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Metal Doped Magnetic Cobalt Ferrite Nanoparticles and Their Nanocomposites Photocatalyst for Degradation of Organic Dye Pollutants: Mini Review

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ABSTRACT

Day by day the increasing pollution is a big threat for the entire environment. The two main pollutions Viz. Air and aqua pollution is more rapidly expanding due to fast urbanization around the world. Although the researchers are working very hard to defeat pollution related problems, but the pollution scenario is becoming very complex issue for all the mankind. Particularly, water pollution is quite serious issue for flora and fauna. There are many methods have been developed so far to treat the industrial, chemical, pharmaceutical, drug and dyes industries pollutant, which discharged through the water stream around the coastal region. The discharged effluent is the major contributing factor for inception of water pollution. The researchers are developing very cheap and effective material to be utilized as catalyst for discrimination of the various pollutants in the form of dyes present under water stream. Mainly metal oxide based semiconducting material have gain more attraction in the field of photocatalysis due to their inherent properties such as tunable band gap, enhanced surface area, excellent thermal stability and good redox mechanism ability. The metal oxide based semiconductors such as Fe3O4, ZnO, CuO, SnO2, TiO2, Fe2O3, ZrO2, NiO, LaFeO3, NiFe2O4, CoFe2O4 etc. are most common and suitable catalysts used in the field of photocatalysis.

The present review is correlated with the utilization of cobalt oxide nanoparticles and nanocomposites in the field of photocatalysis. The CoFe2O4 being magnetic in nature and excellent thermal stability it is extensively as photocatalyst for degradation of common and azo based dyes. The present review elaborates the detailed methods of fabrication of CoFe2O4, their common characterization techniques and extensive utilization of cobalt ferrite in the field of photocatalysis.

Keywords: Magnetic CoFe2O4, Photocatalysis, environmental remediation, Dye degradation.

I. INTRODUCTION

From last few years, the photocatalytic degradation of organic dyes have gained more attention for the treatment of wastewater.[1] Broadly there are three techniques to remove dyes from wastewater that are physical, chemical and biological techniques. Physical processes includes Membrane filteration, adsorption method, Coagulation-flocculation technique, ion-exchange technique, etc while, biological processes make use of aerobic, anaerobic micro-organisms depending on it there are three major type of biological techniques aerobic treatment, anerobic treatment and both aerobic-anaerobic treatment.[2] Organic dyes are relatively stable as they have aromatic system in there structure that provides extra stability so they are not completely mineralized by physical and biochemical method.[3] However, in case of chemical processes "advanced oxidation processes" (AOP) heterogeneous photocatalysis technique have ability to mineralize most of organic dyes completely.[4] Worldwide per year beyond 1,00,000 dyes are commercially made available.[5] Dyes are frequently used in textile, paper, rubber, plastic, food processing, pharmaceutical, leather, cosmetic, etc industries in order to make the products colorful and attractive.[6] These industries discharges effluents that primarily contains dye in fresh water leading it into a wastewater. Organic dyes such as azo dyes, nitro dyes, indigoid dyes, anthraquinone dyes, phthalein dyes, triphenyl methyl dye, nitrated dyes are harmful, toxic and some of them carcinogenic. So it is necessary destroy and clean the wastewater from such toxic dyes.[7] Metal ferrites especially, cobalt ferrite is used to treat wastewater as it have astonishing perperties such as high chemical stability, reasonable heterogeneous catalytic activity, magnetic nature that makes its separation from reaction medium more feasible, high magnetic crystalline anisotropy ($\sim 10^6$ erg/cm^3), high coerecivity, it have good curie temperature 520°C, it have excellent mechanical hardness, low toxic.[8-11] $CoFe_2O_4$ nanoparticals are iron based semiconductors that are n-type and have reasonable band gap of 1.76 eV, but $CoFe_2O_4$ alone don't have ability to total mineralize the organic dyes.[12] Metal doped $CoFe_2O_4$ nanoparticles and their nanocomposites increased the heterogeneous photocatalytic activity drastically.[13] Photocatalytic degradation of rhodamine B (RhB) dye by $CoFe_2O_4$ is 73.0% while Mg doped $CoFe_2O_4$ have degradation efficiency around 99.5%. Band gap of Co-Cu nanoparticles band gap is 1.57eV while sm^{+3} substituted nanoferrites have lower band gap ~1.36eV that makes it more feasible towards degradation of organic dyes.[7]

II. EXPERIMENTAL

Cobalt ferrite, metal doped cobalt ferrite and their nanocomposites can be prepared by different methods such as Sol-gel method, Hydrothermal method, Co-precipitation method, Combustion method, etc. Every method have its own advantages. Preparation of cobalt ferrite and zinc doped cobalt ferrite by Co-precipitation method has been discussed below,

2.1. Materials:

99.9% pure form of reagents ordered from Merck brand chemicals that were used without any futher purification. $Zn(NO_3)_2{\cdot}6H_2O$ (Zinc nitrate hexahydrate), $Co(NO_3)_2{\cdot}6H_2O$ (Cobalt nitrate hexahydrate) and

$Fe(NO_3)_2{\cdot}9H_2O$ (Ferric (III) nitrate nonahydrate) were used as a precursor for zinc, cobalt and iron respectively. Sodium hydroxide pellets (NaOH), and double-distilled water are used as precipitating agent and solvent for the reaction respectively.[14]

2.2. Preparation:

Pure form of Zn doped CoFe2O4 nanoparticles were synthesized with the help of Co-precipitation technique. By taking the cobalt nitrate and ferric nitrate precursors as the ratio of 1:2 respectively. They were dissolved in 100 ml of distilled water and magnetic stirrer inserted into it so obtain homogeneity by constant and vigorous stirring. Drop by drop addition of (4 M) molarity, aqueous NaOH solution is done till the mixture turns into light brown color from colorless solution. Further, the mixture is allowed to stir vigorous for about 6hr. After the magnet stirrer is switch off and the mixture is kept as it for one night to settle down the precipitate in the bottom of the beaker. The precipitate than centrifused 3 times each 20 min. at 3000 rpm and washed by distilled water followed by ethanol till the pH becomes 7. The precipitate is dried on petri dish at temp 80°C using a hot air oven. At last the particles are calcined for 2 hr at 400°C in a muffule furnace. In same way Zn doped $CoFe_2O_4$ are synthesized by co-precipitation method here the only difference is cobalt nitrate wt% is reduced and corresponding wt% of zinc nitrate is added to the reaction medium.[14]

III. RESULTS AND DISCUSSION

Characterization and analysis of cobalt ferrite nanoparticles specially there doped form with metals and their nonacomposite is done by different techniques like X ray diffraction (XRD),[8] XPS,[9] EDAX,[20] FT-IR,[23] UV-Vis,[13] DRS,[16] SEM/TEM,vibrating sample magnetometer VSM,[19] high-resolution transmission electron microscopic (HRTEM),[20] electron paramagnetic resonance (EPR)[25] that indicates the presence of radicals that are useful for the photodegradation of organic dyes such as hydroxide radical ($OH^{\cdot}$) and superoxide radical ($O_2^{\cdot -}$). Among the discussed characterization techniques XRD technique is elaborated in detail below;

3.1 XRD analysis

XRD data was recorded to study the structural properties of syn- thesized MoO_3 nano-rods, $CoFe_2O_4$ Nps and their composite MoO_3/ $CoFe_2O_4$ by X-Ray diffractometer. The diffraction peaks MoO_3 appeared at two theta 25.85°, 29.2°, 35.4°, 41.8°, 43.11°, 45.3°, 46.6°, 48.8°, 50.13° , 51.9° , 53.2° , 56.1° , 57.9° , 61.6° , 67.06° and 68.9° belonging to reflections (210), (300), (310), (224), (320), (410), (404), (008), (500), (330), (420), (280), (334), (430), (610) and (524) respectively and corresponding to JCPDS No. 21–0569 as shown in Fig. 1 (a). The size of molybdenum trioxide was calculated to be 14.1 nm using well known Scherrer's formula given below:

$$\eta = \frac{0.9\lambda}{\beta \cos\theta} \quad - (1)$$

D is the crystallite size of nanomaterial, θ represents the Bragg's angle, λ is the wavelength of used X-rays (Cu Kα 1.5Å) and β is the full width at half maximum value.

The diffraction peaks of $CoFe_2O_4$ appeared at 31.04°, 36.50°, 44.47°, 48.46°, 55.6°, 64.9°, 71.02°, 75.1° and 77.36° with corresponding lattice planes (220), (222), (400), (331), (511), (531), (620), (622), and (444) in Fig. 1 (b). The peaks were matched with JCPDS card No. 22–1086. Extra peaks of iron oxide were also examined by the XRD data which are 50.08°, 59.07° and 68.9° belonging to reflections (024), (018) and (208) respectively matched with the JCPDS card No. 33–0664. The formation of secondary phase of Fe_2O_3 was formed due to the favour- able heating conditions during the synthesis of cobalt ferrite. The size of the cobalt ferrite nano-particles was calculated to be 7.2 nm using the Scherrer formula.

The formation of the nanocomposite $MoO_3/CoFe_2O_4$ was confirmed by XRD analysis as presented in Fig. 1 (c). The synthesized composite possesses characteristic reflections at (300), (204), (310), (410) and (424) with 2θ values of 29.3°, 31.0°, 36.8°, 44.6° and 59.0° respectively, for MoO_3 and matched with the JCPDS No. 21–0569. The characteristic reflections of cobalt ferrite at (222), (331), (511), (531) and (422) corresponding to 2θ values at 38.0°, 47.9°, 55.5°, 65.03° and 53.0° were observed and matched with JCPDS No. 22–1086.[8-9]

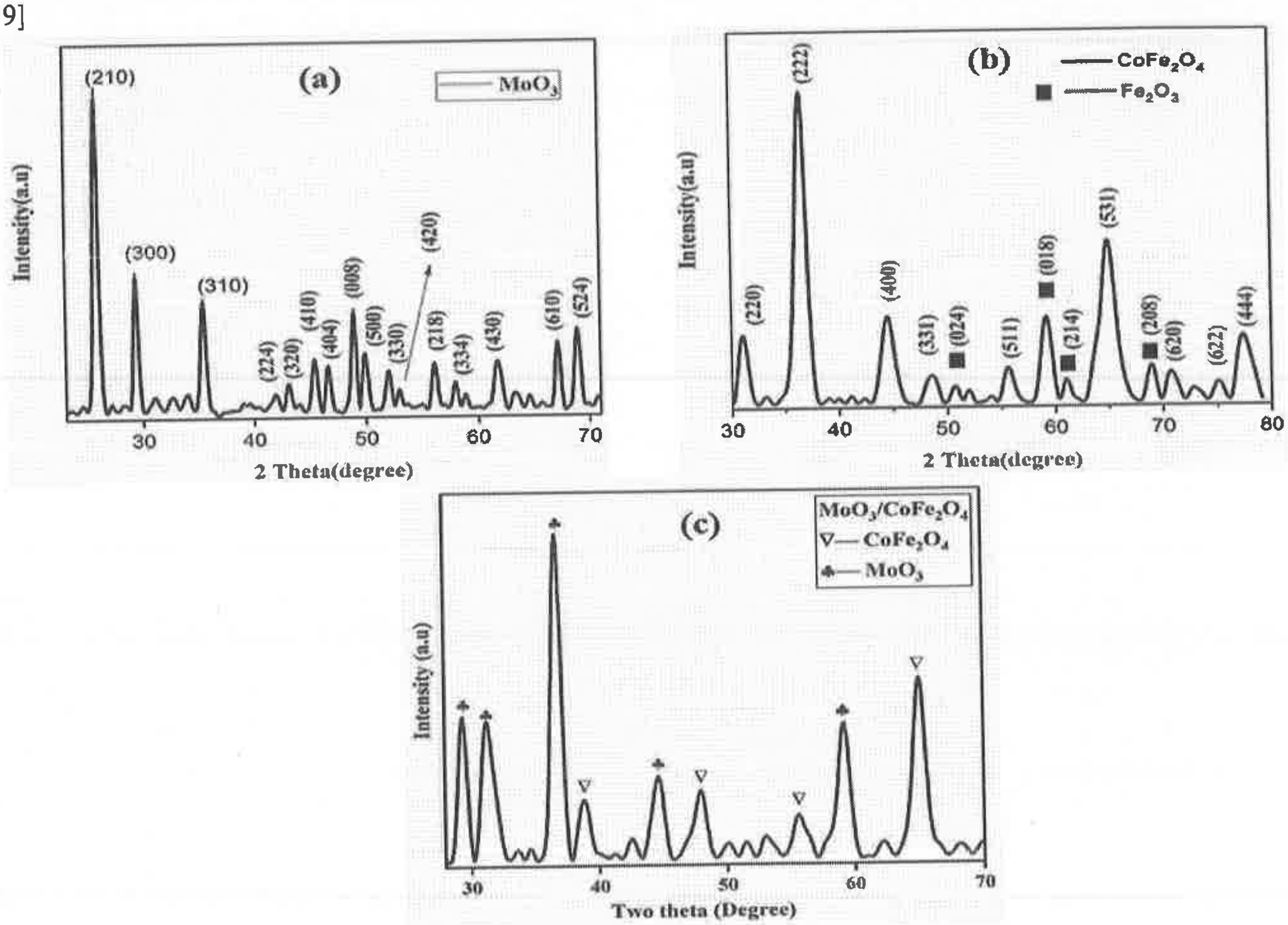

Fig. 1. XRD Diffractogram of (a) MoO_3 (b) $CoFe_2O_4$ and (c) MoO_3/ $CoFe_2O_4$ nanocomposites

Following table presents the list of Photocatalyst, Pollutant (Dye) Degradation efficiency in percentage Irradiation Source and Time;

Sr. No.	Photocatalyst	Pollutant	Effeciency (%)	Irradiation Source and Time		References
1.	$CoFe_2O_4$:Ni (Co-precipitation route)	Methylene Blue	83.41	90min.	Sunlight	[15]
2.	$CoFe_2O_4$ (Modified solvothermal route)	Methylene Blue	80	140min	Tungsten halide lamp (UV-Visible light source)	[16]
3.	Ru doped $CoFe_2O_4$ (Sol-gel method)	Remazol Deep red	-	30min	150 W Xe lamp as visible light source	[17]
4.	Al doped $CoFe_2O_4$ (Sol-gel method)	Methylene blue	93	120min	200 W visible light	[18]
5.	Mg doped $CoFe_2O_4$ (Microwave combustion method)	Rhodamine B	99.5	-	150 W halide lamp (Visible light)	[19]
6.	Sm substituted copper doped $CoFe_2O_4$	Rhodamine B	94.36	270min	Sunlight	[20]
7.	Rh loaded $CoFe_2O_4$	Malachite Green	97	60min.	Xe lamp (400 W)	[21]
8.	$CoFe_2O_4$ (Modified Solvothermal process)	Methylene blue	80	140min	Visible light	[22]
9.	Dy doped $CoFe_2O_4$ (Co-precipitation)	Methyl orange	78.65	2.0 hr.	Visible light	[23]
10.	$CoFe_2O_4$/ZnO (Co-precipitation method)	Acid violet Acid brown	76 63	-	UV lamp (32 W)	[24]
11.	Bi_2O_3/ $CoFe_2O_4$ (Hydrothermal route)	Methyl orange	92	-	300 W Xe arc lamp	[25]
12.	SnO_2-Tio_2/ $CoFe_2O_4$ (Sol gel method)	Rhodamine B	100	90 min	Sunlight	[26]
13.	ZnS-WO_3- $CoFe_2O_4$	Methylene Blue	95.97	180 min	Visible light radiation	[27]
14.	Li-Cr substituted $CoFe_2O_4$	Crystal violet	90.4	60 min	Sunlight	[28]

15.	$CoFe_2O_4/BaTiO_3$ (Sol gel method)	Methylene Blue	99.3	5 hours	Ultraviolet light radiation	[29]
16.	$MoO_3/CoFe_2O_4$ (Co-precipitation method)	Methylene Blue	91	-	Visible light radiation	[30]
17.	$CoFe_2O_4/Fe_2O_3$ (Hydrothermal Process)	Methyl Orange	93	5 hours	Ultraviolet light radiation	[31]
19.	Zn doped cobalt ferrite (Co-precipitation method)	Methylene Blue Rhodamine B Crystal Violet	97 83 91	90 min.	Sunlight	[33]
20.	$CoFe_2O_4$-CeO_2 (Hydrothermal method)	Orange II	98.5	60 min.	Visible light	[34]
21.	Mn doped $CoFe_2O_4$ (Sol-gel auto combustion)	Orange II	85.4	2 hours	Visible light	[35]
22.	$CoFe_2O_4$ (Sol-gel auto combustion)	Reactive Red 195	74	2 hours	UV lamp	[36]
23.	ZrO_2-TiO_2/ $CoFe_2O_4$ (Sol-gel method)	Rhodamine B	99.7	60 min.	UV light	[37]

Degradation of Remazol deep red dye was carried out under 150 W Xe lamp as visible light source where 50 mg catalyst was placed in 100 ml of 60 mg/L dye solution having pH=2.5 using H_2SO_4 addition 0.1ml of 30% H_2O_2 done for 30 min.[15]

Al doped cobalt ferrite a black colored photocatalyst minerize the methylene blue dye in 120 minutes. In 100ml beaker different concentration of catalysts were taken and MB dye of 10 mg/L in same reaction medium pH=11 was maintained.[19]

Rhodium B degradation by Mg doped cobalt ferrite is a fenton type rection in which addition of 30% H_2O_2 is done to generate $OH^{\cdot}$ various volume of dye and catalyst were mixed at constant pH=2 and the catalytic activity initiated in visible source of light and analyzed using UV-Visible spectrometer.[18]

IV. CONCLUSION

In this review, specifically metal doped cobalt ferrites and their nanocomposites that are responsible for the degradation of toxic and hazardous dyes, there comparative study has been carried out Zn doped cobalt ferrite, $CoFe_2O_4/BaTiO_3$ mineralize Methylene blue (MB) most efficiently 97% and 99.3% respectively, whereas, SnO_2-Tio_2/ $CoFe_2O_4$ and ZrO_2-TiO_2/ $CoFe_2O_4$ degrades Rhodamine B (Rh B) in presence of sunlight most efficiently. Orange II is phaotocatalytically destroyed by $CoFe_2O_4$-CeO_2 around 98.5%. There is requirement of a photocatalyst that will mineralize the mixture of dyes at a time with almost same efficiency and comparative less time.

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