# SUPPLEMENTARY INFORMATION

## Versatile Heavy Metals Removal *via* Magnetic Mesoporous Nanocontainers

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## MATERIALS AND METHODS

Tetraethoxysilane, diethyenetriamine pentaaetic diahydride, cetyltrimethyammonium bromide, sodium hydroxide, aminopropyltriethoxysilane, hydrated iron oxide, oleic acid, ammonium nitrate, ether, dimethylformamide, copper(II) chloride, zinc(II) chloride, cadmium(II) bromide tetrahydate, and manganese(II) chloride tetrahydrate were purchased from Sigma-Aldrich. Anhydrous ethanol, and chloroform were purchased from Fisher Chemicals. Palladium(II) chloride, and tin(IV) chloride pentahydrate were purchased from Alfa-Aeser. Thallium(I) sulfate was purchased from Prolabo. Cobalt(II) nitrate pentahydrate, and lead(II) carbonate were purchased from Labosi. Mercury(II) acetate was purchased from Strem Chemicals. Nickel(II) chloride hexahydrate, and chromium(III) chloride were purchased from Chimica.

TEM images were recorded with a JEOL instrument. SEM images were recorded with a FEI instrument. Energy dispersive spectroscopy was performed *via* an FEI scanning electron microscope. The metal ions adsorption capacities were determined from the $K_a$ rays. The magnetic separation was performed *via* a 1 Tesla magnet. IR spectra were recorded on a Perkin-Elmer 100 FT spectrophotometer. Absorption spectra were recorded on a Hewlett-Packard 8453 spectrophotometer. Dynamic light scattering analyses were performed using a Cordouan Technologies DL 135 Particle size analyzer instrument. $^{29}Si$ and $^{13}C$ CPMAS solid state NMR sequences were recorded with a VARIAN VNMRS300, using Q8MH8 and adamantine references respectively.

## EXPERIMENTAL SECTION

**L ligand**. A mixture of diethyenetriamine pentaaetic diahydride (DTPA, 300 mg, $8.0*10^{-1}$ mmol), aminopropyltriethoxysilane (177 mg, $8.0*10^{-1}$ mmol), triethylamine (81 mg, $8.0*10^{-1}$ mmol), and dimethylsufoxide (DMF, 20 mL) was sonicated 20 minutes and stirred 3 h at room temperature. The ligand solution was then used without further purification.

**$Fe_3O_4$NCs**. A mixture of hydrated iron oxide (FeO(OH), 0.181 g), oleic acid (3.180 g), and docosane (5.016 g) was prepared in a 100 mL two neck round bottom flask, and stirred under vacuum for 30 minutes. Then, an argon atmosphere was applied and the mixture was refluxed at 340°C. As soon as the mixture reached 340°C, the reaction was conducted for 1 h 30 min. The solution was cooled down during 1 h without stirring. The resulting black solid was dissolved in pentane (14 mL), mixed with an ether/ethanol solution (30 mL, 2:1, v:v), and centrifugated 10 minutes at 21 krpm (27 krcf). The decomposed organic black solution was removed and the NCs were redispersed in pentane (2 mL), washed with the ether/ethanol solution, and centrifugated. The supernatant was removed, the NCs were stored in chloroform (15 mL), and stabilized with an addition of oleylamine (200 µL).

**$Fe_3O_4$@MSN**. A mixture of cetyltrimethylammonium bromide (250 mg, CTAB), distilled water (120 mL), and sodium hydroxide (875 µL, 2 M) was stirred at 70°C during 50 minutes

 

at 700 rpm in a 250 mL three neck round bottom flask. Then, $Fe_3O_4$NCs dispersed in chloroform were added stepwise (8*500 µL) during 20 minutes. Note that the chloroform is quickly evaporating from the mixture at this temperature, and 40 additional minutes were used at 80°C to stabilize the NCs in the micelles. Finally, tetraethoxysilane (1.2 mL, TEOS) was added to the aforementioned solution, and the condensation process was conducted for 2 h. Afterwards, the solution was cooled at room temperature while stirring; fractions were gathered in polypropylene tubes and collected by centrifugation during 15 minutes at 21 krpm (27 krcf). The sample was then extracted twice with an alcoholic solution of ammonium nitrate (6 $g.L^{-1}$), and washed three times with ethanol, water, and ethanol. Each extraction involved a sonication step of 30 minutes at 50°C in order to remove the CTAB surfactant; the collection was carried out in the same manner. The as-prepared material was dried under air flow few hours.

**$Fe_3O_4$@MSN-L**. $Fe_3O_4$@MSN NPs (100 mg) were sonicated in anhydrous ethanol (4 mL) for 10 minutes, and the ligand was added (4 mL from the DMF solution). The solution was heated to 70°C during 24 h, and centrifugated 10 minutes at 21 krpm (27 krcf). The supernatant was removed and the resulting $Fe_3O_4$@MSN-L NPs were washed once with ethanol, twice with water, once with acetone, and dried under vacuum.

**$Fe_3O_4$@$MSNH_2$**. A mixture of cetyltrimethylammonium bromide (250 mg, CTAB), distilled water (120 mL), and sodium hydroxide (875 µL, 2 M) was stirred at 70°C during 50 minutes at 700 rpm in a 250 mL three neck round bottom flask. Then, $Fe_3O_4$NCs dispersed in chloroform were added stepwise (8*500 µL) during 20 minutes. Note that the chloroform is quickly evaporating from the mixture at this temperature, and 40 additional minutes were used at 80°C to stabilize the NCs in the micelles. Finally, TEOS (1 mL) and aminopropyltriethoxysilane (200 µL) were added to the aforementioned solution, and the condensation process was conducted for 2 h. Afterwards, the solution was cooled at room temperature while stirring; fractions were gathered in polypropylene tubes and collected by centrifugation during 15 minutes at 21 krpm (27 krcf). The sample was then extracted twice with an alcoholic solution of ammonium nitrate (6 $g.L^{-1}$), and washed three times with ethanol, water, and ethanol. Each extraction involved a sonication step of 30 minutes at 50°C in order to remove the CTAB surfactant; the collection was carried out in the same manner. The as-prepared material was dried under air flow few hours.

**$Fe_3O_4$@MSL**. $Fe_3O_4$@$MSNH_2$ NPs (50 mg) were mixed with DMF (500 µL), DTPA (5 mg), and triethyeneamine (100 µL), an d stirred at room temperature for 5 h. The supernatant was removed and the resulting $Fe_3O_4$@MSL NPs were washed once with ethanol, twice with water, once with acetone, and dried under vacuum.

**$Fe_3O_4$@MSL-L**. $Fe_3O_4$@MSL NPs (100 mg) were sonicated in anhydrous ethanol (4 mL) for 10 minutes, and the ligand was added (4 mL from the DMF solution). The solution was heated to 70°C during 24 h, and centrifugated 10 minutes at 21 krpm (27 krcf). The supernatant was removed and the resulting $Fe_3O_4$@MSL-L NPs were washed once with ethanol, twice with water, once with acetone, and dried under vacuum.

**$Fe_3O_4$@MSN-L Metal adsorption**. $Fe_3O_4$@MSN-L NPs (4 mg) were stirred with the metallic complex ($M^{n+}$, 14 mg), in deionized water (1 mL) for 24 h at room temperature. The NPs were separated from the solution *via* a magnet in 10 minutes, and the solution was removed. Then, the NPs were washed thrice with water, twice in ethanol, once in acetone, and dried under vacuum.

**$Fe_3O_4$@MSL Metal adsorption**. $Fe_3O_4$@MSL NPs (4 mg) were stirred with the metallic complex ($M^{n+}$, 14 mg), in deionized water (1 mL) for 24 h at room temperature. The NPs were separated from the solution *via* a magnet in 10 minutes, and the solution was removed. Then, the NPs were washed thrice with water, twice in ethanol, once in acetone, and dried under vacuum.

**$Fe_3O_4$@MSL-L Metal adsorption**. $Fe_3O_4$@MSL-L NPs (4 mg) were stirred with the metallic complex ($M^{n+}$, 14 mg), in deionized water (2 mL) for 24 h at room temperature. The NPs were separated from the solution *via* a magnet in 10 minutes, and the solution was removed. Then, the NPs were washed thrice with water, twice in ethanol, once in acetone, and dried under vacuum.

**Selectivity study of the metal adsorption**. $Fe_3O_4$@MSL NPs (26 mg) were stirred with all the metallic complexes ($M^{n+}$, 20 mg each), in deionized water (20 mL) for 2 h at room temperature. The NPs were separated from the solution *via* a magnet in 10 minutes, and the solution was removed. Then, the NPs were washed thrice with water, twice in ethanol, once in acetone, and dried under vacuum.

**Nanocontainers desorption** The acidic desorption of the metal ions was performed at pH 4.5, at which the DTPA ligand is protonated (pKa values of about 5.6 and 6.1 according to *Martell and Smith,1989*). Thirty minutes of sonication is used for an efficient desorption at 25°C. In these conditions, the DTPA-$Mn^{n+}$ complexes are not stable anymore, and the ions could be collected in the aqueous supernatant. The NPs were then washed three times in water, and the supernatant fractions were gathered for inductively-coupled plasma (ICP) measurement. According to our preliminary results with ICP measurements on the aqueous removal solution, 90 to 99 of the copper and zinc ions could be removed from the nanocontainers after one desorption process.

 

NANOMATERIAL CHARACTERIZATIONS

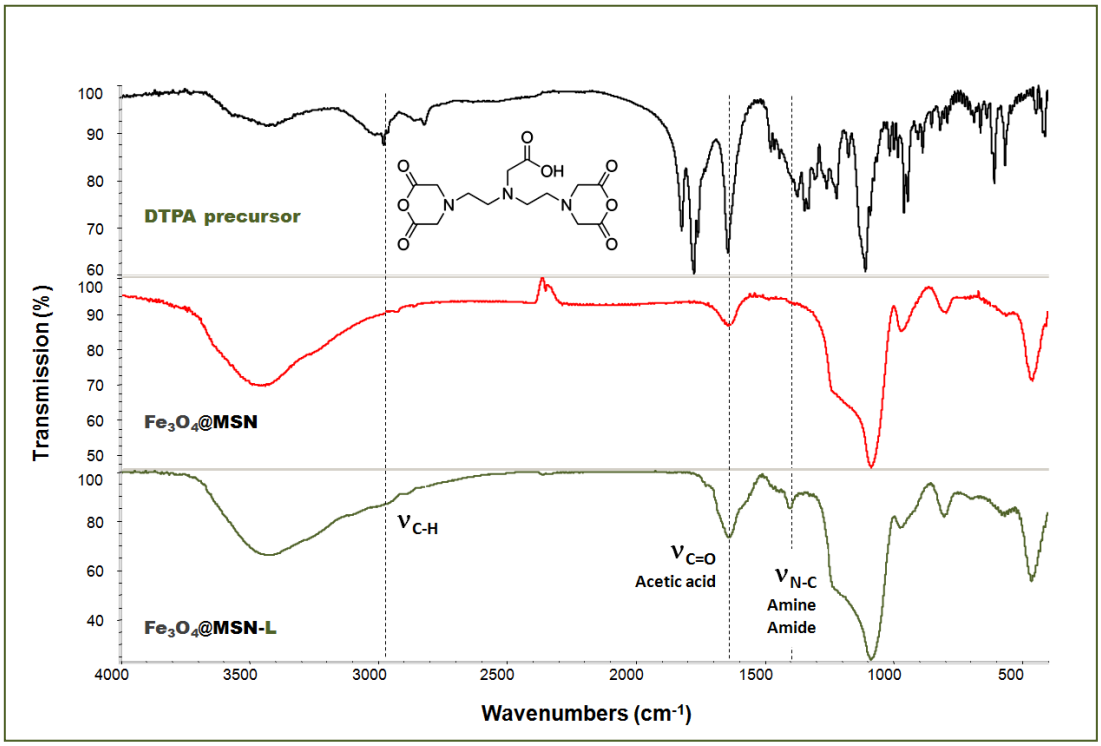


**Figure S1** IR spectra of the ligand precursor, the $Fe_3O_4$@MSN NPs, and $Fe_3O_4$@MSN-L NPs, demonstrating the ligand functionalization.

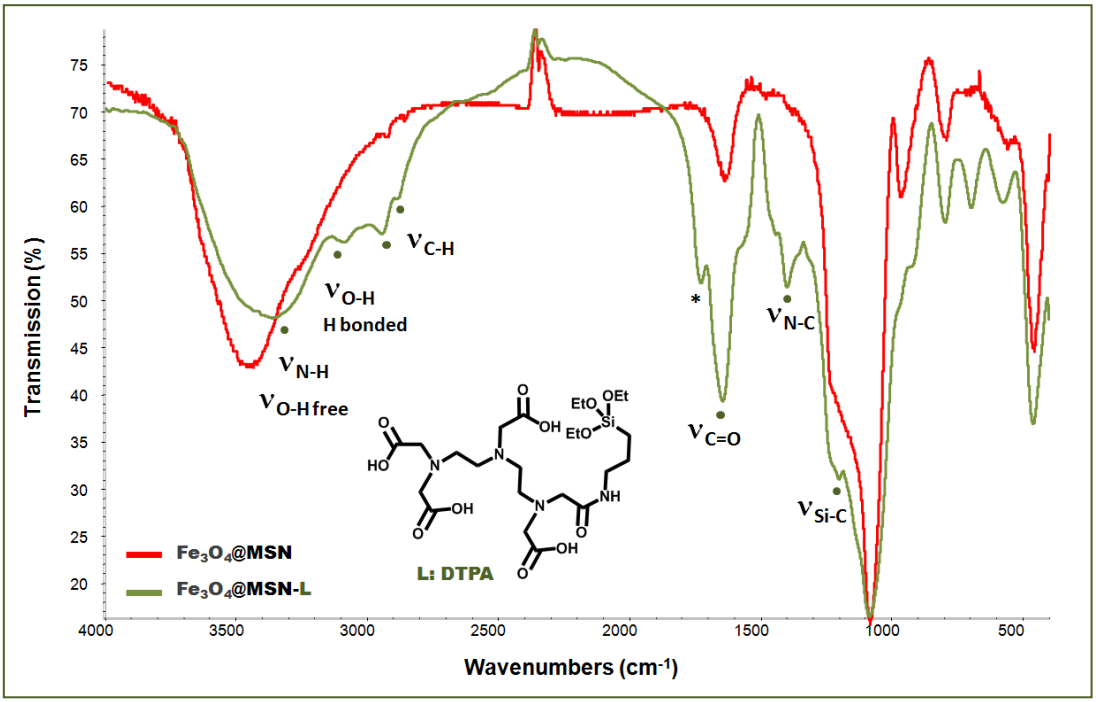


**Figure S2.** IR spectra of the surfactant-free $Fe_3O_4$@MSN, and $Fe_3O_4$@MSN-L NPs with a higher ligand functionalization, further demonstrating the DTPA ligand functionalization.

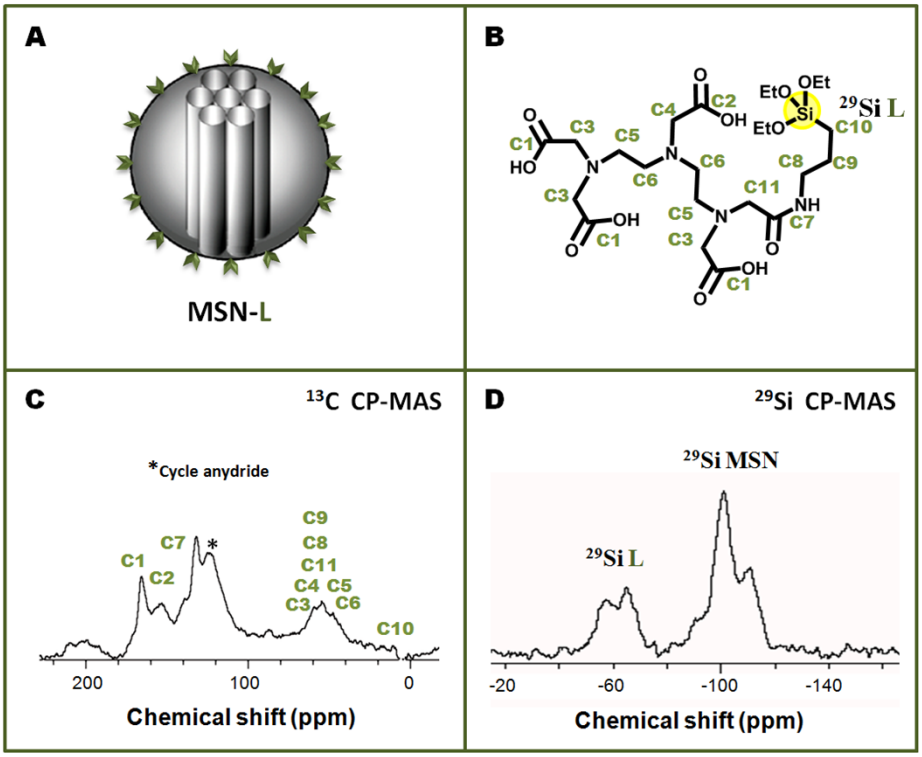


**Figure S3.** Solid state NMR $^{13}C$ and $^{29}Si$ CPMAS spectra (C-D) of a MSN-L NPs control (A), further confirming the alkoxysilylation of the ligand (B) on the MSN surface.

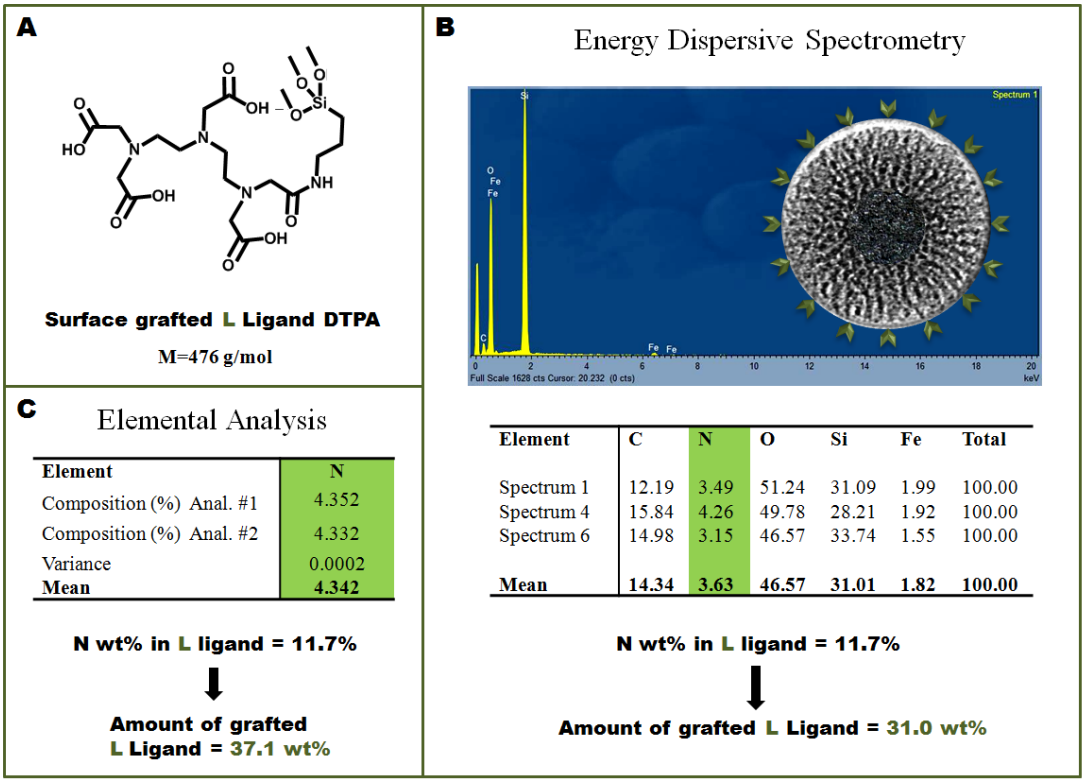


**Figure S4.** Molecule model used to calculate the ligand grafting on the NPs (A). EDS composition measurement of $Fe_3O_4$@MSN-L (B), and the combustion elemental measurement of nitrogen (C).

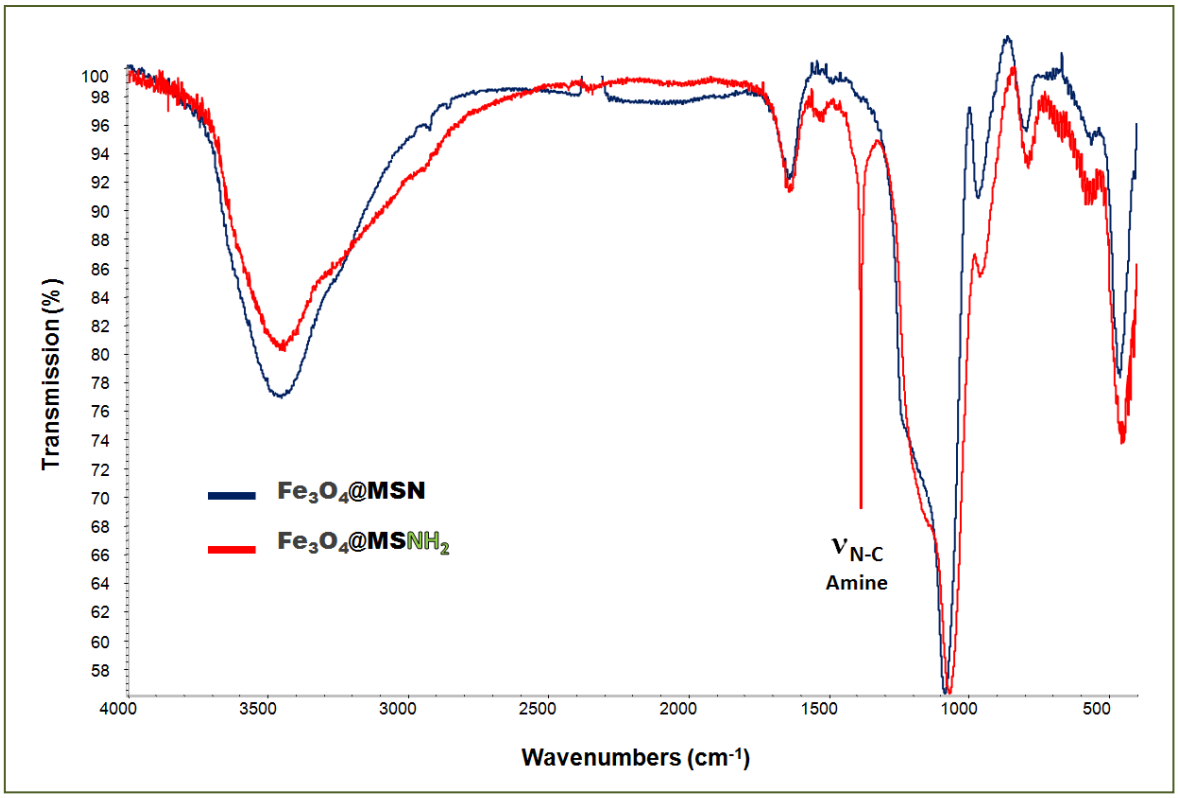


**Figure S5.** IR spectra of the $Fe_3O_4$@MSN NPs, and $Fe_3O_4$@$MSNH_2$ NPs demonstrating the successful encapsulation of amino groups in the MSN framework.

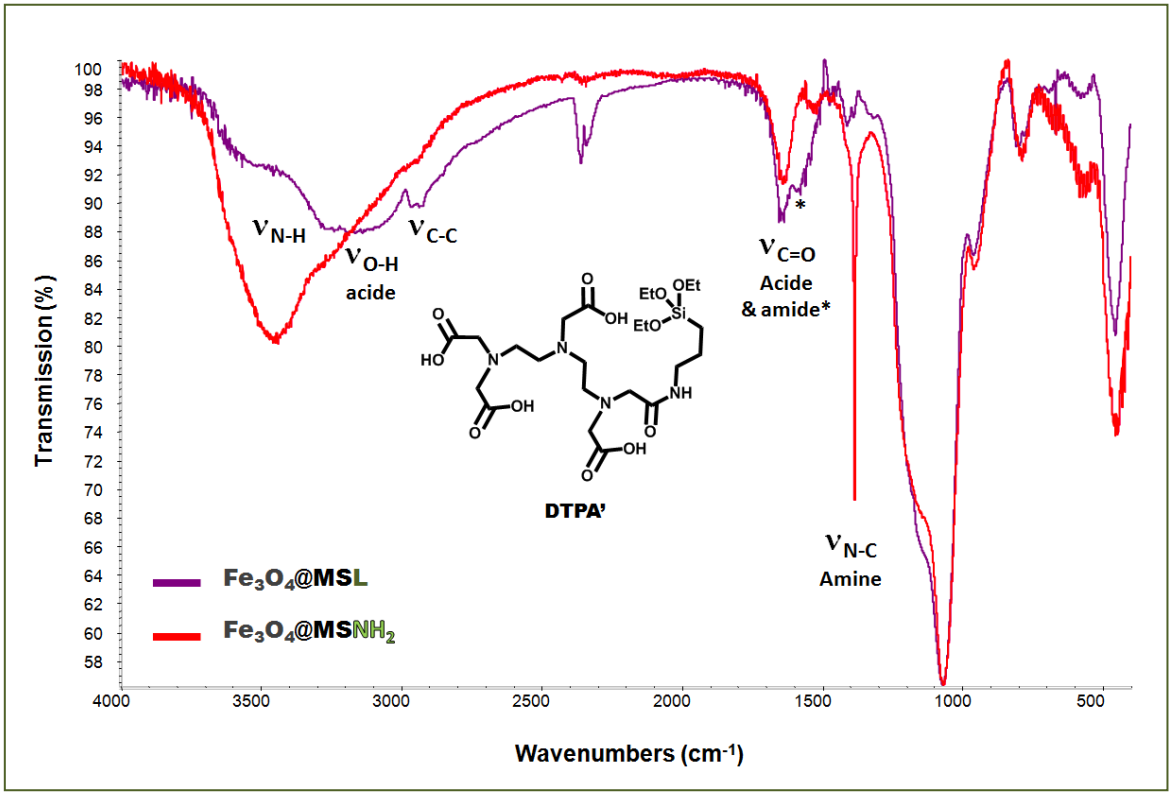


**Figure S6.** IR spectra of the $Fe_3O_4$@$MSNH_2$ NPs, and $Fe_3O_4$@MSL NPs demonstrating the successful grafting of the ligand in the MSN framework.

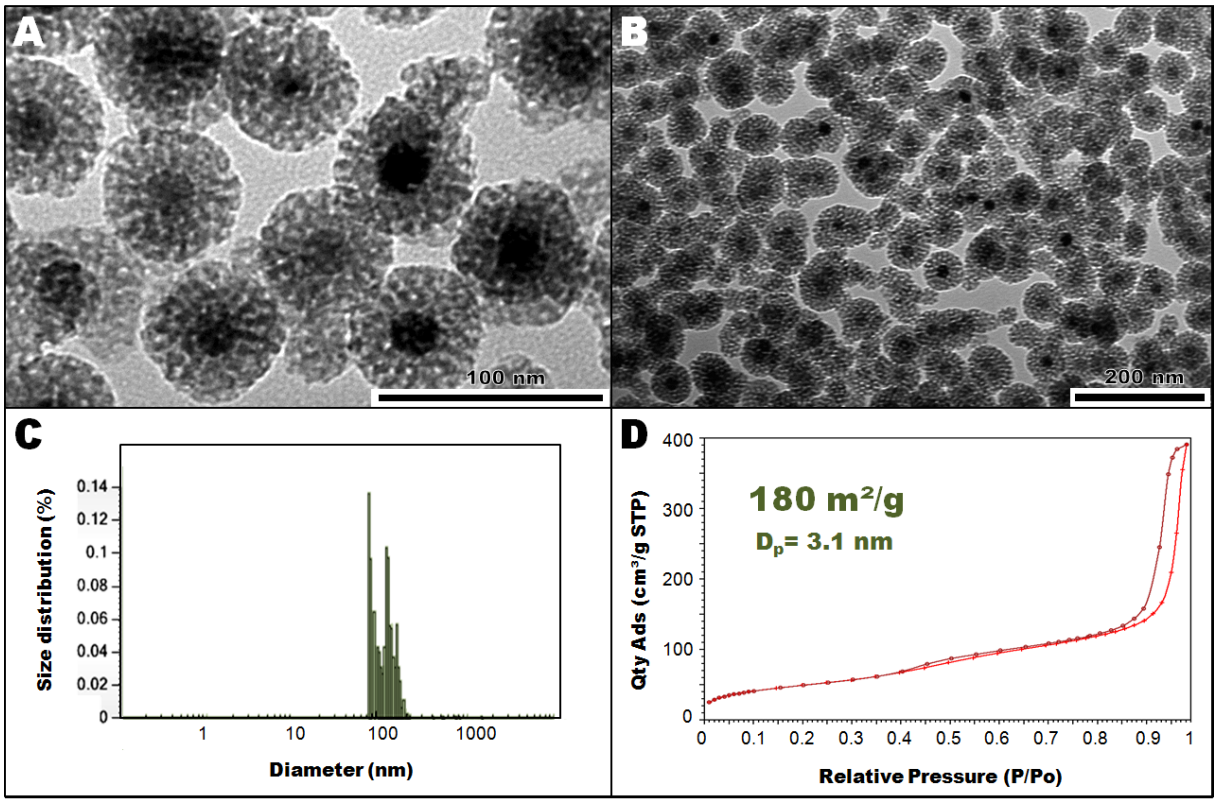


**Figure S7.** Transmission electron microscopy images (A-B), DLS size distribution (C), and $N_2$-adsorption-desorption analysis of $Fe_3O_4$@$MSNH_2$ NPs (D).

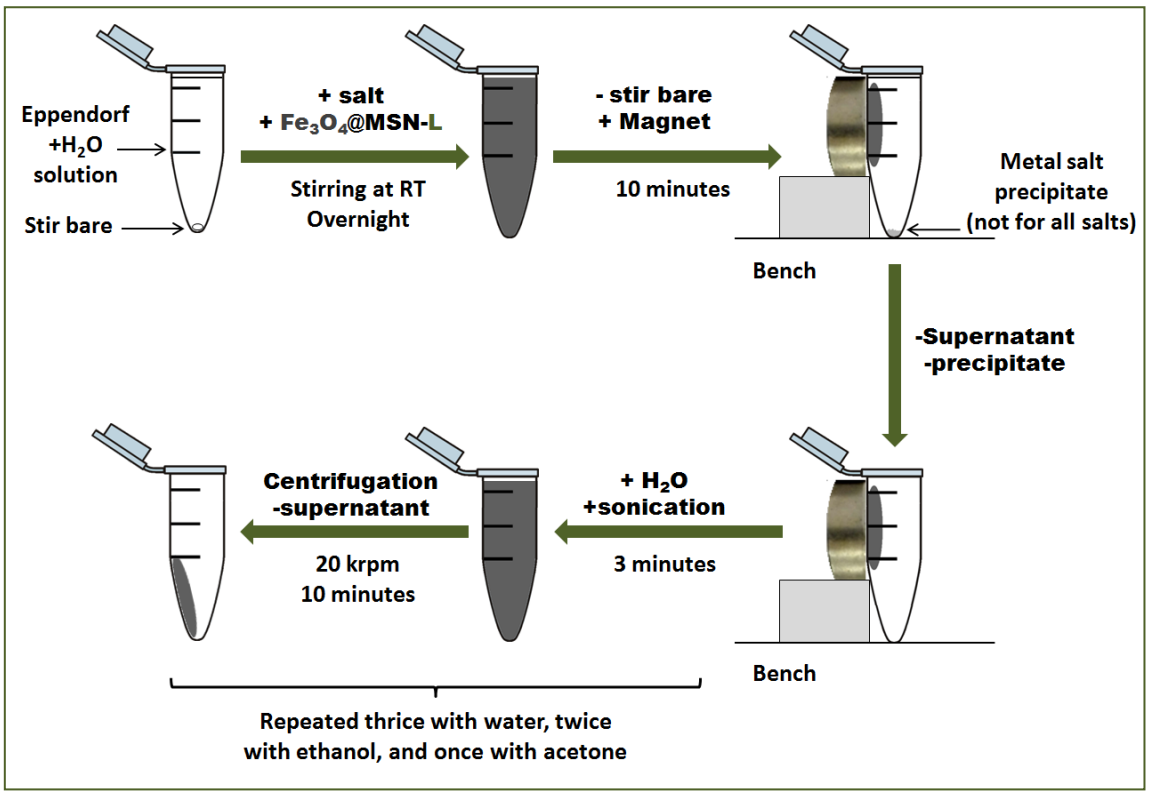


**Figure S8.** Schematic representation of the experimental settings of the heavy metal removal.

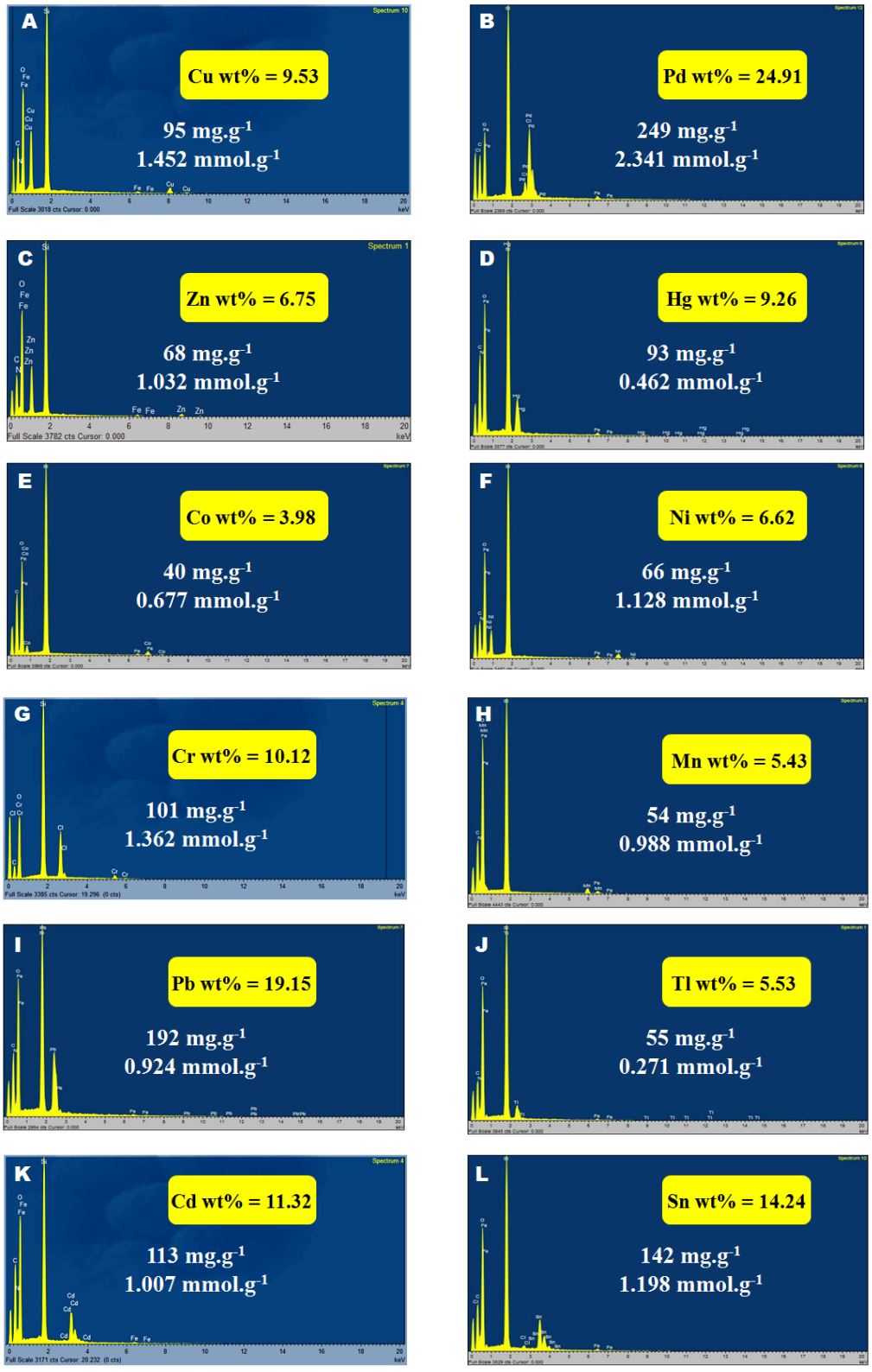


**Figure S9.** EDS spectra of $Fe_3O_4$@MSN-L∩$M^{n+}$ and adsorption capacities (A-L).

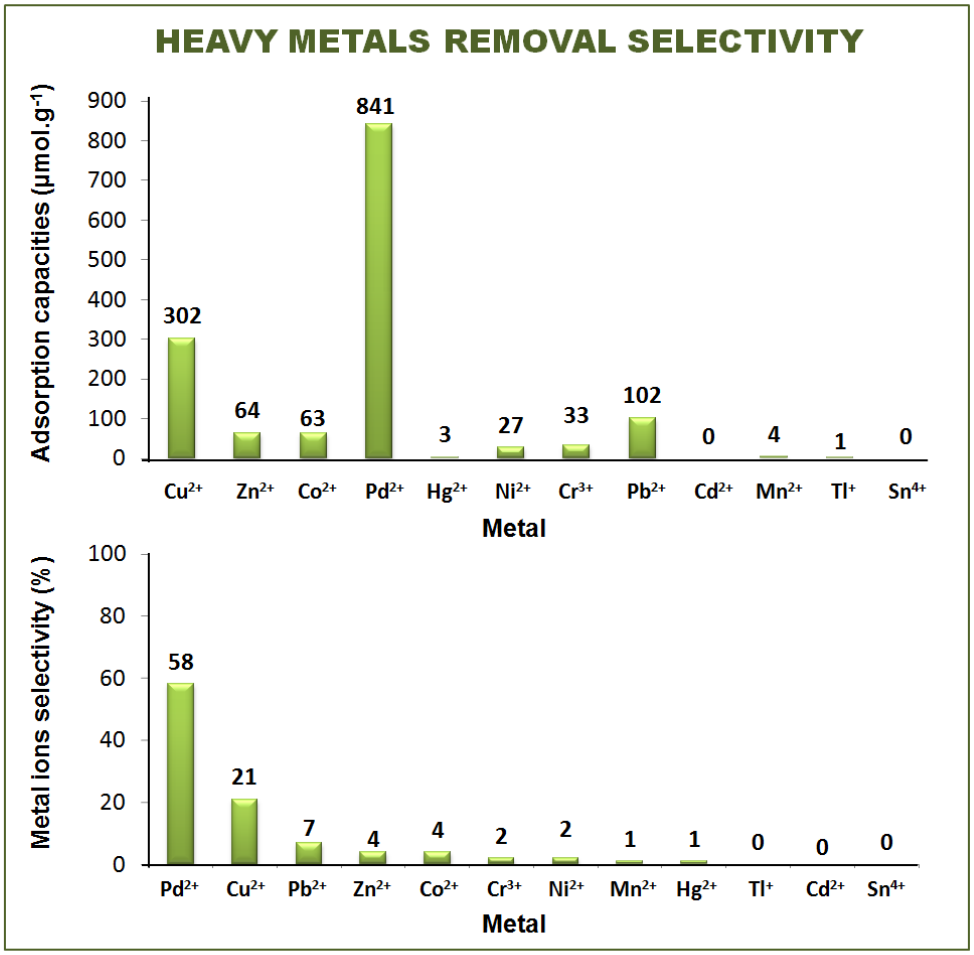


**Figure S10.** Selectivity of the metal ion removals by the nanocontainers. EDS was used to determine the adsorption capacities.

**Table S1-11.** EDS statistics of $Fe_3O_4$@MSN-L∩$M^{n+}$compositions for all metals expect the magnetic chromium.

| FM-L∩$Cu^{2+}$ | C | N | O | Si | Fe | Cu | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 23.18 | 6.41 | 31.04 | 27.38 | 2.06 | 9.92 | 100.00 |
| Spectrum 2 | 25.44 | 7.88 | 41.06 | 17.64 | 1.82 | 6.14 | 100.00 |
| Spectrum 3 | 22.12 | 6.05 | 33.42 | 26.13 | 2.30 | 9.98 | 100.00 |
| Spectrum 4 | 28.66 | 6.86 | 34.72 | 20.08 | 1.51 | 8.17 | 100.00 |
| Spectrum 5 | 26.40 | 9.02 | 40.20 | 16.67 | 1.06 | 6.64 | 100.00 |
| Spectrum 6 | 27.92 | 8.56 | 31.62 | 17.72 | 0.53 | 13.64 | 100.00 |
| Spectrum 7 | 24.88 | 6.48 | 32.11 | 22.28 | 1.32 | 12.93 | 100.00 |
| Spectrum 8 | 25.30 | 5.34 | 34.51 | 19.83 | 1.50 | 13.52 | 100.00 |
| Spectrum 9 | 31.55 | 5.80 | 33.28 | 19.34 | 1.77 | 8.26 | 100.00 |
| Spectrum 10 | 24.91 | 7.68 | 40.68 | 17.70 | 2.88 | 6.14 | 100.00 |
| **Mean** | **26.04** | **7.01** | **35.26** | **20.48** | **1.68** | **9.53** | **100.00** |
| Std. deviation | 2.75 | 1.23 | 3.90 | 3.69 | 0.66 | 2.97 | |
| Max. | 31.55 | 9.02 | 41.06 | 27.38 | 2.88 | 13.64 | |
| Min. | 22.12 | 5.34 | 31.04 | 16.67 | 0.53 | 6.14 | |

| FM-L∩$Zn^{2+}$ | C | N | O | Si | Fe | Zn | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 26.45 | 8.65 | 38.95 | 18.23 | 1.18 | 6.55 | 100.00 |
| Spectrum 2 | 33.59 | 10.89 | 37.67 | 9.46 | 0.17 | 8.22 | 100.00 |
| Spectrum 3 | 33.03 | 7.09 | 37.93 | 14.82 | 1.68 | 5.45 | 100.00 |
| Spectrum 4 | 24.47 | 6.35 | 35.98 | 23.74 | 1.31 | 8.16 | 100.00 |
| Spectrum 5 | 25.10 | 8.53 | 43.65 | 16.02 | 1.27 | 5.43 | 100.00 |
| Spectrum 6 | 29.02 | 8.45 | 40.60 | 14.87 | 1.94 | 5.11 | 100.00 |
| Spectrum 7 | 25.26 | 7.82 | 36.15 | 20.71 | 1.31 | 8.75 | 100.00 |
| Spectrum 8 | 27.85 | 8.57 | 41.44 | 15.68 | 1.20 | 5.27 | 100.00 |
| Spectrum 9 | 24.61 | 7.53 | 39.88 | 21.04 | 1.07 | 5.88 | 100.00 |
| Spectrum 10 | 22.03 | 5.65 | 33.44 | 28.43 | 1.77 | 8.67 | 100.00 |
| **Mean** | **27.14** | **7.95** | **38.57** | **18.30** | **1.29** | **6.75** | **100.00** |
| Std. deviation | 3.77 | 1.45 | 2.98 | 5.37 | 0.49 | 1.52 | |
| Max. | 33.59 | 10.89 | 43.65 | 28.43 | 1.94 | 8.75 | |
| Min. | 22.03 | 5.65 | 33.44 | 9.46 | 0.17 | 5.11 | |

| FM-L∩$Co^{2+}$ | C | N | O | Si | Fe | Co | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 28.59 | 13.21 | 40.85 | 11.92 | 0.62 | 4.82 | 100.00 |
| Spectrum 2 | 28.92 | 10.63 | 42.21 | 12.97 | 0.70 | 4.57 | 100.00 |
| Spectrum 3 | 25.12 | 5.98 | 34.27 | 26.56 | 2.32 | 5.76 | 100.00 |
| Spectrum 4 | 33.34 | 7.68 | 39.42 | 15.20 | 1.05 | 3.31 | 100.00 |
| Spectrum 5 | 29.62 | 7.80 | 42.92 | 15.90 | 0.98 | 2.79 | 100.00 |
| Spectrum 6 | 26.06 | 7.55 | 45.47 | 17.13 | 1.16 | 2.64 | 100.00 |
| Spectrum 7 | 36.01 | 4.54 | 34.99 | 19.32 | 1.04 | 4.11 | 100.00 |
| Spectrum 8 | 28.14 | 6.83 | 44.71 | 16.85 | 0.60 | 2.86 | 100.00 |
| Spectrum 9 | 35.62 | 4.90 | 27.57 | 22.70 | 3.27 | 5.93 | 100.00 |
| Spectrum 10 | 32.58 | 9.54 | 41.07 | 12.32 | 1.44 | 3.05 | 100.00 |
| **Mean** | **30.40** | **7.86** | **39.35** | **17.09** | **1.32** | **3.98** | **100.00** |
| Std. deviation | 3.80 | 2.66 | 5.54 | 4.68 | 0.85 | 1.24 | |
| Max. | 36.01 | 13.21 | 45.47 | 26.56 | 3.27 | 5.93 | |
| Min. | 25.12 | 4.54 | 27.57 | 11.92 | 0.60 | 2.64 | |

| FM-L∩$Pd^{2+}$ | C | N | O | Si | Fe | Pd | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 20.00 | 6.98 | 33.33 | 14.67 | 0.94 | 24.08 | 100.00 |
| Spectrum 2 | 25.97 | 5.50 | 30.02 | 16.88 | 0.96 | 20.67 | 100.00 |
| Spectrum 3 | 22.70 | 7.56 | 32.17 | 12.16 | 0.90 | 24.50 | 100.00 |
| Spectrum 4 | 22.40 | 7.30 | 30.51 | 13.22 | 0.61 | 25.98 | 100.00 |
| Spectrum 5 | 16.35 | 4.53 | 29.49 | 15.57 | 1.20 | 32.87 | 100.00 |
| Spectrum 6 | 31.23 | 5.98 | 27.62 | 11.99 | 1.36 | 21.81 | 100.00 |
| Spectrum 7 | 20.52 | 7.26 | 36.86 | 15.51 | 1.07 | 18.77 | 100.00 |
| Spectrum 8 | 19.71 | 7.40 | 36.22 | 15.27 | 1.44 | 19.96 | 100.00 |
| Spectrum 9 | 16.57 | 4.18 | 25.50 | 16.01 | 4.19 | 33.55 | 100.00 |
| Spectrum 10 | 19.24 | 5.41 | 28.96 | 17.61 | 1.83 | 26.94 | 100.00 |
| **Mean** | **21.47** | **6.21** | **31.07** | **14.89** | **1.45** | **24.91** | **100.00** |
| Std. deviation | 4.46 | 1.26 | 3.62 | 1.89 | 1.02 | 5.10 | |
| Max. | 31.23 | 7.56 | 36.86 | 17.61 | 4.19 | 33.55 | |
| Min. | 16.35 | 4.18 | 25.50 | 11.99 | 0.61 | 18.77 | |

| FM-L∩$Hg^{2+}$ | C | N | O | Si | Fe | Hg | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 31.69 | 6.62 | 33.44 | 13.26 | 1.08 | 13.91 | 100.00 |
| Spectrum 2 | 22.38 | 7.32 | 39.83 | 17.72 | 1.87 | 10.89 | 100.00 |
| Spectrum 3 | 22.15 | 6.55 | 43.00 | 19.28 | 1.58 | 7.43 | 100.00 |
| Spectrum 4 | 18.33 | 4.30 | 43.36 | 26.19 | 1.72 | 6.10 | 100.00 |
| Spectrum 5 | 21.60 | 4.96 | 45.65 | 21.90 | 1.35 | 4.55 | 100.00 |
| Spectrum 6 | 29.14 | 8.98 | 36.34 | 13.75 | 1.26 | 10.53 | 100.00 |
| Spectrum 7 | 22.47 | 7.23 | 36.32 | 14.02 | 1.25 | 18.71 | 100.00 |
| Spectrum 8 | 30.14 | 8.34 | 38.22 | 15.21 | 1.06 | 7.04 | 100.00 |
| Spectrum 9 | 19.80 | 4.61 | 44.10 | 22.52 | 1.89 | 7.07 | 100.00 |
| Spectrum 10 | 19.23 | 4.85 | 39.97 | 27.75 | 1.80 | 6.40 | 100.00 |
| **Mean** | **23.69** | **6.37** | **40.02** | **19.16** | **1.49** | **9.26** | **100.00** |
| Std. deviation | 4.82 | 1.64 | 3.98 | 5.27 | 0.32 | 4.33 | |
| Max. | 31.69 | 8.98 | 45.65 | 27.75 | 1.89 | 18.71 | |
| Min. | 18.33 | 4.30 | 33.44 | 13.26 | 1.06 | 4.55 | |

| FM-L∩$Ni^{2+}$ | C | N | O | Si | Fe | Ni | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 22.35 | 7.46 | 40.14 | 22.24 | 1.78 | 6.04 | 100.00 |
| Spectrum 2 | 26.43 | 6.34 | 41.36 | 17.33 | 1.24 | 7.30 | 100.00 |
| Spectrum 3 | 27.54 | 6.79 | 34.54 | 16.79 | 3.57 | 10.77 | 100.00 |
| Spectrum 4 | 23.70 | 3.68 | 46.31 | 20.87 | 1.12 | 4.33 | 100.00 |
| Spectrum 5 | 25.55 | 7.90 | 38.09 | 20.30 | 1.74 | 6.42 | 100.00 |
| Spectrum 6 | 24.06 | 6.95 | 40.95 | 21.18 | 1.44 | 5.41 | 100.00 |
| Spectrum 7 | 22.02 | 6.05 | 32.35 | 26.99 | 2.31 | 10.28 | 100.00 |
| Spectrum 8 | 25.82 | 7.62 | 42.62 | 18.14 | 1.14 | 4.66 | 100.00 |
| Spectrum 9 | 26.43 | 8.68 | 43.31 | 15.55 | 1.01 | 5.02 | 100.00 |
| Spectrum 10 | 24.36 | 5.81 | 43.20 | 19.39 | 1.27 | 5.97 | 100.00 |
| **Mean** | **24.83** | **6.73** | **40.29** | **19.88** | **1.66** | **6.62** | **100.00** |
| Std. deviation | 1.83 | 1.39 | 4.24 | 3.29 | 0.78 | 2.24 | |
| Max. | 27.54 | 8.68 | 46.31 | 26.99 | 3.57 | 10.77 | |
| Min. | 22.02 | 3.68 | 32.35 | 15.55 | 1.01 | 4.33 | |

| **FM-L∩$Pb^{2+}$** | **C** | **N** | **O** | **Si** | **Fe** | **Pb** | **Total** |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 21.49 | 6.84 | 38.44 | 16.22 | 0.88 | 16.13 | 100.00 |
| Spectrum 2 | 21.43 | 6.53 | 38.09 | 16.31 | 0.93 | 16.72 | 100.00 |
| Spectrum 3 | 21.38 | 7.49 | 38.32 | 14.89 | 1.01 | 16.90 | 100.00 |
| Spectrum 4 | 18.46 | 5.49 | 35.00 | 17.92 | 0.87 | 22.26 | 100.00 |
| Spectrum 5 | 17.72 | 5.01 | 35.38 | 17.66 | 1.15 | 23.08 | 100.00 |
| Spectrum 6 | 22.13 | 7.00 | 35.48 | 13.83 | 0.80 | 20.75 | 100.00 |
| Spectrum 7 | 23.05 | 7.85 | 36.41 | 13.39 | 0.74 | 18.57 | 100.00 |
| Spectrum 8 | 23.36 | 7.49 | 36.07 | 14.07 | 0.73 | 18.29 | 100.00 |
| Spectrum 9 | 22.75 | 6.63 | 36.43 | 14.54 | 0.55 | 19.09 | 100.00 |
| Spectrum 10 | 22.91 | 7.29 | 34.93 | 14.70 | 0.52 | 19.66 | 100.00 |
| **Mean** | **21.47** | **6.76** | **36.45** | **15.35** | **0.82** | **19.15** | **100.00** |
| Std. deviation | 1.92 | 0.90 | 1.37 | 1.59 | 0.20 | 2.34 | |
| Max. | 23.36 | 7.85 | 38.44 | 17.92 | 1.15 | 23.08 | |
| Min. | 17.72 | 5.01 | 34.93 | 13.39 | 0.52 | 16.13 | |

| **FM-L∩$Cd^{2+}$** | **C** | **N** | **O** | **Si** | **Fe** | **Cd** | **Total** |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 26.81 | 7.83 | 38.55 | 13.14 | 0.55 | 13.13 | 100.00 |
| Spectrum 2 | 24.99 | 7.19 | 41.09 | 13.38 | 0.78 | 12.57 | 100.00 |
| Spectrum 3 | 22.68 | 6.77 | 40.63 | 20.76 | 0.32 | 8.84 | 100.00 |
| Spectrum 4 | 26.00 | 7.78 | 40.20 | 14.29 | 0.49 | 11.23 | 100.00 |
| Spectrum 5 | 20.04 | 5.42 | 40.60 | 23.46 | 0.49 | 9.98 | 100.00 |
| Spectrum 6 | 21.32 | 5.49 | 40.77 | 22.47 | 0.37 | 9.58 | 100.00 |
| Spectrum 7 | 21.18 | 5.37 | 39.93 | 23.20 | 0.32 | 10.00 | 100.00 |
| Spectrum 8 | 25.97 | 6.84 | 36.26 | 14.61 | 0.61 | 15.71 | 100.00 |
| Spectrum 9 | 20.57 | 5.06 | 40.90 | 23.44 | 0.22 | 9.80 | 100.00 |
| Spectrum 10 | 26.22 | 6.46 | 40.82 | 13.60 | 0.55 | 12.35 | 100.00 |
| **Mean** | **23.58** | **6.42** | **39.98** | **18.24** | **0.47** | **11.32** | **100.00** |
| Std. deviation | 2.67 | 1.03 | 1.50 | 4.75 | 0.17 | 2.11 | |
| Max. | 26.81 | 7.83 | 41.09 | 23.46 | 0.78 | 15.71 | |
| Min. | 20.04 | 5.06 | 36.26 | 13.14 | 0.22 | 8.84 | |

| **FM-L∩$Mn^{2+}$** | **C** | **N** | **O** | **Si** | **Fe** | **Mn** | **Total** |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 22.71 | 6.65 | 41.41 | 22.76 | 1.63 | 4.84 | 100.00 |
| Spectrum 2 | 23.30 | 7.19 | 39.79 | 22.43 | 2.43 | 4.87 | 100.00 |
| Spectrum 3 | 22.29 | 6.04 | 39.29 | 24.73 | 2.14 | 5.50 | 100.00 |
| Spectrum 4 | 23.27 | 7.65 | 43.70 | 20.01 | 1.66 | 3.73 | 100.00 |
| Spectrum 5 | 22.55 | 6.07 | 39.78 | 24.95 | 1.80 | 4.85 | 100.00 |
| Spectrum 6 | 24.18 | 6.49 | 38.95 | 23.08 | 2.22 | 5.08 | 100.00 |
| Spectrum 8 | 22.22 | 3.63 | 33.14 | 32.58 | 1.50 | 6.92 | 100.00 |
| Spectrum 9 | 26.97 | 7.54 | 38.29 | 17.44 | 0.72 | 9.04 | 100.00 |
| Spectrum 10 | 27.16 | 8.44 | 42.00 | 17.19 | 1.14 | 4.07 | 100.00 |
| **Mean** | **23.85** | **6.63** | **39.59** | **22.80** | **1.69** | **5.43** | **100.00** |
| Std. deviation | 1.92 | 1.37 | 2.95 | 4.64 | 0.54 | 1.63 | |
| Max. | 27.16 | 8.44 | 43.70 | 32.58 | 2.43 | 9.04 | |
| Min. | 22.22 | 3.63 | 33.14 | 17.19 | 0.72 | 3.73 | |

| FM-L∩$Sn^{4+}$ | C | N | O | Si | Fe | Sn | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 14.56 | 0.00 | 42.82 | 27.46 | 1.82 | 13.33 | 100.00 |
| Spectrum 2 | 16.21 | 0.00 | 43.69 | 25.78 | 1.21 | 13.11 | 100.00 |
| Spectrum 3 | 24.63 | 5.46 | 43.85 | 11.92 | 0.63 | 13.52 | 100.00 |
| Spectrum 4 | 24.96 | 5.91 | 44.98 | 12.01 | 0.58 | 11.56 | 100.00 |
| Spectrum 5 | 21.36 | 0.00 | 44.19 | 16.37 | 1.44 | 16.65 | 100.00 |
| Spectrum 6 | 19.71 | 2.69 | 43.06 | 16.76 | 1.41 | 16.37 | 100.00 |
| Spectrum 7 | 21.40 | 0.00 | 44.66 | 16.41 | 1.09 | 16.44 | 100.00 |
| Spectrum 8 | 20.68 | 4.72 | 41.11 | 18.74 | 1.53 | 13.22 | 100.00 |
| Spectrum 9 | 20.45 | 6.65 | 41.44 | 17.58 | 1.25 | 12.63 | 100.00 |
| Spectrum 10 | 22.09 | 0.00 | 43.16 | 18.04 | 1.14 | 15.57 | 100.00 |
| **Mean** | **20.60** | **2.54** | **43.30** | **18.11** | **1.21** | **14.24** | **100.00** |
| Std. deviation | 3.25 | 2.86 | 1.27 | 5.06 | 0.38 | 1.84 | |
| Max. | 24.96 | 6.65 | 44.98 | 27.46 | 1.82 | 16.65 | |
| Min. | 14.56 | 0.00 | 41.11 | 11.92 | 0.58 | 11.56 | |

| FM-L∩$Tl^{+}$ | C | N | O | Si | Fe | Tl | Total |
|---|---|---|---|---|---|---|---|
| Spectrum 1 | 22.63 | 8.37 | 44.00 | 18.52 | 1.12 | 5.37 | 100.00 |
| Spectrum 2 | 25.78 | 6.17 | 31.77 | 26.16 | 1.77 | 8.35 | 100.00 |
| Spectrum 3 | 27.47 | 7.39 | 33.04 | 23.59 | 1.62 | 6.89 | 100.00 |
| Spectrum 4 | 26.45 | 9.03 | 40.84 | 17.30 | 1.49 | 4.89 | 100.00 |
| Spectrum 5 | 24.65 | 9.21 | 42.40 | 17.34 | 2.10 | 4.30 | 100.00 |
| Spectrum 6 | 23.51 | 7.38 | 42.26 | 20.74 | 1.05 | 5.06 | 100.00 |
| Spectrum 7 | 27.88 | 9.24 | 38.27 | 17.61 | 1.28 | 5.72 | 100.00 |
| Spectrum 8 | 30.24 | 9.92 | 41.55 | 13.14 | 0.98 | 4.17 | 100.00 |
| Spectrum 9 | 33.57 | 13.07 | 35.85 | 11.73 | 0.72 | 5.07 | 100.00 |
| Spectrum 10 | 24.06 | 7.17 | 38.58 | 22.58 | 2.11 | 5.51 | 100.00 |
| **Mean** | **26.62** | **8.69** | **38.86** | **18.87** | **1.42** | **5.53** | **100.00** |
| Std. deviation | 3.34 | 1.93 | 4.15 | 4.51 | 0.47 | 1.25 | |
| Max. | 33.57 | 13.07 | 44.00 | 26.16 | 2.11 | 8.35 | |
| Min. | 22.63 | 6.17 | 31.77 | 11.73 | 0.72 | 4.17 | |

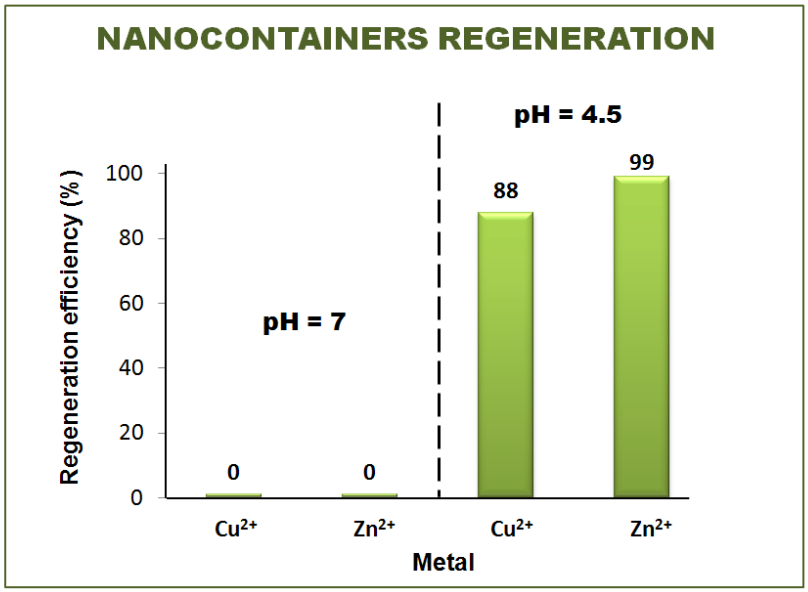


**Figure S11.** Nanocontainers regeneration determined by inductively-coupled plasma measurements of the collected acidic solution used for the copper and zinc ion desorptions.

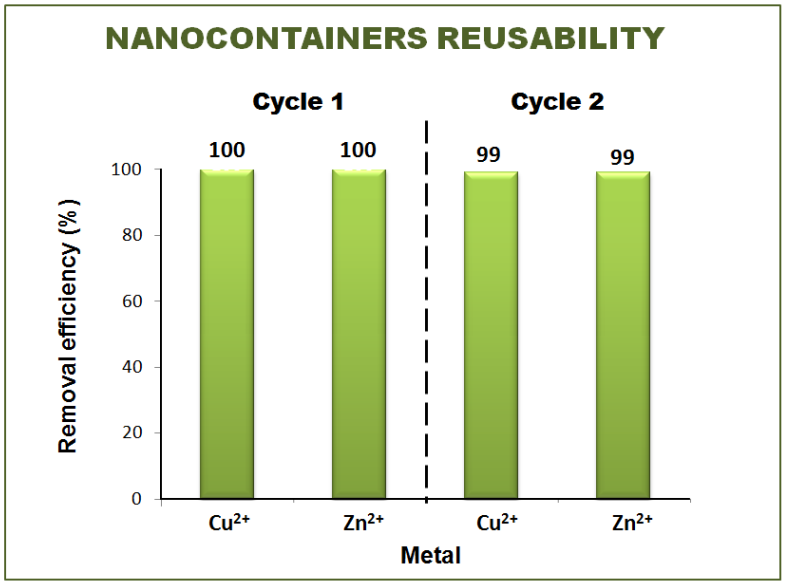


**Figure S12.** Reusability of the nanocontainers after regeneration for heavy metals removal.

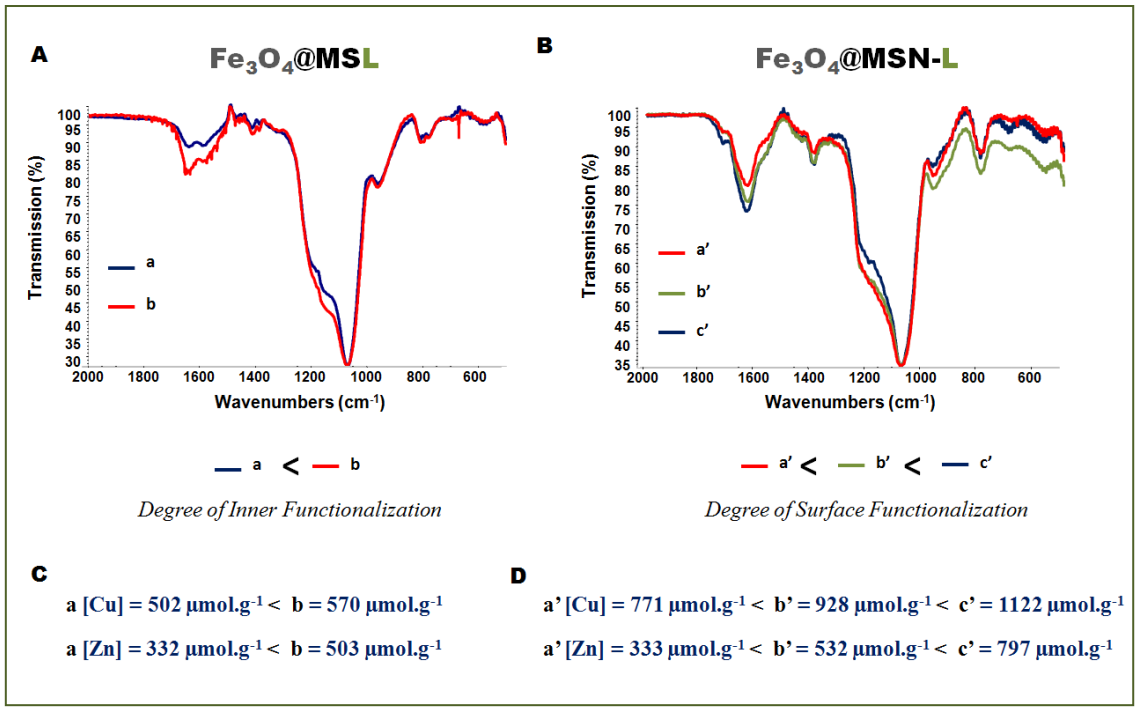


**Figure S13.** Study of the influence of the degree of ligand functionalization, in the pores or on the surface, on the heavy metals removal. IR spectra of the $Fe_3O_4$@MSL, and $Fe_3O_4$@MSN-L NPs with different degrees of functionalization (A-B). EDS metal adsorption capacities of $Fe_3O_4$@MSL∩$Mn^{n+}$ (C), and $Fe_3O_4$@MSN-L∩$Mn^{n+}$ (D), demonstrating the influence of the ligand in the pollutant removal.

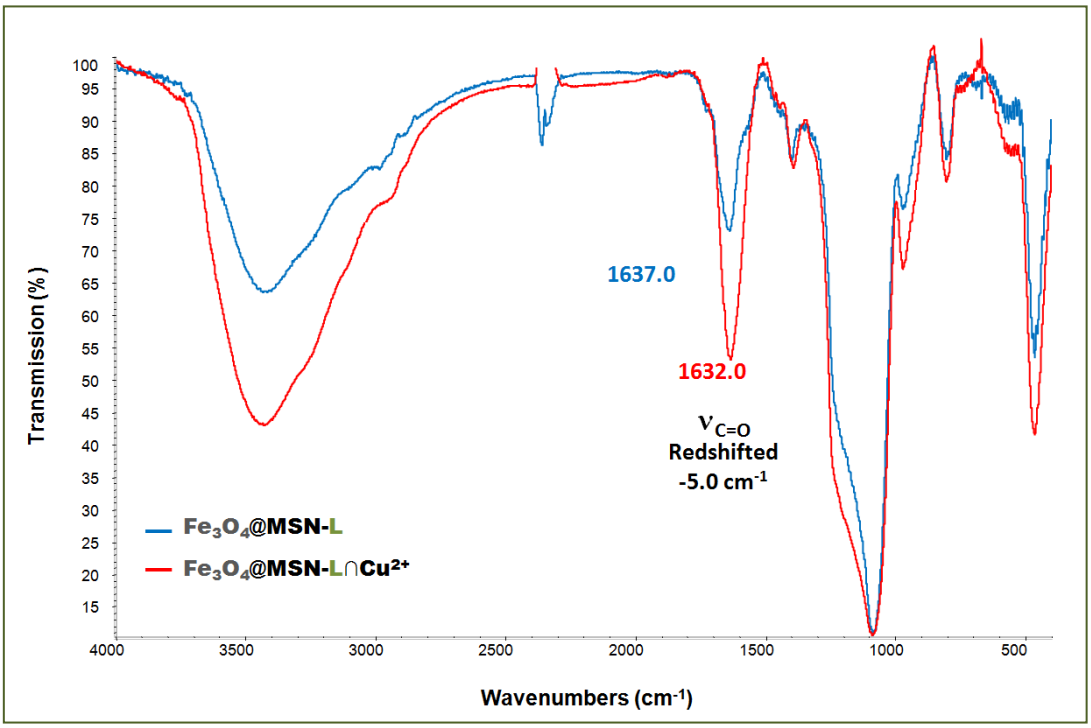


**Figure S14.** IR spectra of the $Fe_3O_4$@MSN-L, and $Fe_3O_4$@MSN-L∩$Cu^{2+}$ NPs, confirming the successful complexation of copper ions.

 

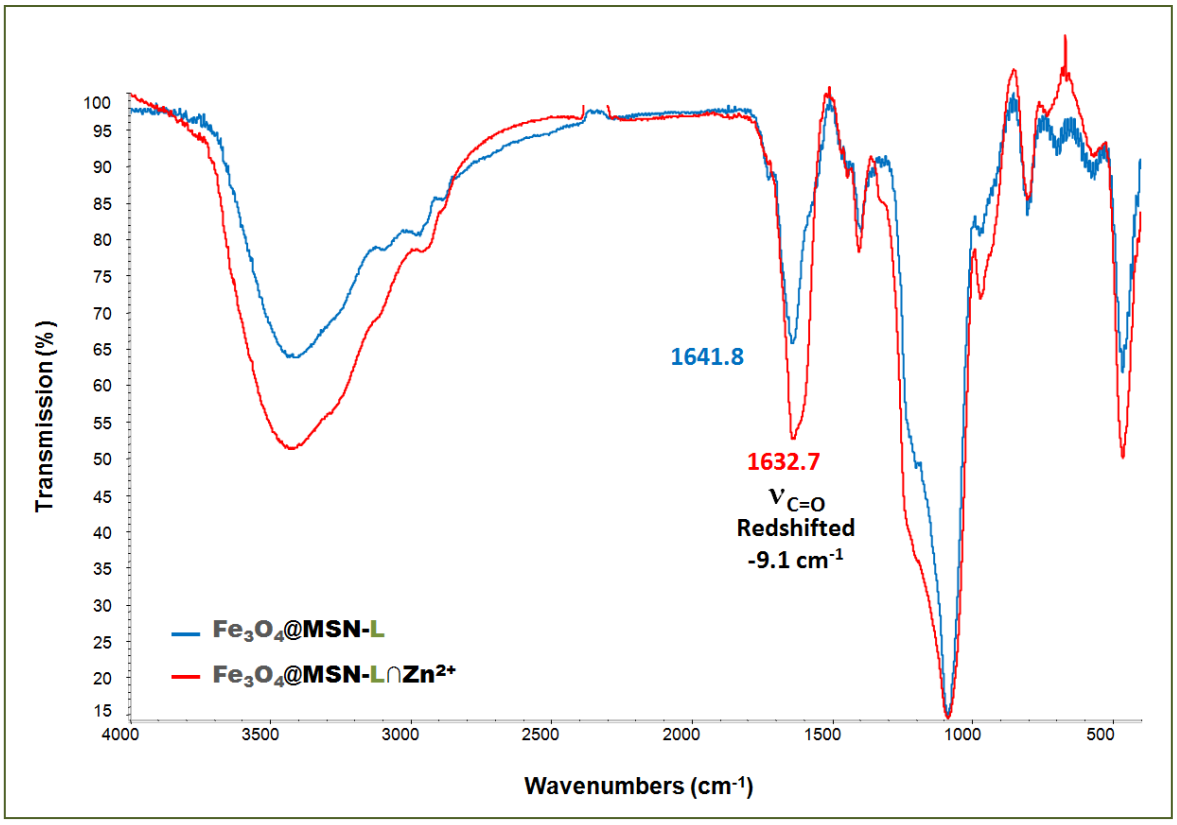


**Figure S15.** IR spectra of the $Fe_3O_4$@MSN-L, and $Fe_3O_4$@MSN-L∩$Zn^{2+}$ NPs, confirming the successful complexation of zinc ions.

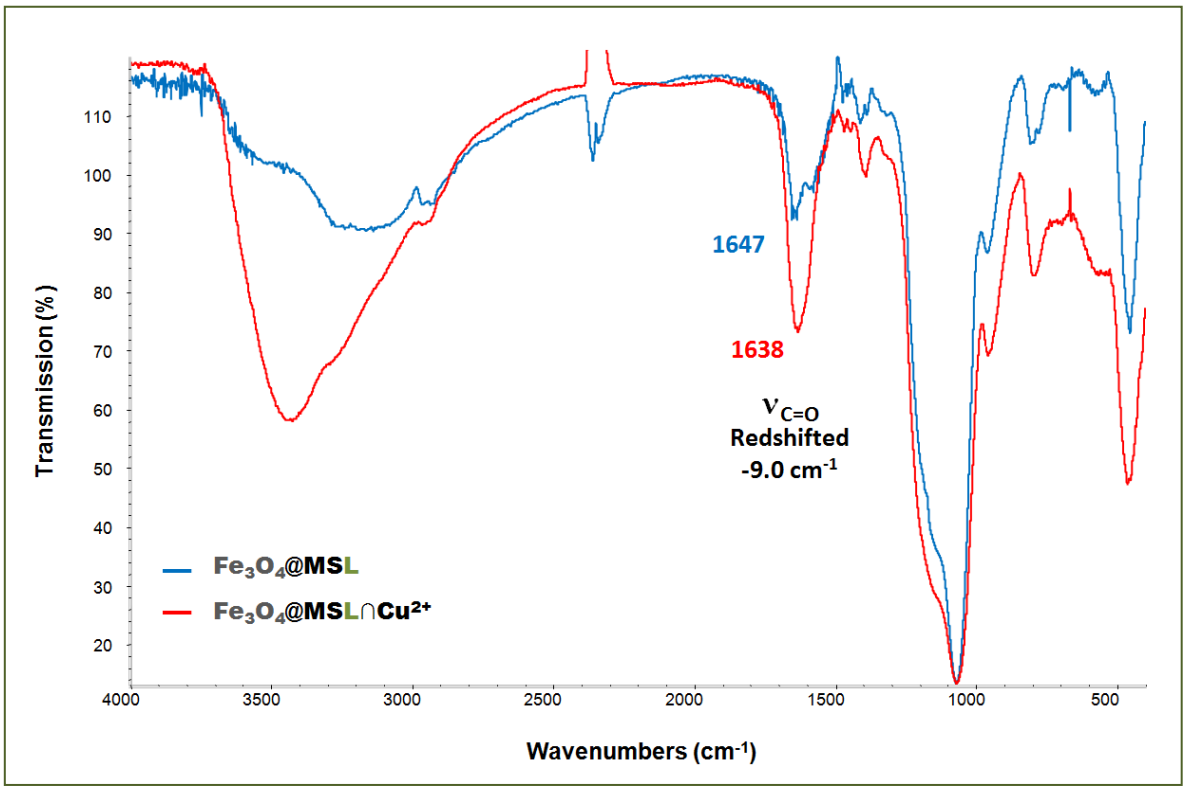


**Figure S16.** IR spectra of the $Fe_3O_4$@MSL, and $Fe_3O_4$@MSL∩$Cu^{2+}$ NPs, confirming the successful complexation of copper ions.

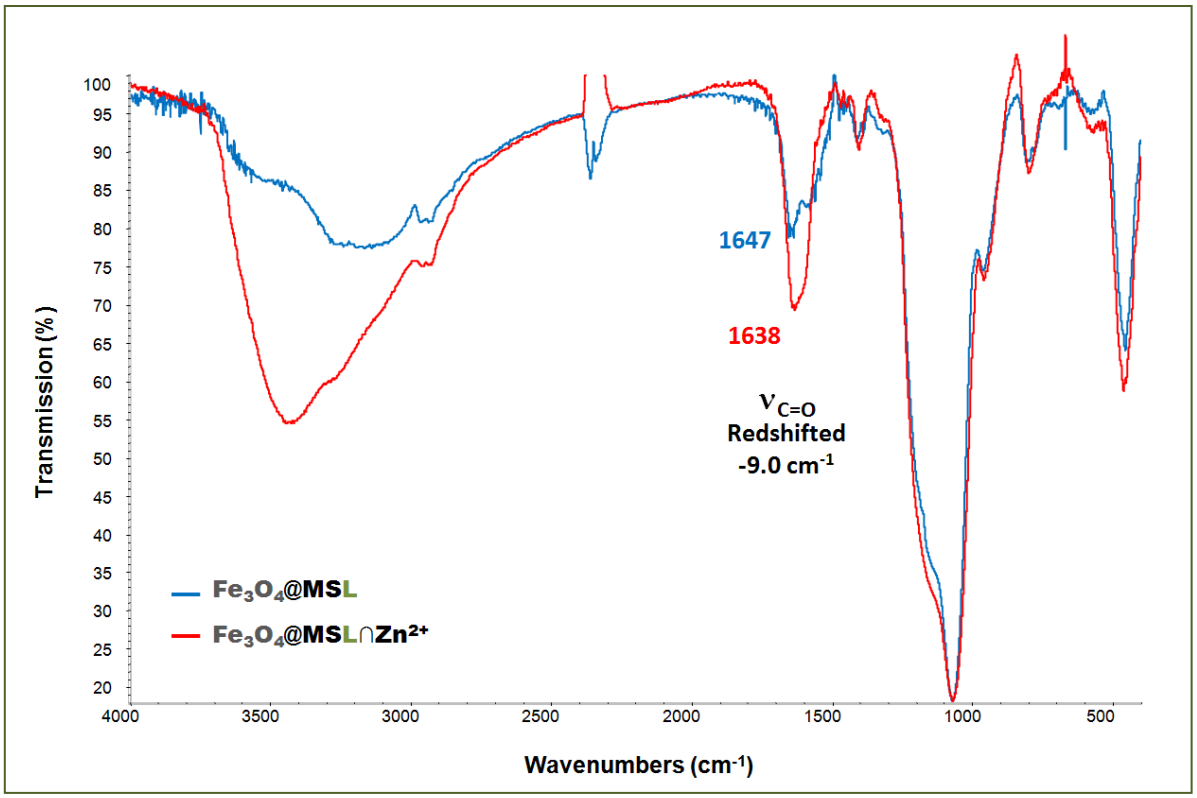


**Figure S17.** IR spectra of the $Fe_3O_4$@MSL, and $Fe_3O_4$@MSL∩$Zn^{2+}$ NPs, confirming the successful complexation of zinc ions.