 1,10-phenanthroline form complex with Co (II) [12]. Nickel (II) forms different types of complexes with 1,10-phenanthroline, Tris phenanthroline nickel complex shows light red color while bis phenanthroline nickel shows light blue color [13]. Dithizone (diphenylthiocarbazone) abbreviated as (DZ) has sulfur and nitrogen donor atoms to form chelates with metals. It is a valuable reagent as it forms colored products with metals and can be used in low concentrations. In 1882 dithizone was prepared by Emil Fischer [14]. Dithizone is an efficient metal chelating agent; it separates metals in trace amounts. Mohammad Saraji and coworkers prepared nickel (II) dithizone complex [15]. Sulfanilamide (SNM) ligand has sulfur, oxygen and nitrogen donor atoms to form stable chelates with metals. It is known because of its remarkable bioactivities. It was the first drug used against bacterial infections.

Organically functionalized sol-gels because of their tunable porosity and surface layer composition have been the area of interest for researchers. Silica is usually selected as inorganic support due to its high surface area and pore size ranges from micro to macroporous. Silica in amorphous state is biogenic. It was the first material used for bioencapsulation. It shows high thermal and chemical stability. The organic part is an active reagent that is incorporated in silica framework for the removal of metal ions [16]. Alkoxysilanes are usually used to form sol-gel silica because they are stable and easy to handle [17].

Previously metal chelating agents such as 4-(2-thiazolylazo) resorcinol, 1-(2-Thiazolylazo)-2-naphthol, Diethyl ammonium diethyl dithiocarbamate, 5-methyl-4-(thiazol-2-yl diazenyl) benzene-1, 3-diol, trithiane, dithizone, dithiocarbamate, Schiff base and dimethylglyoxime have been used for the preparation of modified silica for the sorption of metal ions [18–25].

In this work we report functionalization of sol-gel with efficient chelating agents as well as their metal complexes by simple doping method at room temperature. High heating costs can be avoided on large scale production and application of these materials. They show good mechanical stability and fast equilibration in metal sorption. Efficient chelating agents and their metal complexes have been selected due to their analytical, industrial, biological, catalytic and other applications [29–33]. The synthesized materials can be utilized in biomedical and catalysis.

Experimental

2.1 Apparatus and Instruments

IR Spectra of the materials were recorded on ATR spectrophotometer. Functionalized Gels were analyzed by EDS Oxford 7573 combined with SEM, Atomic absorption spectrophotometry (AAS) was used to determine the amount of nickel (II) in solutions.

2.2 Chemicals and reagents:

In this work analytical grade reagents and absolute solvents purchased from Sigma Aldrich and Merck were used. Deionized water was used in all adsorption experiments.

2.3 Synthesis of Metal Complexes

(i) Ni Complex of Ethylenediamine

0.6 g $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ was dissolved in 2.5 mL of distilled water, added 4 mL of ethylenediamine. Stirred for 10 minutes. Added 15 mL of acetone slowly. Green color of solution changed to violet color. The mixture was cooled in ice bath, filtered and washed with acetone. Dried and weighed.

M.F: $[(\text{C}_2\text{H}_8\text{N}_2)_3\text{Ni}] \text{Cl}_2$, M.P: 233–238 °C, Color: Violet, Yield: 81 %. IR: N-H $3320 - 3164 \text{ cm}^{-1}$, CH_2 asymmetric stretch 2947 cm^{-1} , CH_2 symmetric stretch 2886 cm^{-1} , NH_2 scissoring 1590 cm^{-1} , C-N 1152 cm^{-1} , Calculated; $[(\text{C}_2\text{H}_8\text{N}_2)_3\text{Ni}] \text{Cl}_2$ (309.69 g/mol) (weight %) C: 23.24; H: 7.74; N: 27.12; Cl: 22.90; Ni: 19, Found; C: 23.58; H: 7.75; N: 27.17; Cl: 22.89; Ni: 18.61

(ii) Ni 1,10-Phenanthroline Complex

1mmol (0.237g) of Ni (II) chloride was dissolved in small amount of methanol and 3 mmol (0.594 g) of 1,10-phenanthroline was dissolved in a small amount of methanol. In hot methanolic solution of 1,10-phenanthroline added nickel chloride solution drop by drop while stirring and heated over water bath to concentrate the solution. The product was dried and weighed.

M.F: $[(\text{C}_{12}\text{H}_{10}\text{N}_2\text{O}_3)_3\text{Ni}] \text{Cl}_2$, M.P: 310 °C, Color: Pink, Yield: 61 %. IR: C = N 1624 cm^{-1} , C = C 1515 cm^{-1} , Calculated; $[(\text{C}_{12}\text{H}_{10}\text{N}_2\text{O}_3)_3\text{Ni}] \text{Cl}_2$ (820.2 g/mol) (weight %) C: 53.83; H: 3.76; N: 11.45; Cl: 23.21; Ni : 7.7, Found; C: 54.52; H: 3.91; N: 11.91; Cl: 21.91; Ni: 7.75

(iii) Ni Dithizone Complex

1mmol (0.256 g) of dithizone was dissolved in 50 mL of ammonia at room temperature. 0.5 mmol (0.131 g) of $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was dissolved in 12.5 mL of distilled water, added drop by drop to dithizone solution by vigorous stirring. Precipitates of the metal complex were filtered, dried and weighed.

M.F: $[(\text{C}_{13}\text{H}_{12}\text{N}_4\text{S})_2 \text{Ni}] (\text{NO}_3)_2$, M.P: 215 °C, Color: Brownish black, Yield: 51 %. IR (N = N 1594 cm^{-1} , C = S 1214 cm^{-1} , N-H scissor 1495 cm^{-1} , Calculated; $[(\text{C}_{13}\text{H}_{12}\text{N}_4\text{S})_2 \text{Ni}] (\text{NO}_3)_2$ (695.3g/mol) (weight %) C: 44.6; H: 3.4; N: 18.8; S: 10.1; O: 14.7; Ni : 8.4, Found; C: 44.21; H: 3.0; N: 19.04; S: 10.55; O: 14.8; Ni: 8.4

(iv) NiDMG Complex

1g of $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ was dissolved in distilled water and small amount of ammonium hydroxide was added to make the solution basic, color of solution changed from green to blue. 0.1 g of dimethylglyoxime was added in 10 mL of absolute ethanol (1 % DMG solution). Both solutions were mixed. Rosy red precipitates were filtered, dried and weighed.

M.F: $(\text{C}_4\text{H}_7\text{N}_2\text{O}_2)_2\text{Ni}$, M.P: 293 °C, Color: Rosy red, Yield: 64 %. IR C = N 1570 cm^{-1} , Calculated; $(\text{C}_4\text{H}_7\text{N}_2\text{O}_2)_2\text{Ni}$ (288.69 g/mol) (weight %); C: 33.26; H: 4.85; N: 19.40; O: 22.17, Ni: 20.32, Found; C: 33.51; H: 4.96; N: 19; O: 21.90; Ni: 20.63

(v) Tetrakis sulfanilamide nickel (II) chloride

2.37 g of $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$ and 6.80 g of p-amino benzene sulfonamide were dissolved in methanol separately. Both solutions were mixed at 65–70 °C and mixture was stirred for 2 hours in presence of pH 10 buffer. Change in color from dark green to light green and precipitation at room temperature showed the complex formation. Precipitates were filtered and washed with methanol. Recrystallization was done with ethanol.

M.F: $[\text{Ni}(\text{C}_6\text{H}_8\text{N}_2\text{O}_2\text{S})_2]\text{Cl}_2$, M.P: 270 °C, Color: light green; Yield: 65 %, IR; $3500 - 3300\text{ cm}^{-1}$ (-NH stretch for primary amines), $1640 - 1560\text{ cm}^{-1}$ (-NH bend for primary amines), 1600 and 1475 cm^{-1} (C = C aromatic stretch), 3000 cm^{-1} (= CH stretch), 1335 and 1150 cm^{-1} (S = O stretch), $1350 - 1000\text{ cm}^{-1}$ (C-N stretch), 400 cm^{-1} (Ni-NH₂ stretch) **Calculated**, $[\text{Ni}(\text{C}_6\text{H}_8\text{N}_2\text{O}_2\text{S})_2]\text{Cl}_2$ (474.01 g/mol) (weight %); C: 30.3; H: 3.40; N: 11.8 ; O: 13.5, Ni:19.35; Cl : 14.9 ; S: 6.75, **Found**, C: 30.1; H: 3.50; N: 11.7 ; O: 13.6, Ni:19.30; Cl : 15.1 ; S: 6.70

2.4 Synthesis of Functionalized Xerogels

(i) Synthesis of Sol-Gels Functionalized with Chelating Agents

5 mL tetraethoxysilane and 5 mL distilled water were added to 10 mL ethanol solution of 0.005 M chelating agent containing appropriate amount of ammonium fluoride as catalyst in a 100 mL beaker. A transparent gel was formed. It was dried at room temperature for two days and then placed in oven at 45 °C till constant weight. Then the gel was crushed, ground and sieved. Undoped chelating agent was removed by soaking the gel in distilled water. After removing water, the gel was dried again. Blank xerogel was also prepared by the same procedure without adding the chelating agent.

(ii) Synthesis of Sol-Gels Functionalized with Metal Complexes

1 mL tetraethoxysilane and 1mL distilled water were added to 5 mL ethanol solution of 0.002 M complex containing 0.002 M ammonium fluoride as catalyst in a 100mL beaker. Gel was formed in few seconds. It

was dried at room temperature for two days to evaporate all the solvents, then placed in oven at 45 °C till constant weight. The dried gel was crushed, ground and sieved. Undoped complex was removed by soaking the gel in distilled water. After removing water, the gel was dried in oven.

2.5 Sorption of Ni (II) ions

Sorption experiments were performed by the batch method at room temperature by letting undoped/doped sol-gel (50 mg) in contact with 20 mL of Ni (II) solutions of neutral pH in labelled test bottles. To study sorption isotherms, Ni (II) solutions of 15–50 mg/L were prepared and 20 mL of each solution was taken in the test bottles having pre-weighed sorbents. The suspensions were shaken, centrifuged and analyzed for final metal concentrations on atomic absorption spectrophotometer. The amount of metal sorbed (q_e in mg/g) was determined using the following equation:

$$q_e = C - C_e \times \frac{V}{m}$$

Where C and C_e are metal concentrations (mg/L) before and after sorption respectively, m is the amount of sorbent in grams and V is the suspension volume in liters.

The above experiments were repeated using blank gel as well.

Results And Discussion

3.1 Physical Properties

Chelating agent's metal complexes are brightly coloured. Their colours and melting points are different from respective chelating agents. Blank gel is white in color. Doped gels of chelating agents and their metal complexes are coloured having the same colour as that of chelating agent/metal complex showing no chemical change in the process of doping.

3.2 IR Studies

IR spectra of blank and doped gels presented in Fig. 2 showed characteristic absorptions of silanol groups (Si-OH), siloxane linkages (Si-O-Si), and chelating agent's functional groups. Absorption at 3304 cm^{-1} and 1626 cm^{-1} is due to silanol groups (Si-OH) whereas absorption at 1066 cm^{-1} is due to siloxane linkages (Si-O-Si). Ethylenediamine nickel complex shows $\text{-CH}_2\text{-}$ asymmetric stretch at 2947 cm^{-1} and symmetric stretch at 2886 cm^{-1} . Absorption in the range $3320 - 3164\text{ cm}^{-1}$ is assigned to -NH_2 and absorption at 1152 cm^{-1} to C-N groups. 1,10 Phenanthroline shows C = C and C = N absorptions at 1503 cm^{-1} and 1616 cm^{-1} respectively. On complexation these absorptions shift at 1515 cm^{-1} and 1624 cm^{-1} respectively. This shifting is an indication of nitrogen coordination to metal [26]. Dithizone does not show any signal for N-H stretching due to internal hydrogen bonding whereas on complexation it shows absorption at 3250 cm^{-1} . Absorption band of N = N is observed at 1590 cm^{-1} . In metal complex this peak is shifted to 1594 cm^{-1} . N-H scissoring is observed at 1496 cm^{-1} in dithizone and shifts to 1495 cm^{-1} in

the metal complex [27]. C = S absorption is observed at 1212 cm^{-1} in dithizone. On complexation this band shifts to 1214 cm^{-1} . In DMG a band observed at 3196 cm^{-1} for OH groups is absent in its complex showing hydrogen bonding in the complex. C = N absorption in the range of $1625 - 1500\text{ cm}^{-1}$ is absent in ligand due to intramolecular hydrogen bonding and it has been observed in metal complex at 1570 cm^{-1} [28].

3.3 Scanning Electron Microscopy (SEM)

SEM of blank and doped gels showed porous structure. Gels functionalized with chelating agent showed partly stick together particles of roughly spheric form. Gels functionalized with metal complex showed tubular structure. Functionalized gels showed shrinkage of pores as compared to blank gel. After sorption of metal ions complete shrinkage of pores of xerogels is observed. SEM images of blank and functionalized gels are presented in Fig. 3. Sol-gel silica was also functionalized with metal complexes for comparison with chelating agent functionalized gels with sorbed Ni (II) ions. Same amount of Ni (II) was observed in both, showing high efficiency of the process.

3.4 Energy Dispersive X-Ray Spectroscopy (EDS)

EDS of blank gel, sol-gel functionalized with Ni-DMG complex and DMG functionalized sol-gel with adsorbed Ni are presented in Fig. 4. EDS of sol-gel functionalized with Ni-DMG complex and DMG functionalized sol-gel with adsorbed Nickel are taken for comparison. Both samples show the same amount of nickel.

3.5 XRD Studies

XRD spectra of blank and modified gels showed pure state of amorphous structure. No disorder or crystallinity was observed due to undoped chelating agent as evidenced in Fig. 5. The main diffraction peak after 2θ ; 20° is observed for amorphous silica. Silica in amorphous state is biogenic. It has been used in bioencapsulation. Crystalline silica is highly toxic. The synthesized amorphous silica sol-gel can be used in biomedical applications as well.

3.6 Sorption Studies

Sorption of Ni (II) by blank (BG) and functionalized xerogels has been provided in Fig. 6. Blank gel shows negligible sorption. Functionalized gels show high sorption capacity. Sorption data shows successful incorporation of chelating agents. DMGSG shows the highest sorption capacity for Ni (II) ions. 10 mg Ni (II) was adsorbed by functionalized silica having 20 mg DMG, showing Ni/DMG ratio of 1:2.

3.7 Desorption of Ni (II) from functionalized sol-gels

A fast quantitative desorption of metal ions is essential for a high efficiency of adsorbent in metal removal mode. It was found that 0.1 molar HNO_3 was fairly effective for complete recovery of Ni(II) ions from the gels. The sol-gels loaded with known amounts of metal ions were washed with deionized water. After washing and drying they were shaken with 20 mL of 0.1 molar HNO_3 solution for a minute. The concentration of released metal was determined using atomic absorption spectrophotometer. After the

complete recovery of the gels, they were washed with enough deionized water for repeated use. 100 % elution of metal ions were achieved for the functionalized gels under study. They exhibited chemical stability and efficiency for several sorption-desorption-regeneration cycles for each batch of sorbent.

Conclusions

The chelating agents and their metal complexes did not show any chemical change after entrapment in the solid cages of the xerogels. The chelating agents retained their activity for the target metal ions. The entrapped chelating agents played major role in the sorption of metal ions by the functionalized gels. Thus, the simple preparation of doped gels, fast sorption of Ni (II), desorption and recyclability increase their commercial scope. All the sorption experiments were performed at room temperature and neutral pH. Thus, it is possible to use chelating agents entrapped into sol-gels for analytical and industrial applications. XRD showed amorphous nature of blank and functionalized gels. Amorphous silica is biogenic and can be utilized in biomedical.

Declarations

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Conflicts of interest/Competing interests

We do not have any conflicts of interest to declare

Availability of data and material

The data required to reproduce the above findings cannot be shared at this time due to technical/time limitations

Code availability (NA)

Authors' contributions

All authors have contributed significantly

Ethics approval

The accepted principles of ethical conduct have been followed

Consent to participate

Agreed

Consent for publication

Agreed for publication

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Figures

Figure 1

Structures of complexes (a) Ni Complex of Ethylenediamine (b) Tris (1, 10-Phenanthroline) Nickel (II) (c) Bis (Dithizone) Nickel (II) (d) Bis (Dimethylglyoxime) Nickel (II) (e) Tetrakis sulfanilamide nickel (II) chloride

Figure 2

IR spectrum of (a) blank gel, (b) DMG, (c) NiDMG complex, (d) DMG modified gel, (e) NiDMG complex modified gel

Figure 3

SEM image of (a) blank gel at high resolution, (b) blank gel at low resolution, (c) DMG modified gel at high resolution, (d) DMG modified gel at low resolution, (e) NiDMG complex modified gel at high resolution, (f) NiDMG complex modified gel at low resolution

Figure 4

EDS Scan of (a) Blank Gel (b) NiDMG Complex Modified Gel (c) DMG modified Gel with adsorbed Ni (II)

Figure 5

XRD Scan of Blank and DMG modified Gel

Figure 6

Sorption of Ni (II) mg/g (from 15 to 50 mg/L Ni (II) solution)