Preparation and Characterization of Al2O3/SiO2 Nanoparticles for Enhanced Photocatalytic Applications

Sofia Mendes[*1], Miguel Almeida[2], Ana Costa[3], and Rui Gomes[4]

[*1]Department of Chemical Engineering, University of Porto, Porto 4200-465, Portugal
[2]Department of Materials Engineering, University of Lisbon, Lisbon 1649-004, Portugal
[3]Institute of Nanotechnology, University of Coimbra, Coimbra 3000-300, Portugal
[4]Department of Physics, University of Aveiro, Aveiro 3810-193, Portugal

ABSTRACT

Nanoparticles Aluminum oxide (Al_2O_3)/ Silicon dioxide (SiO_2)photocatalyst was successfully synthesized by urea decomposition & ultrasonic assisted method using metal nitrate as precursors in the presence of sunlight. The as-synthesized samples were characterized by X-ray diffraction (XRD), Fourier transform infrared (FTIR), and UV-Vis spectroscopy. The x-ray diffraction pattern indicated that as-synthesized sample had a crystal size for $Al_2O_3$with finest particle size of the catalyst (30.096 nmappr.) was obtained at 600°C calcination temperature. Fourier transform infrared spectra confirmed the presence of hydroxyl group and Al-O bond vibration in the catalyst. Experimental result of the Al_2O_3/ SiO_2photocatalyst calcined at 600°C for 2hr, exhibited photocatalytic activity of under sunlight irradiation, the constants of malachite green dye degradation. In the present study, the synthesized nanoparticles used to for the degradation of the dye by direct sunlight exposer.

KEYWORDS: Al_2O_3/SiO_2 Nanoparticles, Photocatalytic.

I. INTRODUCTION

The synthetic dyes are toxic chemicals which can generate more colors and also hazardous to the environment [1]. In general, approx. 35–45% of these dyes remain inthe waste waters [2]. Presence of these dyes diminishes the photosynthesis and causes many serious health problems forhumanity [3]. To overcome these problems, the waste water from those industries must be treated before their discharge [4]. Variousphysical and chemical methods have been used for toxic removal from waste waters [5]. One of these methods is metal oxides photocatalysis and it has proven to be an effective in treating wastewater [6]. The search for low cost and efficient photocatalyst is still continuing. Among many organic pollutants, malachite green (MG) is one of pollutant color for environment undesirable which effects on aesthetic of environment [7]. The photocatalytic activity of Al_2O_3 can improved by the addition of SiO_2 which increase the available surface area of the catalyst, allowing an increase adsorption of polluted dyes [8]. The SiO_2 Surface turns improve the Al_2O_3/ SiO_2 photocatalytic activity compared to Al_2O_3[9] The properties of Al_2O_3/ SiO_2 mixed oxides are depends upon the preparations methods [10]. The preparation includes of urea decomposition method for Al_2O_3 and precipitation method for SiO_2. These two methods are very effective and conventional also the less time consuming.Al_2O_3/ SiO_2 mixed oxides exhibit promising photocatalytic activities due to their environmental friendly behavior, low catalyst cost, high specific surface area, high crystallinity and solar energy application and thus, could be an alternative material for environmental application and wastewater treatment [11]. Al_2O_3/ SiO_2 was used as photo catalyst under visible radiation for degradation of MG to get clean water.

II. MATERIALS AND METHODS

Synthesis of photocatalyst

Aluminum oxide (Al_2O_3)

The Al_2O_3, nanoparticles powder was prepared by Urea decomposition method. The Urea was corresponded to total volume ratio of metal nitrate, ratio of 1:2. In each case, aluminum nitrate dissolved in stoichiometric amounts of water, 10% then mixed with vigorous stirring at room temperature (55°C). The prepared slurry was left to stand for the formation of solid. After the solidification was completed, the solid was kept for 2 days at room temperature and sample was dried at 75°C for 36 h. After grinding the dried samples, they were calcined at 600°C for 2 h. Nano sized materials of the catalyst were analyzed.

Silicon dioxide (SiO_2)

Tetraethyl orthosilicate (TEOS) 0.1 M and Ammonia (NH_4) Solutions were mixed. The resultant solution immediately sonicated at different temperatures (according to experimental design) for 30 min using an

ultrasonic generator with the output power set to 200W with a frequency of 20 kHz.After sonication, to complete the reaction, the solution was maintained at the reaction temperature for 15 min.the final precipitate calcination at 600˚ C for 2 hours in muffle furnace.

Photocatalytic degradation studies.

Photocatalytic study of the synthesized Al_2O_3/ SiO_2, Nanoparticles mixer powder were evaluated by decolorization of malachite green (MG)dye in aqueous solution. The experiments were carried out in the presence of visible light irradiation without any catalyst (blank), with catalyst in dark and in the presence of Al_2O_3/ SiO_2, photo catalyst. The photocatalytic reaction carried out in the glass beaker with sunlight direct exposer for 5hr. Reaction was set up by adding 0.3 g of Al_2O_3/ SiO_2 Nanoparticlespowder into 100 mL of MG solution the suspension was magnetically stirred in dark for 20 min to obtain adsorption/ desorption equilibrium before irradiating the sunlight in the beaker. Then the was exposed to sunlight, 9 mL of the sample was withdrawn for 1 hrtime interval over irradiation. The suspension was centrifuged at 1000 rpm for 10 min and filtered to remove the catalyst particles before measuring absorbance. The absorbance of the clear solution was measured at a λmax of 660 nm for quantitative analysis. Percentage degradation of MG dye was calculated using the following relation:

$$\% \text{ degradation} = \frac{A_0 - A_t}{A_0} \times 100$$

Where A_0 is absorbance of dye at initial stage A_t is absorbance of dye at time t.

Characterization:

FTIR is carried out from our institute using Parkin Elmer FTIR instrument ranges from 400 cm^{-1} 4000 cm^{-1}. X-ray powder diffraction (XRD) analysis was carried out with Goniometer Ultima IV using a Cu Kα radiation (λ=1.54060Å) operating at 40 kV and 40 mA.Absorbance carried out by spectrophotometer.

III. RESULTS AND DISCUSSION

FTIR Studies:

Fourier transform infrared Spectroscopy (FTIR) was used identify vibrational modes of different functional group. In the figure.1 the silica spectra shown as can be observed. The peaks appearedat 412.08 cm^{-1}, 598.0 cm^{-1}& 412.08 cm^{-1}, which are related to vibrations of the O-Si-O bands, bending vibration of Si-O and antisymmetric stretching vibration of O-Si-O band respectively. also, the absorption peak at 3460$cm^{-1}$is due to water molecules and the peak at 1637 cm^{-1}belongs to the OH bending vibration.

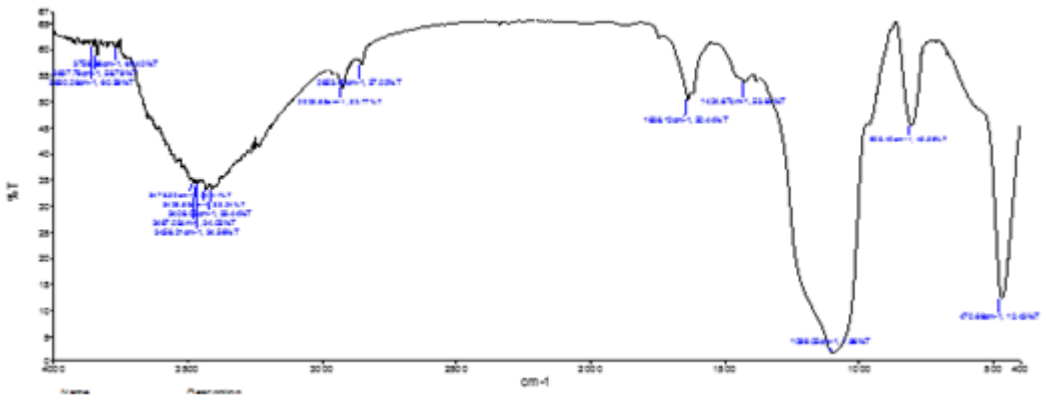

Figure 1. FTIR of SiO_2 nanoparticles

The reaction of aqueous solutions of urea with $Al(NO_3)_3$, produces clear white colored oxide, Al_2O_3. The formation of this oxide upon the heating of an aqueous mixture of $Al(NO_3)_3$. For the reaction mechanisms, an oxidation process for aluminum urea complex occurs during the decomposition of urea into ammonia, carbon dioxide and hydrogen chloride gases. The infrared spectra of synthetic oxide product are shown in Fig2. The

infrared spectra of the obtained products show bands due to characteristic groups of urea (carbonyl and amide groups) at 1102 cm^{-1} and 1629 cm^{-1}, the bands associated to the O-H are observed at 3429 cm^{-1} is due to moisture absorbed during measurement of spectra.

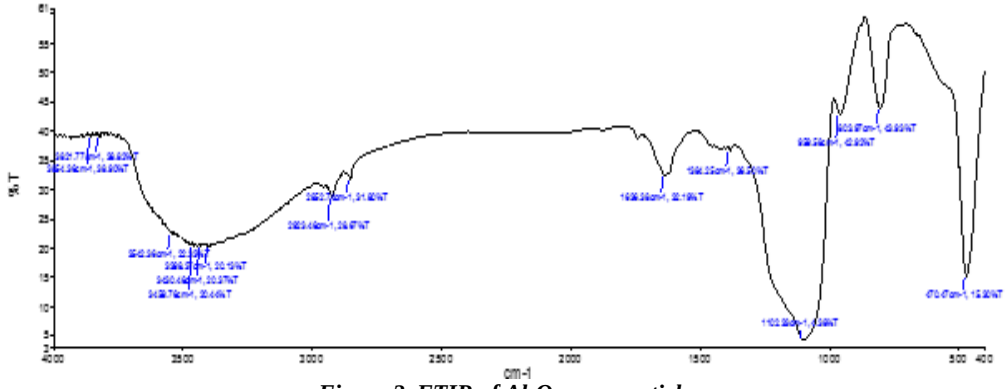

Figure 2. FTIR of Al_2O_3 nanoparticles

XRD Studies:

Silica NPs:The powder X-ray diffraction pattern of nanosilicaobtained by ultrasound assisted method shown in fig.3. the strong peak at angle 2 θ of 22º confirms the amorphous nature of silica. By using Scherrer's formula the particles size silica is found 15.9 nm approx.

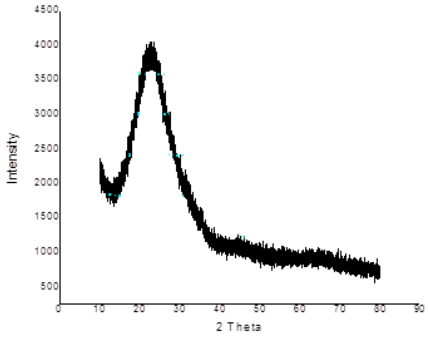

Figure 3. XRD of SiO_2 nanoparticles

Al_2O_3Nps: The phase formation and orientation of Al_2O_3nanoparticleswere investigated using X–ray diffraction in the ranges (20-80deg). X-ray diffraction patterns of nanoparticles with shown in figure 2. It was found that the presence of Al_2O_3 at temperatures 600ºC. The XRD results also reveal the structural results for work and the values obtainedusing the Scherrer equation: $D=k\lambda/\beta\cos\theta$ where D is the crystallite size, λ is the wavelength of the Cukα radiation, k is a constant equal to unity, β is corrected peak width at half maximum intensity and θ is peak position (68.69º used for all lines). Crystallite size of Al_2O_3-NPs increases. The decomposition process is highly affected by the molar ratio.

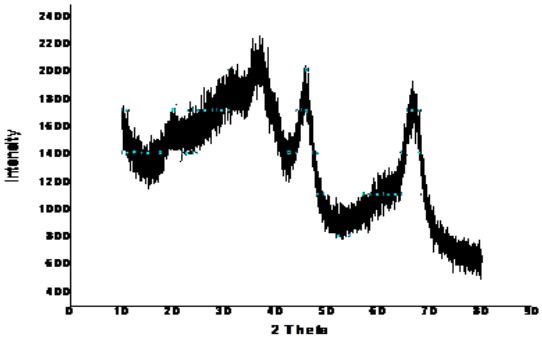

Figure 4. XRD of Al_2O_3 nanoparticles

Photocatalytic degradation studies:

The photocatalytic activity of Al_2O_3/ SiO_2nanoparticles was evaluated by the degradation of MG dye in aqueous solution. The decolorization of the MG dye was examined under three different conditions (treatments): sunlight

irradiation without any catalyst (blank solution), in the presence of catalyst without light irradiation (in dark) and in the presence of Al_2O_3/ SiO_2nanoparticles photo catalyst under sunlight irradiation, respectively. For the blank experiment (in the absence of the catalyst) under sunlight irradiation, almost insignificant degradation of the dye was observed. In the presence of Al_2O_3/ SiO_2nanoparticles. The formation of electrons-holes pairs are responsible for enhancing the oxidation and reduction reactions with the MG dye, which might be adsorbed on the surface of the Al_2O_3 nanoparticles to give the necessary products. The experimental results show that when the dye solution is exposed to sunlight irradiation for 5 hr in the presence of Al_2O_3/ SiO_2nanoparticles. The degradation of MG dye as function of time under sunlight irradiation in the presence of Al_2O_3/ SiO_2nanoparticles as shown in table 1. Accordingly, the degradation efficiency of MG dye under the sunlight was found to be much larger than the degradation efficiency as compare to blank and dark treatment. This enhancement under sunlight in the presence of Al_2O_3 nanoparticles, the first one could be the fact that the Al_2O_3/ SiO_2nanoparticles prepared by the urea decomposition method has a high specific surface area, that could give more active surface sites to absorb water molecules and to form active •OH and HOO• radicals by trapping the photo generated holes. This free active radical drive the photo degradation reactions and eventually leads to the decomposition.of organic pollutants in aqueous solution. Under sunlight irradiation, MG molecules are absorbed on the surfaces of nanoparticles and produced electrons. These electrons are captured by the surface adsorbed O2 molecules to yield O2•− and HO2• radicals, which makes more chance to touch with dye molecules and giving a faster reaction speed then, the MG molecules could be mineralized in time by the super oxide radical ions. Therefore, it can be concluded that the smaller crystalline size of nanoparticlesis favorable for the reduction of O2 and oxidation of H2O molecules by trapping electrons and holes, which improves the photocatalytic activity.

Table 1. Measurement of absorbance of suspension (Al_2O_3/ SiO_2 NPs and MG dye)

Time in hours	Absorbance Intensity (Approx.)
2	2.2
3	0.5
4	0.4
5	0.3

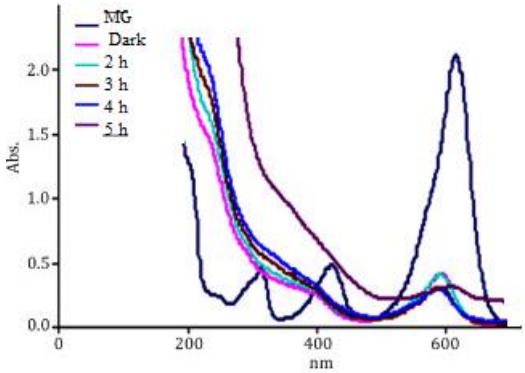

Figure 5. Graphical representation of Absorption under UV Visible

IV. CONCLUSION

The phase of Al_2O_3/ SiO_2 nanoparticles can successfully be synthesized by urea decomposition & ultrasonic assisted method using aluminum nitrate, at room temperature then the burnt product was calcined at 600^oC for 2h for Al_2O_3 NPs and ultrasonic assisted for silica Nps. The prepared sample was characterized by using different tools; FTIR, XRD, andUV. Al_2O_3/ SiO_2NPs with average crystallite size 30.096 nm & 15.9 nm approx.was obtained at 600^oC.The decomposition process is highly affected by the molar ratio. The produced Al_2O_3/ SiO_2 NPs showed photocatalytic activity by degradation of 85 % approx. of the MG dye, under sunlight irradiation, respectively, within 5 h. in overall studies it is concluded that the Al_2O_3/ SiO_2 NPs showed photocatalytic activity and it can be used as best degradation agent.

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VI. REFERENCES

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