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Photocatalytic Degradation Study of Coralene Blue BGFS Azo Dye by Using Synthesized $SrAl_2O_4$ Nanoparticles and UV Light

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Abstract

Photocatalysis is an emerging method in dye degradation in wastewater treatment, which provides an alternative for energy saving technique by using renewable solar energy and UV light. The $SrAl_2O_4$ nanoparticle as photocatalyst was synthesized by solution combustion method with acetamide as fuel. These nanoparticles were characterized by XRD, SEM, and UV-absorption spectroscopy. Under artificial UV light, the photocatalytic activity of the synthesized nanoparticles was employed to degrade Coralene Blue BGFS azo dye by altering parameters such as dye pH, dye concentration, and catalyst concentration. The maximum photocatalytic degradation of Coralene Blue BGFS was recorded 84.27% at acidic condition (pH 4).

Keywords: Coralene Blue BGFS, $SrAl_2O_4$, Nanoparticle, UV light, Photocatalytic Activity

Introduction

The azo dyes are always found with one or more azo bond linkage (-N=N-) which is used regularly about 70% out of 2,000 species of synthetic dyes (Gopalappa *et al.*, 2012; Madhusudhana *et al.*, 2012). The overuse of more than 70% use among other industrial dyes with applications in paint, paper and most especially textile industries (Oyetade *et al.*, 2022). Despite their vast use, azo dyes remain an industrial dye of global threat and difficulty in the treatment of their corresponding effluents. The degradation of azo dyes in industrial wastewaters has therefore received increasing attention. Traditional physical techniques (adsorption on activated carbon, ultra-filtration, reverse osmosis, coagulation by chemical agents, ion exchange on synthetic adsorbent resins, etc.) have been used for the removal of dye pollutants (Krutzler *et al.*, 1999; Mai *et al.*, 2002). These methods only succeed in transferring organic compounds from water to another phase, thus creating secondary pollution. This will require a further treatment of solid-wastes and regeneration of the adsorbent which will add more cost to the process (Mailhot *et al.*, 2000). AOPs are promising methods for the remediation of wastewaters containing azo dye traces like coloring matters and disrupting chemicals. Advanced oxidation process is a set of processes involving the production of very reactive oxygen species able to destroy a wide range of organic compounds (Munter 2001).

Photocatalysis is a process of using energy from light to excite nanoparticles to degrade dyes and produces less harmful by products. These substances are mostly semiconductors. In recent years the usage of photocatalysts has increased due to their potential applications to remove the environmental pollutants. Recently $MgZnAl_2O_5$ nanoparticles have been used as effectively used under UV light to degrade coralene yellow 7GD and found to be very effective in treating the coloured solution (Madhusudhana *et al.*, 2022). Hence, the current research work is mainly focused on the development of efficient photocatalysts to degrade the pollutants from the environment.

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Materials and Methods

Materials and Reagents

The chemicals used for the synthesis of strontium aluminate ($SrAl_2O_4$) nanoparticles are [Strontium Nitrate $Sr(NO_3)_2$ (99.0% AR) from Loba Chemie Pvt. Ltd, Palghar and Aluminiun nitrate $Al(NO_3)_3$ (99.0% AR) and fuel Acetamide $CH_3.CO.NH_2$ (99.5%) from Thomas Baker Pvt. Ltd, Mumbai]. The Coralene blue BGFS dye used for the photocatalytic study was purchased from Colourtex Limited, Surat, Gujarat. The structure of the dye is given in Figure 1.

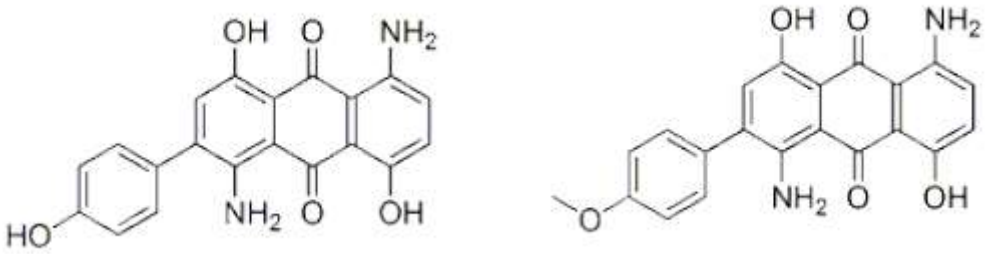

Figure 1: Molecular Structure of Coralene Blue BGFS dye

Synthesis of $SrAl_2O_4$

The nanoparticles were prepared by using solution combustion method. A mixture of stoichiometric amounts of Strontium nitrate (23.27gm), Aluminum nitrate (82.52gm), and fuel Acetamide (23.62gm) was dissolved in a minimum quantity of water in a silica crucible (100 mm size). The mixture solution was introduced into the muffle furnace which was preheated at 600°C. The solution undergoes dehydration and catches fire by spreading throughout the mass, finally yielding $SrAl_2O_4$ nanoparticles. The mixture then froths and swells, forming foam which ruptures with a flame and glows to in candescence. The obtained $SrAl_2O_4$ was crushed in a mortar to make the $SrAl_2O_4$ amorphous. Thus, $SrAl_2O_4$ is formed. According to propellant chemistry the reaction is as shown (Madhusudhana *et al.*, 2012):

$$Sr(NO_3)_2 + 22Al\,(NO_3)_2 + 40\,CH_3CONH_2 \rightarrow 11\,SrAl_2O_4 + 80CO_2 + 100H_2O + 64N_2$$

Figure 2: Synthesis of nanoparticles

Synthesis of $SrAl_2O_4$

X-ray diffraction is one of the most important characterization tools used in solid state chemistry and materials science. XRD is an easy tool to determine the size and the shape of the unit cell for any compound X-ray diffraction is a versatile, non-destructive that reveals detailed information about the chemical composition and crystallographic structure of natural and manufactured materials. XRD was performed by Rigaku diffractometer using Cu-Kα radiation (1.5406 Å) in a θ-2θ configuration (Girase *et al*., 2011). According to the Debye Scherrer's formula:

Debye Scherrer's formula $D = K\lambda/\beta\cos\theta$ (Eq. 1)

Where D = Thickness of the crystallite K = 0.94 the Scherrer's constant (dependent on crystalline shape) λ = X-ray wavelength β = the peak width at half-maximum (FWHM) θ = the Bragg diffraction angle

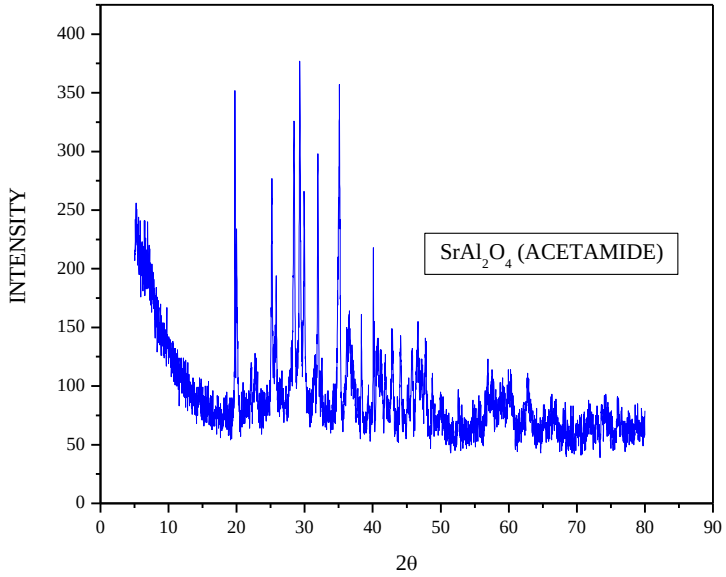

Figure 3: X-ray Diffraction of $SrAl_2O_4$ (Acetamide)

The powdered sample of $SrAl_2O_4$ (Acetamide) nanoparticle was examined by XRD and analysis was carried out on fresh sample to assess the purity of the expected phases and the degree of crystallization i.e., size, composition and crystal structure. XRD was performed by Rigaku diffractometer using Cu-Kα radiation (1.5406 Å) in a θ-2θ configuration. According to the XRD the average crystallite size of $SrAl_2O_4$ (Acetamide) was found to be 51nm.

SEM Study of $SrAl_2O_4$

Illustrates SEM photographs of single crystals of $SrAl_2O_4$ (Acetamide). The photographs revealed combination of cluster, Sharpe edge, and plate like structure and tube like structure morphology which looks like a colony. The enlarged image shows the uneven size and shape of the different nano-particles, which also revels the thick attachment of nanoparticles over one another.

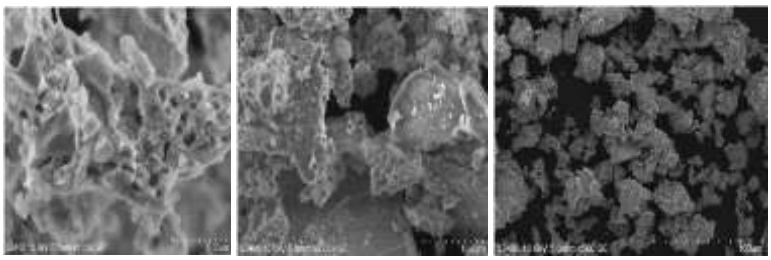

Figure 4: SEM photographs of $SrAl_2O_4$ nanoparticles

UV Vis Spectroscopy

The optical absorption is a significant tool to get optical energy band gap of crystalline and amorphous materials. The elemental absorption corresponds to the electron jump from valence band to the conductivity band. The spectrum reveals that, the Strontium aluminate nanoparticles absorption in the visible radiation with an above wavelength 400 nm. The value of optical band gap (OBG) is calculated from the TAUC's relation.

$$[\alpha h\upsilon] = B[h\upsilon - Eg]^n$$

Where, 'hυ' is the photon energy, 'B' is the constant and 'n' is the power factor and that takes 1/2, 2, 3/2 and 3 allowed direct, allowed indirect, forbidden direct and forbidden indirect transitions respectively. The OBG of the Strontium aluminate nanoparticle found to be 3.27eV.

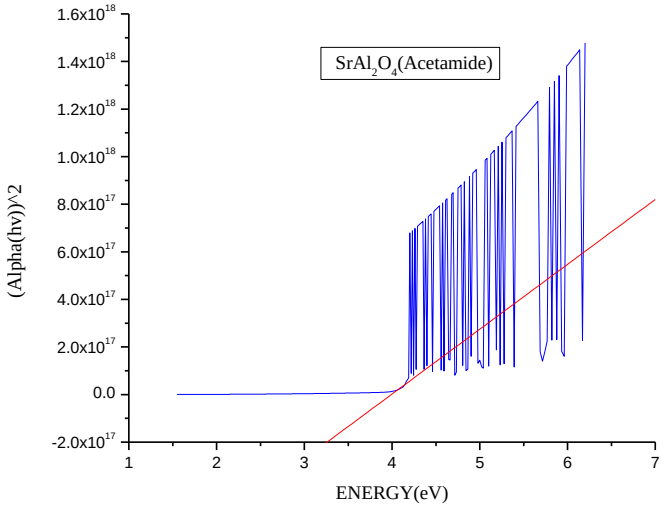

Figure 5: UV absorption spectra of $SrAl_2O_4$ (Acetamide)

Results and Discussion

Experimental Procedure

Initially, photocatalytic experiments were carried out in the presence of UV light of wave length 254nm. The UV-1900i spectrophotometer was used for the determination of absorbance in the range of 200 to 800nm. The λ_{max} of Coralene Blue BGFS was found to be 526 nm. In all photocatalytic experiments, 100ml of 50ppm Coralene Blue BGFS aqueous solution was taken in 100ml borosil beakers. A known concentration of synthesized $SrAl_2O_4$ (Acetamide) was added to test the photocatalytic activity in presence of UV light. Further experiments were conducted based on the degradation results obtained from the photocatalytic activity of the catalysts.

Effect of Catalyst Concentration on Coralene Blue BGFS using $SrAl_2O_4$ (Acetamide) Nanoparticles

The effect of catalyst concentration on the photocatalytic degradation was studied over a range of the catalyst amount from 0.1 to 1.5g/100ml for Coralene Blue BGFS. Solution. The synthesized nanoparticle has shown an appreciable result. Where, $SrAl_2O_4$ (Acetamide) of average size 51nm showed maximum of 81.85% at 0.5g/100ml, in 300 minutes.

The increase in degradation rate can be explained in terms of availability of active sites on the catalyst surface and UV light penetration into the suspension as a result of increased

screening effect and scattering of light. A further increase in the catalyst amount beyond the optimum dosage for all the nanoparticles decreases the photodegradation by some margin. This may be due to overlapping of adsorption sites as a result of overcrowding owing to collision with ground state catalyst (Santhosh *et al*., 2020a; Santhosh *et al.*, 2020b). Since, the photodegradation was most effective at 81.85% at 0.5g/100ml for $SrAl_2O_4$ (Acetamide) nanoparticle dosages, further experiments were continued with same dosages.

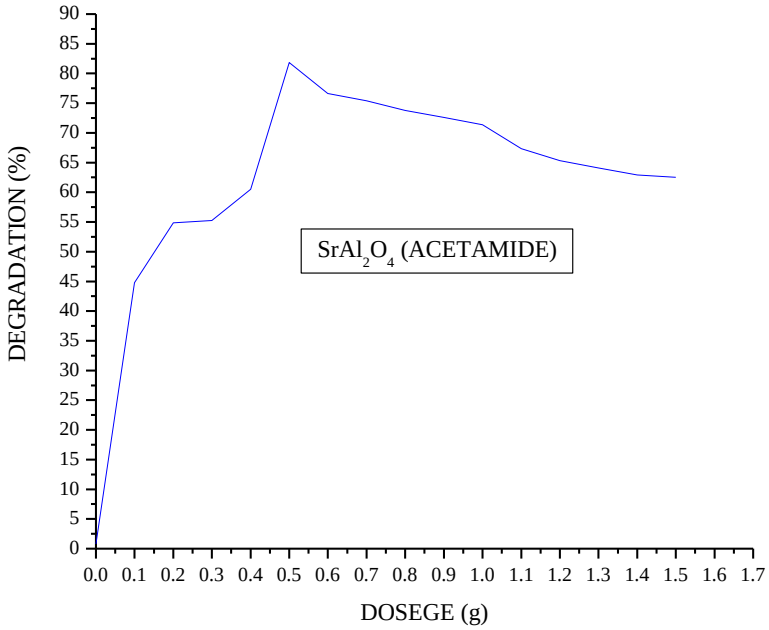

Figure 6: Effect of catalyst concentration on Coralene Blue BGFS at 300 minutes of 50ppm/L, pH=7 $SrAl_2O_4$ (Acetamide)

Photo 1: Effect of catalyst concentration on Coralene Blue BGFS at 300 minutes of 50ppm/L, pH=7 $SrAl_2O_4$ (Acetamide).

Mechanism of the Photocatalytic Degradation

Nanoparticles + UV → (e^-_{CB} + h^+_{VB}) (Eq. 2)

$e^-_{CB} + O_2 \rightarrow O_2^{\bullet -}$ (Eq. 3)

$H_2O + O_2^{\bullet -} \rightarrow OOH^{\bullet} + OH^-$ (Eq. 4)

$2OOH^{\bullet} \rightarrow O_2 + H_2O_2$ (Eq. 5)

$O_2^{\bullet -}$ + Coralene Blue BGFS → Coralene Blue BGFS -$OO^{\bullet}$ (Eq. 6)

$O\ OH^{\bullet} + H_2O + e^-_{CB} \rightarrow H_2O_2 + OH^-$ (Eq. 7)

$H_2O_2 + e^-_{CB} \rightarrow OH^{\bullet} + OH^-$ (Eq. 8)

$H_2O_2 + O_2^{\bullet -} \rightarrow OH^{\bullet} + OH^- + O_2$ (Eq. 9)

$OH^{\bullet}/O_2^{\bullet -}$/Nano-particles$^{\bullet +}$ + Coralene Blue BGFS →
→ Coralene Blue BGFS degradation (Eq. 10)

The mechanism of photocatalytic activity of nanoparticles can be predicted. Under sunlight irradiation, nanoparticle molecules get excited and transfer electrons to the conduction band (Eq. 2). An electron in the conduction band of the nanoparticles can reduce molecular oxygen and produce the super oxide radical (Eq. 3). Molecular oxygen, adsorbed on the surface of the photocatalysts prevents the hole-electron pair recombination process (Di-Paola *et al*., 2003). Recombination of hole-electron pair decreases the rate of photocatalytic degradation. This radical may form hydrogen peroxide or organic peroxide in the presence of oxygen and organic molecule (Eq. 4, 5, 6). Hydrogen peroxide can be generated in another path (Eq. 7). Hydrogen peroxide can form hydroxyl radicals which are powerful oxidizing agents (Eq. 8, 9). The radicals produced are capable of attacking dye molecules and degrade them (Eq. 10).

Effect of pH

In order to study the effect of pH on the degradation efficiency of $SrAl_2O_4$ (Acetamide) as catalysts, the experiments were carried out at pH ranging from 2 to11. The results showed that pH significantly affected the degradation efficiency.

The degradation rate of Coralene Blue BGFS for $SrAl_2O_4$ (Acetamide) increased from 72.58% to 84.27% from pH 2 to pH 4 and decreased to 72.58% at pH 11 in 300 minutes for 0.5g/100ml. The maximum degradation rate for nanoparticles was achieved at pH 4. As the generation of OH radical is more in acidic medium maximum degradation was achieved at pH 4. These OH radicals are the main oxidizing species responsible for photocatalytic degradation. On greater than optimum pH, the decrease in degradation efficiency can be explained on the basis of the amphoteric nature of the catalysts. Here the catalyst surface becomes negatively charged for higher pH value, which causes the electrostatic repulsion between the catalyst and negatively charged dyes (Turchi and Ollis, 1990).

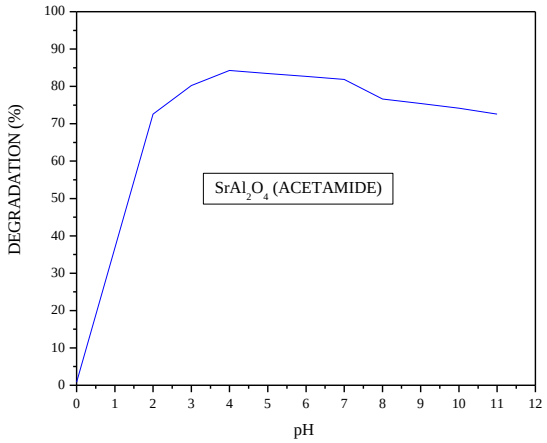

Figure 7: Effect of pH on Coralene Blue BGFS at 300 minutes 50ppm/L, pH=7, $SrAl_2O_4$ (Acetamide)

Photo 2: Effect of pH on Coralene Blue BGFS at 300 minutes 50ppm/L, pH=7, $SrAl_2O_4$ (Acetamide)

Effect of Initial Dye Concentration

The experiments were conducted to study the effect of initial dye concentration by varying the Coralene Blue BGFS concentration from 50, 60, 70 & 80ppm. The results obtained for $SrAl_2O_4$ (Acetamide) 50ppm was 84.27%, 60ppm was 79.36%, 70ppm 73.35% and 80ppm was 69.90%. These experiments illustrated that the degradation efficiency was directly affected by the concentration. The decrease in the degradation with an increase in dye concentration was ascribed to the equilibrium adsorption of dye on the catalyst surface which results in a decrease in the active sites. This phenomenon results in the lower formation of $OH^{\cdot}$ radicals which were considered as primary oxidizing agents of the organic dye. According to Beer Lambert law, as the initial dye concentration increases, the path length of photons entering the solution decreases. This results in the lower photon absorption of the catalyst particles, and consequently decrease photocatalytic reaction rate (Byrappa *et al*., 2006; Madhusudhana *et al.*, 2018).

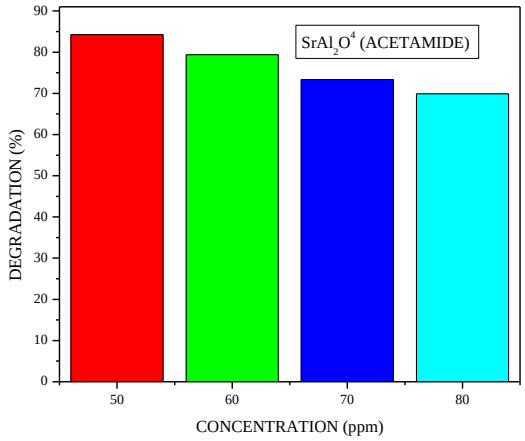

Figure 8: Effect of initial dye concentration on the photocatalytic degradation of Coralene Blue BGFS by $SrAl_2O_4$ (Acetamide) 50, 60, 70 & 80ppm pH=0.5g/4

Photo 3: Effect of initial dye concentration on the photocatalytic degradation of Coralene Blue BGFS by $SrAl_2O_4$ (Acetamide) 50, 60, 70 & 80ppm pH=0.5g/4

Effect of UV Light Irradiation on Coralene Blue BGFS

UV light irradiation generates the photons required for the electron transfer from the valence band to the conduction band of a semiconductor photocatalyst. The energy of a photon is related to its wavelength and the overall energy input to a photocatalytic process is dependent on the light intensity. Therefore, the effects of both intensity and wavelength are important. In the present study, the effect of the UV light intensity was studied by keeping the constant wavelength (526 nm).

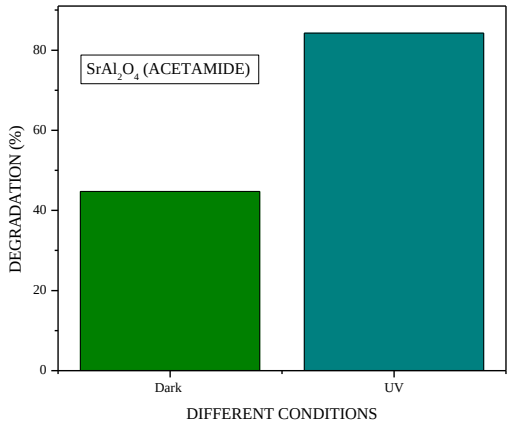

Figure 9: Effect of light source on the photocatalytic degradation of Coralene Blue BGFS for $SrAl_2O_4$ at 300 minutes

Photo 4: Effect of light source on the photocatalytic degradation of Coralene Blue BGFS by $SrAl_2O_4$ (Acetamide) at 300 minutes. 50ppm, pH=0.5g/4

These results clearly showed that, photodegradation occurs more efficiently in presence of UV light. Under UV light excitation of catalysts takes place rapidly than in absence of light. The experiment demonstrated that, both UV light and photocatalyst are needed for the effective destruction of Coralene BGFS as it has been established that the photocatalytic degradation of organic matter in the dye solution is initiated by the photo excitation of the semiconductor, followed by the formation of electron hole pair on the surface of the catalyst (Madhusudhana *et al.,* 2017; Madhusudhana *et al.,* 2011).

Conclusion

In the present study, the $SrAl_2O_4$ nanoparticles were prepared by solution combustion method using acetamide as a fuel and the result revealed that, the average particle size was found to be 51nm and band gap was found to be 3.27eV and these nanoparticles can be utilized in industries and medical applications. The photocatalytic activity of synthesized nanoparticles was used to degrade the Coralene Bule BGFS under artificial UV light and by varying the parameters such as, pH of the dye, dye concentration and catalyst concentration. The optimal catalyst concentration was found to be 0.5g/100ml with dye concentration 50ppm and pH 4 was maintained by 0.1 N HCl and NaOH. The maximum degradation was found to be 84.27% in 300 min at pH 4. The method of photocatalytic degradation of Coralene Blue BGFS is more helpful in treating Coralene Blue BGFS dye containing textile effluents. This method does not cause any harm to the environment and easily adoptable dye degradation technique under UV light and this can be applicable in large scale.

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