Electronic Supplementary Information

Experimental and computational modeling studies on silica-embedded NiO/MgO nanoparticles for adsorptive removal of organic pollutants from wastewater

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S1. Synthesis of silica-embedded NiO and MgO nanoparticles

The following subsections show the in-house preparation methods for synthesising three different types of silica-embedded NiO and MgO nanoparticles which were developed as promising nanosorbcats for the removal of organic pollutants in wastewater.

S1.1 Silica-embedded NiO nanoparticles

Silica-embedded NiO nanoparticles preparation method can be summarized under the following major steps. First, 4.3 g of colloidal silica (Ludox AS-40, SiO_2 40% (w/w)) was added to a 1 L glass beaker containing 200 mL of deionized water. The mixture was agitated using a magnetic stirrer (300 rpm) for 2 min until a homogenous solution was obtained. Then, while keeping the magnetic stirring at 300 rpm, 16.8 g of $Ni(NO_3)_2.6H_2O$ (precursor for NiO nanoparticles) was added to the mixture. After complete dissolution of the nickel salt, 13.0 g of granulated household sucrose was added to the mixture. Sucrose has two main functions in this preparation method. First, it helps to distribute the Ni within the aqueous mixture, so it works like a complexing agent for the Ni and silica. Second, it acts like a temperature reducer for the synthesis of the dried powder (auto-ignition process). Afterwards, 1.5 mL of HNO_3 70% (w/w) was added dropwise to the mixture. The mixture was magnetically stirred at 300 rpm and 293 K for 30 min to produce a homogeneous blackish solution. The beaker was then placed in the oven

at 373 K for 48 h to evaporate the water. On the third day, a dried black solid material looking like a sponge cake was obtained. This solid material was re-placed in the oven at 523 K for 12 h. This step was carried out to ensure that there was no free water left in the solid material that could affect the auto-ignition process, and to ensure that all the amorphous materials, atomically speaking, got closer to each other, for the next step. Eventually, the solid material was taken out from the oven and then grinded very carefully in the fume hood until a homogenous powder was obtained and then calcined in the oven at 1073 K for 3 h in a flow of air and ramping at 5 K/min. This was done to release the carbonaceous part of the material allowing the production of the Silica-embedded NiO nanoparticles with around 25 nm crystalline domain size for NiO and less than 100 nm for the silica. The main framework components of the resultant nanoparticles structure are SiO_2 and NiO as confirmed by infrared spectroscopy and the X-ray structural identification analyses, respectively. Thus, the prepared silica-embedded NiO nanoparticles were labeled as SiO_2-NiO.

S1.2 Silica-embedded MgO nanoparticles

Silica-embedded MgO nanoparticles preparation method went through the following major steps. 4.0 mL of NH_4OH 27% (w/w) was added dropwise to a 1 L glass beaker containing 300 mL of deionized water, then, 4.0 g of H_2SiO_3 was added to the mixture and agitated using a magnetic stirrer for 10 min at 300 rpm. After that, 20.0 g of granulated household sucrose was added to the mixture. Then, 24.0 g of $Mg(NO_3)_2.6H_2O$ (precursor for MgO nanoparticles) was added to the mixture. After complete dissolution of the magnesium salt, 4.0 mL of HNO_3 70% (w/w) was added dropwise to the mixture. The mixture was magnetically stirred at 300 rpm and 293 K for 30 min to produce a homogeneous brownish solution. The beaker was then placed in the oven at 373 K for 48 h to evaporate the water. On the third day, a dried brown solid material was

obtained. This solid material was re-placed in the oven at 523 K for 12 h. At the end, the solid material was taken out from the oven and then grinded very carefully in the fume hood until a homogenous powder was obtained and then calcined in the oven at 1073 K for 3 h in a flow of air and ramping at 5 K/min. It is worth mentioning here that after calcination the powder material turned completely from brown to white color. The main framework components of the resultant nanoparticles structure are SiO_2 and MgO as confirmed by infrared spectroscopy and the X-ray structural identification analyses, respectively. Thus, the prepared silica-embedded MgO nanoparticles were labeled as SiO_2-MgO.

S1.3 Silica-embedded NiO and MgO nanoparticles

For the sake of studying the possible synergistic effect of bimetallic nanoparticles towards adsorption of cationic and anionic pollutants, silica-embedded NiO and MgO (at ratio of 0.5:0.5) nanoparticles were also prepared in this study. The preparation method is summarized as follows: 5.1 g of colloidal silica (Ludox AS-40, SiO_2 40% (w/w)) was added to a 1 L beaker containing 225 mL of deionized water. The mixture was agitated using a magnetic stirrer for 2 min until a homogenous solution was obtained. Then, while keeping the magnetic stirring, 9.9 g of $Ni(NO_3)_2.6H_2O$ (precursor for NiO nanoparticles) and 9.1 g of $Mg(NO_3)_2.6H_2O$ (precursor for MgO nanoparticles) were added to the mixture. After complete dissolution of the nickel and magnesium salts, 15.0 g of granulated household of sucrose and 1.5 mL of HNO_3 70% (w/w) were added to the mixture. The mixture was magnetically stirred at 300 rpm and 293 K for 30 min to produce a homogeneous greyish solution. The beaker was then placed in the oven at 373 K for 48 h to evaporate the water. Afterwards, the solid material was re-placed in the oven at 523 K for 12 h. Finally, the solid material was taken out from the oven and then grinded very carefully in the fume hood until a homogenous powder was obtained and then calcined in the

oven at 1073 K for 3 h in a flow of air and ramping at 5 K/min. The main framework components of the resultant nanoparticles structure are SiO_2, NiO, and MgO as confirmed by infrared spectroscopy and the X-ray structural identification analyses, respectively. Thus, the bimetallic prepared silica-embedded NiO and MgO nanoparticles were labeled as SiO_2-$(Ni_{0.5}Mg_{0.5})O$.

S2. Detailed calculations for the NiO and MgO contents

S2.1 SiO₂-NiO nanoparticles

For calculating the molecular weight M_w of SiO_2-NiO nanoparticles, the following stoichiometry should be taken into consideration, $Ni_2SiO_4 = 2NiO + SiO_2.Ni(NO_3)_2.6H_2O$. 16.8 g,

$$M_w = 58.7 + 2 \times 14 + 6 \times 16 + 6 \times 18 = 290.81 \ g/mol$$

$Ludux\ 40\ wt\%\ SiO_2:4.267g$

$\#g\ of\ SiO_2 = 4.267g \times 0.4 = 1.7068\ g\ SiO_2$

$$\#mole\ SiO_2 = \frac{1.7068\ g\ SiO_2}{60g/mol\ SiO_2} = 0.02845\ mol\ SiO_2$$

$\#mole\ Ni = \#mole\ NiO = \#moleNi(NO_3)_2.6H_2O$

$$= \frac{16.8\ g}{290.81g/mol} = 0.0578\ mol\ Ni$$

$$Ratio\ of\ \frac{Ni}{Si} = \frac{0.0578\ mol\ Ni}{0.02845\ mol\ Si} = 2.03 \cong 2$$

$\#g\ of\ NiO = 0.0578\ mol\ Ni \times 74.7\ \frac{g}{mol}NiO = 4.32g\ NiO$

$\%\ of\ NiO\ in\ the\ prep\ sample\ of\ SiO_2 - NiO = \dfrac{4.32}{4.32 + 1.7068} \times 100\% = 71.67\% \cong 72\%$

S2.1 SiO₂-MgO nanoparticles

For calculating the molecular weight M_w of SiO_2-MgO nanoparticles, the following stoichiometry should be taken into consideration, $Mg_2SiO_4 = 2MgO + SiO_2$.

$Mg(NO_3)_2.6H_2O = 24\ g,\ M_{w1} = 24.3 + 14 \times 2 + 16 \times 6 + 6 \times 18 = 256.3g/mol$

$H_2SiO_3 = 4\ g$

$$\#g\ SiO_2 = 4\ g\ H_2SiO_3 \times \frac{60\ (g/mol)\ SiO_2}{78\ (g/mol)\ H_2SiO_3} = 3.0769\ g\ SiO_2$$

$$\#mol\ SiO_2 = \frac{3.0769\ g\ SiO_2}{60\ (g/mol)\ SiO_2} = 0.0513\ mol\ SiO_2$$

$\#mole\ Mg = mole\ of\ MgO = mole\ of\ Mg(NO_3)_2.6H_2O = 256.3 = 0.0936\ mol\ Mg$

$$Ratio\ of\ \frac{Mg}{Si} = \frac{0.0936}{0.0513} = 1.83$$

$$\#g\ of\ MgO = 0.0936\ mol\ Mg \times \frac{40.3\ g}{1\ mol} MgO = 3.7721\ g$$

$$\%\ of\ MgO\ in\ the\ prepared\ sample\ of\ SiO_2 - MgO = \frac{3.7721}{3.7721 + 3.0769} = 55.1\%$$

S2.1 SiO$_2$-(Ni$_{0.5}$Mg$_{0.5}$)O nanoparticles

For calculating the molecular weight M_w of SiO$_2$-(Ni$_{0.5}$Mg$_{0.5}$)O nanoparticles, the following stoichiometry should be taken into consideration, $(Ni_{0.5}Mg_{0.5})O - SiO_2 = NiO + MgO + SiO_2$;

$Mg(NO_3)_2.6H_2O{:}9.1\ g, M_{wt} = 256.41\ g/mol$

$Ni(NO_3)_2.6H_2O{:}9.9\ g, M_{wt} = 290.81\ g/mol$

$Ludux\ 40\ wt\%\ SiO_2{:}5.112\ g \rightarrow \#g\ SiO_2 = 5.112 \times 0.4 = 2.0448\ g\ SiO_2$

$$\#mol\ SiO_2 = \frac{2.0448\ g}{60\ g/mol} = 0.03549\ mol\ MgO$$

$$Ratio\ of\ \frac{Ni}{Si} = \frac{0.03404}{0.03408} = 0.9988 \cong 1, Ratio\ of\ \frac{Mg}{Si} = \frac{0.03549}{0.03408} = 1.04 \cong 1$$

$\#g\ of\ NiO = 0.03404 \times 74.7 = 2.542\ g\ NiO,$

$\#g\ of\ MgO = 0.03549 \times 40.3 = 1.4302\ g\ MgO$

$$\%\ of\ NiO = \frac{2.5428}{2.5428 + 1.4302 + 2.04448} = 0.4225 = 42.25\%$$

$$\%of\ MgO = \frac{1.4302}{6.0178} 0.2380 \cong 23.8\%$$

Table S1. Estimated NiO and MgO percentage contents in the prepared nanoparticles.

Nanoparticles	NiO (%)	MgO (%)
SiO$_2$-NiO	72	-
SiO$_2$-MgO	-	55
SiO$_2$-(Mg$_{0.5}$Ni$_{0.5}$)O	42	24