

# Functional metal sulfides and selenides for the removal of hazardous dyes from Water



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## ABSTRACT

Water contamination by organic dyes, is among the most alarming threats to healthy green environment. Complete removal of organic dyes is necessary to make water healthy for drinking, cooking, and for other useful aspects. Recently use of nanotechnology for removing organic dyes, became fruitful because of high surface to volume ratio and adsorption properties. Among these materials, metal chalcogenides emerge as new class of active materials for water purification. In this review article, we gathered information related to sulfide and selenide based nanomaterials which include metal sulfides and selenides, their binary composites, and use of different capping agents and dopants for enhancing photocatalysis. We have discussed in detail, about adsorption power of different dyes, relative percentage degradation, reaction time and concentration.

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## 1. Introduction

Increasing global warming threatens the whole world, earth is facing uneven climatic changes in the form of droughts, heat waves, severe storms, rising seas and floods all over the world [1–3]. Major hazard to ecosystem is the discharge of different dyes from different sources (e.g., paper and pulp industries, textile industries [4], dye and dye intermediates industries, tanner, pharmaceutical industries, craft and bleaching industries), which are introducing huge amount of dyes in natural water as well as in waste water treatment systems [5]. Dyeing and fishing industries are producing almost 50–100 L wastewater/kg [6], Vinu et al. [7], stated that almost 10–15% of 700 thousand tons of 10 thousand different types of dyes are discharged, without any proper physico-chemical treatment in different water systems, and are increasing day by day [7]. More than one million ton of different kinds of organic dyes are produced worldwide annually, and there are more than 100,000 commercially available dyes [8–10]. Reactive dyes have very low fixation rate, hence they can be considered as major contributor towards water pollution. Fixation rate of a dye is influenced by the extent of hydrolysis [11], toxicity of dyes impart negative impact on environment, which in turn effect human health. The negative impact include both non-biodegradability, and high color absorbing property influencing continual decrease in aquatic life by blocking sunlight [12]. Their removal is necessary, because most of the dyes and their

breakdown products are toxic, carcinogenic or mutagenic [13]. Organic dyes causes irritation of respiratory tract, eyes, skin, asthma, sore throat and allergic contact dermatitis [14]. The persistent dyes will remain in the water for a long period of time, because they are non-biodegradable (only decolorize and slowly mineralized in anaerobic environment, and also resistant to light) [15]. Hence, azo dyes can be easily subjected to decolorization with the splitting of azo bonds under anaerobic conditions, in turn the breakdown products are susceptible to degradation under aerobic conditions [16]. Due to the toxicity associated with use of dyes, some dyes such as Sudan Orange G, Para red, Sudan Red 7B and Rhodamine B, are not allowed to use in foods according to the European Parliament and council directive, 94/36/EC, on colors for use in food stuffs (EFSA 2005) [17]. If the dose of methylene blue exceeds 5 mg/kg, it may cause serious serotonin toxicity [18]. The effect of common cosmetic dye on human lip fibroblast was investigated in a culture system. It was observed, that Rhodamine B above 25 µg/ml, significantly decrease the number of cells after 72 h culture. In a similar way, the cell number decreased after 48 h with 50 µg/ml [19]. The LD 50 for malachite green is 80 mg/kg for mouse. Malachite green causes an increase in liver DNA adducts, and also the lung adenomas [20]. Eventually, 20 mg of Congo red intake during gestation period in rats is known to produce hydronephrosis and hydrocephalus in almost 15% of rat offspring's [21]. Paper and textile industries introduced several methods for the removal of organic dyes, but traditional methods are not efficient in decolorizing hazardous dyes [22]. The wastewater treatments for dye removal can be categorized into 3 major types i.e. chemical [23], physical [24,25] and biological treatments [24,26–29].

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## 2. Dye removal techniques

Liquid phase adsorption is the most well-known process for dye removal from waste water. The adsorption is very efficient equilibrium separation process, if the sorbent is cheap in terms of cost, simplicity to design, flexibility and insensitivity to toxic pollutants. The decolorization process greatly depends on physico-chemical factors (e.g. interaction between sorbent and dye, reaction time, particle size of the catalyst, surface area of sorbent, temperature and pH) [30]. Another, widely used method to remove dyes from different water systems is the use of activated carbon [31,32], but use of activated carbon in removing dyes is somewhat critical because of its high cost and regeneration problems of carbon after complete decolorization of dye [2]. The real difficulty in regeneration is the desorption of adsorbed dye from surface of activated carbon. Additionally, several filtration techniques are widely used for dye removal including microfiltration, ultrafiltration, and sometimes reverse osmosis [33,34]. However, membrane filtration processes are associated with few disadvantages i.e. they can only be applied in small flow rates, require membrane to push the waste water flow through membrane filtration, and are costly as well. Biodegradation is introduced as an alternative to both physical and chemical process, several different kinds of viruses, bacteria and algae are used for this purpose, involving microbial degradation, fungal decolorization, bioremediation systems and adsorption by microbial biomass [35,36]. However, biodegradation is limited, because most of the chemicals are toxic and harmful for microorganisms themselves and decreases the number of active microorganisms [35].

Chemical treatment include coagulation and flocculation for the removal of dyes from different water systems. However, disadvantages associated with chemical treatment are, high cost of the coagulating and flocculating agents, pH dependence of some dyes for their removal and waste disposal after accumulation of concentrated sludge. Additionally, secondary pollutants may arise by excessive use of some chemicals. Chemical treatment involves, ion exchange process which is not efficient in removing several types of dyes, rate of reaction is limited by the rate of diffusion, and solvents used for regeneration are very expensive [26].

## 3. Photo catalytic degradation of dyes

Photocatalytic degradation of dyes is gaining much interest in the recent research, because of the advantages associated with it over the traditional methods [2,37,38], as it is considered to be the green environmental solution for better future environment [39], which mainly include oxidative decomposition and purification of waste water [40,41]. In the last few decades, several photocatalytic systems including metal chalcogenides emerged as efficient photocatalyst for photochemical degradation of organic dyes.

## 4. Metal sulfides for dye degradation

### 4.1. FeS<sub>2</sub> nanomaterials

Large surface area of catalyst is of prime concern for effective degradation of organic dyes, which is associated with small size of the catalyst used for degradation process. Nanomaterials possess properties, which are quite different than bulk material, different research groups have developed methods to synthesize FeS<sub>2</sub> nanomaterials. One such research group, reported the degradation of Rose Bengal (RB) with mesoporous FeS<sub>2</sub> nanoparticles, and were quite successful in evaluating sharp decrease in the absorption maxima at 546 nm for an aqueous solution of RB. Degradation was successfully carried out by exposing 20 ppm aqueous solution of RB dye containing FeS<sub>2</sub> nanoparticles to 100 W glass lamp. FeS<sub>2</sub> nanoparticles used were mesoporous in nature with an average diameter of 40.33 Å having high surface area of

49.53 m<sup>2</sup>/g which greatly enhance the adsorption of dye over catalyst which in turn enhances degradation process [42].

Dutta et al. [43], reported similar results for RB. However, additionally they also investigated the FeS<sub>2</sub> photocatalytic activity against methylene blue (MB). The enhanced photodegradation of dyes with FeS<sub>2</sub> nanoparticles was evaluated with the help of UV-spectrophotometer from 400 to 600 nm for RB (initial concentration  $6.6 \times 10^{-5}$  M) and 600–750 nm for MB (initial concentration  $6 \times 10^{-6}$  M), by exposing aqueous solutions to incandescent tungsten halogen lamp (200 W), sharp decrease in the absorption maxima for both dyes was observed with passage of irradiation time.

### 4.2. CoS nanomaterials

CoS are usually unstable and undergo aggregation soon after formation, hence no such report of using CoS for photodegradation of dyes appear until now. However, hybrid materials containing CoS are reported by one such group, comparing the degradation of MB with CoS and their hybrids by incorporating CoS in AIMCM-41. In UV-visible absorption spectra, CoS provide strong absorption edges at wavelength of 320 and 350 nm, while CoS/AIMCM-41 has strong absorption edges at 290 and 360 nm. The red shift in case of CoS/AIMCM-41 favors degradation of MB under the application of UV irradiation with respect to CoS sample. On the other hand, shifting of absorption edge from 320 nm for CoS bulk to 290 nm for CoS/AIMCM-41 was observed. This blue shift is ascribed to the decrease in particle size which enhances the photodegradation (Fig. 1). Almost, 34% of dye molecules were degraded after irradiating the sample for 70 min with CoS nanoparticles (average size 25 nm), while 90% of the dye molecules were decolorized and degraded with hybrid CoS/AIMCM-41 (average size 8 nm) after exposing 0.32 ppm of sample at pH 2–12, for 60 min of irradiation under the application of UV light. The optimum pH at which the highest degradation was observed is at 9, because surface of the catalyst is negatively charged in basic medium, which favors degradation process [44]. Another group reported degradation of methylene blue (MB) and methyl red (MR), by CoS nanoparticles doped PAN (Polyacrylonitrile) hybrid in a simple photochemical reactor. The hybrid provided a continual decrease in characteristic absorbance peaks i.e. 663 nm for MB and 430 nm for MR [45].

### 4.3. CuS nanomaterials

Naturally, copper sulfide exist in different phases, which can be easily synthesized by controlling molar ratios of copper and sulfur i.e. CuS (Covellite) dispersed particles [46], Cu<sub>7</sub>S<sub>4</sub>-CuS hexagonal plates [47], Cu<sub>9</sub>S<sub>5</sub> (Digenite) octahedron [48] and Cu<sub>2</sub>S (Chalcocite) [49]. Among

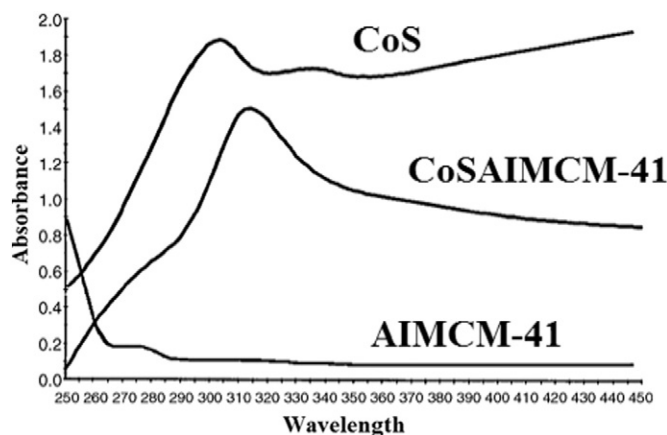


Fig. 1. Absorption spectra for the set of CoS samples. Modified with the permission of Sh. Sohrabnezhad et al. J. Hazard. Mater. 170 (2009) 184–190.

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