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Application of AgNPs-Doped MIL-53 in The Catalytic Reduction of Methylene Blue MB Dye

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ABSTRACT

This work focuses on the preparation of MIL-53 material by hydrothermal route. The obtained material was used as a support for AgNPs silver nanoparticles. Several percentages of AgNPs (0.5%, 1%, 2%, and 3%) were dispersed on the surface of MIL-53 to study their catalytic behavior. The obtained solids were characterized by different characterization methods in order to determine their physicochemical properties. The parent material and its AgNPs-modified counterparts were tested as catalysts for the catalytic reduction of MB dye under the presence of NaBH4. The obtained results clearly showed the presence of AgNPs on the surface of MIL-53 after its chemical modification. The obtained results showed that the reduction of MB is favored by catalysts having a high content of AgNPs. A high conversion of MB dye to Leuco-MB was obtained in reaction time which did not exceed 5 min.

Keywords: MIL-53, AgNPs, MB dye, catalytic reduction.

1 Introduction

Wastewater pollution has become a topical problem following industrial discharges. Among the most harmful discharges, dyes have been considered dangerous pollutants due to their toxicity. It has been shown in several studies that low traces of dyes can cause serious and fatal problems for all living species. Different strategies have been established to reduce the pollution of contaminated water. Among the most used methods, there are adsorption, photocatalysis, oxidation, reduction, flocculation-coagulation and other methods. Catalytic reduction has seen wide application due to the speed of the process and also the products formed can be used in several chemicals. The presence of nanoparticles and their sizes are the most important parameters to activate the reduction of organic pollutants. However, due to aggregation problems, metal nanoparticles cannot be used directly. One of the most used methods is to disperse them on solid supports. Recently a new discovery of porous materials of metal-organic framework type having remarkable textural properties. Due to their surface properties, these materials have been applied in several fields such as adsorption, separation, catalysis, and catalytic support. The main objective of this work is the preparation of silver nanoparticles supported on MIL-53. To optimize the best MB dye reduction conditions, several percentages of AgNPs (0.5%, 1%, 2%, and 3%) were dispersed on the surface of MIL-53. The obtained results were correlated between the contents of silver nanoparticles and the reducing agent $NaBH_4$.

2 Experimental

2.1 Preparation of MIL-53@AgNPs

The MIL-53 material was synthesized hydrothermally using a method well described in the literature[1,2]. After the purification of MIL-53, this material was modified with an $AgNO_3$ solution using different percentages 0.5%, 1%, 2%, and 3%. The obtained suspensions were washed, then filtered and dried. The

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obtained materials were modified with a 1M $NaBH_4$ [3,4]solution to transform the Ag^+ species into AgNPs. The obtained materials MIL-53@AgNPs(0.5%), MIL-53@AgNPs(1%), MIL-53@AgNPs(2%), and MIL-53@AgNPs(3%) were named according to the percentage of AgNPs doped in each material.

2.2 Catalytic reduction of MB dye

The MB dye reduction protocol is very similar to our previously published work[3,5,6]. Generally, 3mg and 2mL of MB solution (0.1 mM) was introduced into a cuvette then approximately 1.5 mL of freshly prepared $NaBH_4$ solution (15 mM) was added, the reaction mixture was monitored by UV-vis.[3,5,6]

3 Results and Discussion

The different catalysts were characterized by XRD analysis (results not shown). It was found that as the concentration of $AgNO_3$ increases, the characteristic peaks of AgNPs increase due to the formation of AgNPs layers on the MIL-53 surface. The catalytic application of these catalysts in the reduction of MB dye showed interesting results. Initially, MB adsorption tests and blank test ($NaBH_4$ + MB solution) were used to confirm the role of our catalytic system. It was found that the different catalysts did not exhibit MB adsorption affinity. While the blank test without catalyst showed no catalytic reaction. After the addition of a few traces of MIL-53@AgNPs(2%) catalyst, a drastic decrease in MB dye intensity was observed and total conversion of MB was obtained (see Figure 1a and b). The role of our catalytic system lies in its ability to transport electrons from the donor NaBH4 to the dye acceptor MB dye, which subsequently leads to the transformation of MB dye to Leuco-MB. It was found that the content of AgNPs plays a very important role in the MB reduction process. The higher AgNPs content lead to the faster reduction of MB dye (results not shown). Under all the conditions used in this study it was found that the MIL-53@AgNPs(2%) was the best catalyst which completely reduced the MB dye in a reaction time of 5 min.

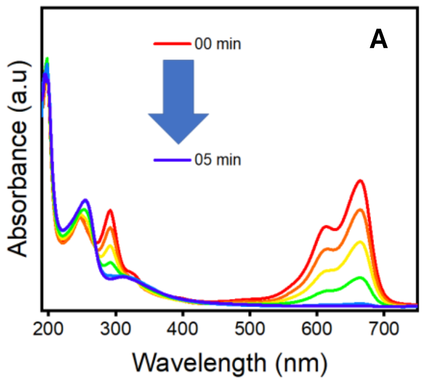

Figure1 UV-vis of MB dyes during their reduction

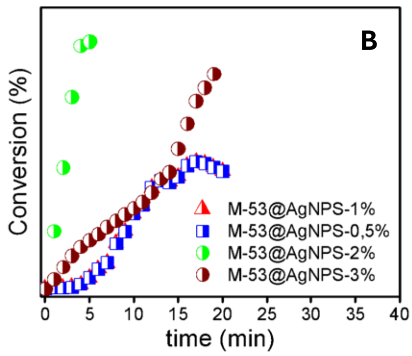

Figure2 MB Conversion by different catalyst M-53@AgNPS-(0.5,1,2,3%)

4 Conclusions

In conclusion, the use of MIL-53 presents more advantages, it makes it possible to obtain good dispersion of AgNPs, limitation of the aggregation phenomenon and ultrafine particles of AgNPs were obtained. The application of this system in the reduction of MB dye as a model reaction showed that the reaction in the presence of $NaBH_4$ alone does not lead to any transformation. After the addition of the catalyst MIL-53@AgNPs(2%), a complete reduction of MB to Leuco-MB was obtained within 5 minutes. The use of catalysts containing a low content of AgNPs does not lead to a significant reduction following the presence of weak sites on the surface of MIL-53.

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