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Photocatalytic Effect of Nano BaO on Brilliant Green-a Cationic Dye

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Abstract **- The photocatalytic characteristics of BaO metal oxide nanoparticles (Nps) were well studied against Brilliant Green dye and discussed. Solution combustion method was employed in the synthesis of BaO NPs. The average particle size of BaO NPs was analyzed with SEM and XRD methods. Effect of various parameters like diverse catalyst dosage, pH, dye concentration and different irradiations implied in the photo degradation of Brilliant Green (BG) dye were studied. The BaO NPs proved to be remarkable and 97.25% of colored solution of Brilliant Green dye degraded at a pH 4 and catalyst dosage of 0.8g/100 ml. The BaO nanoparticles proved to be very effective in photo-catalyzing Brilliant green and cost effective.**

Index terms **- Brilliant Green, BaO, Nanoparticles (NPs), Photo degradation Introduction**

I.INTRODUCTION

Industries such as textile, cosmetic, plastic, paper and food use dyes as coloring agents [1]. "One of the major source of water contamination are textile industries and partially treated effluent containing chemicals are released to the receiving water bodies" [2]. Gallons of effluent from textile mills discharged into the water bodies are hazardous. These chemicals are full of color and synthetic chemicals produced as byproduct of dyeing. Presence of naphtha, nitrates, soaps, compounds and heavy metals and certain solvents, separation agents, or dispersing agents make the effluent highly toxic [3].

"The advent of a new class of processes known as advanced oxidation processes (AOPs) has resulted from research and development in the field of color removal, particularly in the form of heterogeneous photocatalysis, in which light energy is turned into chemical energy" [4]. "Due to its visible character, photocatalysis is not only an interesting instrument for energy conversion but also for environmental purification" [5]. A large degree of aromaticity present in modern dye molecules increase their stability and conventional biological treatments are ineffective for decolorization [6-7]. "The photocatalysis processes can breakdown a large variety of organic compounds to CO_2, water and mineral salts as the degradation products" [8]. Brilliant green (BG) is a triphenylmethane-based dye that has a wide range of applications in leather, textile, and biological sectors [9]. However, BG polluted water has a negative impact on people causing hypertension, heart, lung, and kidney problems, as well as carcinogenic to living organisms [10]. In this research study, Barium Oxide (BaO) nanoparticles are used as photocatalysts against Brilliant green dye. Effect of various parameters like diverse catalyst dosage, pH, dye concentration and different irradiations implied in the photo degradation of Brilliant Green (BG) dye were also studied.

II. CHEMICALS AND MATERIALS

A. Chemicals

Brilliant green (λmax 623nm-625nm) Fig.1 (a) and (b) was obtained from Sigma Aldrich, Mumbai, India. Chemicals like, Barium Nitrate ($Ba(NO_3)_2$ (99% A. R.), Urea (NH_2CONH_2) (99.5%), HCl (LR, 35.0%) and NaOH (LR, ≥ 97%), Buffer tablets of pH 4, pH 7.2 and pH 9 were purchased from Hi-Media Chemicals, Mumbai.

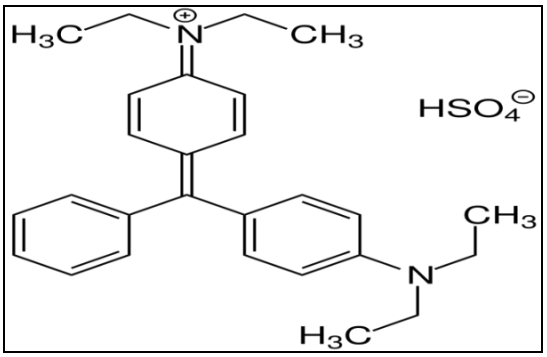

Fig.1. (a) Structural formula of Brilliant green (BG).

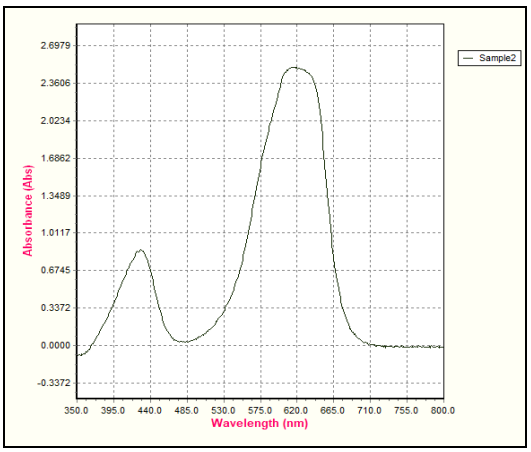

Fig. 1. (b) Lambda Max of Brilliant Green

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B. Materials

The λmax and relative absorbance of the dye solution with respect to time was determined by UV-VIS single beam spectrophotometer-119 (Systronics) [11]. The pH tester (J lab pH Meter 0 to 14 pH Range) was procured from Eutech Instrument, Mumbai. The digital lux meter TES 1332A (TES, Taiwan). Muffle furnace (Labline-11000 C) was used for the preparation of photocatalysts.

III. NANOPARTICLES SYNTHESIS

Heterogeneous photo catalysis concentrates on the dissociation of dyes into simpler molecules of CO_2, H_2O and mineral acids by using metal oxide nanoparticles as catalysts [12-14]. The calculated amount of Barium Nitrate, $Ba(NO_3)_2$ (15.67g) was dissolved in a silica crucible (100 cm3) with a minimum amount of water and Urea (NH_2CONH_2) (6g). The mixture was transferred to a muffle furnace that had been heated to 600°C. Initially, the solution boils and dehydrates, followed by breakdown and the release of gases (N_2 and CO_2) [15]. The residue is formed when the solution combination burns. The gases produced not only produce tiny metal oxide particles, but also aid to disperse the heat that prevents the product from sintering. As a result, the combustion process was finished in a few of minutes [16]. The redox mixture technique (urea) combustion reaction for the production of BaO may be expressed as:

$$3Ba(NO_3)_2 + 5NH_2CONH_2 \rightarrow$$

$$3BaO + 5CO_2 + 10H_2O + 8N_2 \qquad (1)$$

IV. CHARACTERIZATION OF BAO NANOPARTICLES

Barium oxide is an ionic compound having a chemical formula BaO. It is a non-flammable white hygroscopic chemical. BaO nanoparticles have the following properties:

A. XRD Studies of BaO

TABLE 1. THE COMPUTED VALUES OF XRD PARAMETERS OF BAO

Nano particle	D(nm)	d (Å)	a (Å)	δ (1016) (lines/m2)	V (Å)3
BaO	6.0469	2.734	9.025	2.734	735.091

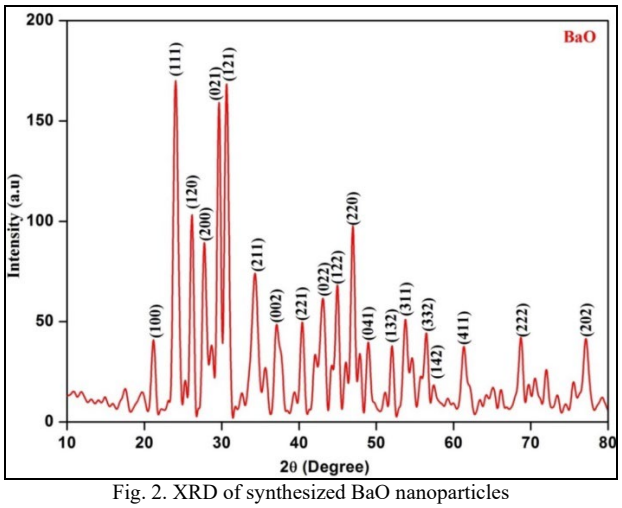

Fig. 2. XRD of synthesized BaO nanoparticles

Debye-Scherrer formula,

$$\text{average crystalline size, } D = \frac{0.9\,\lambda}{\beta \cos\theta} \qquad (2)$$

d-spacing values were calculated using X-ray diffraction peaks by the following relation,

$$d_{hkl} = \frac{\lambda}{2 \sin\theta} \qquad (3)$$

Dislocation density was determined using Williamson-Smallman relation,

$$\delta = \frac{1}{D^2} \qquad (4)$$

Table 1. shows the XRD patterns recorded for the barium oxide specimens. In Fig.2 the XRD peaks occur at 2θ =21.21, 23.9, 26.12, 27.7, 29.7, 30.5, 33.8 and 47.2 with the hkl values (1 0 0), (1 1 1), (1 2 0), (2 0 0), (0 2 1), (1 2 1) (2 1 1) and (2 2 0) respectively. It resembles the presence of body centered structure of BaO nanoparticles with the lattice parameter a=b=9.025A˚ and c=6.508A˚. The obtained values are in agreement with the (JCPDS File No: 26-0178). The presence of BaO phases in the present specimens with tetragonal crystal structures is therefore confirmed by the comparison. The pattern has been indexed and the 'd' values calculated for the lines. A comparison of the variables is presented in table 1. XRD study reveals that the average crystallite size achieved was at 6.04 nm and crystallinity of 98.21%.

B. Sem

Scanning Electron Microscope pictures of BaO nanoparticles have shown scattered crystals with irregular shapes. The magnified images also have shown a sharp edged, uneven, an irregular texture and are tightly bonded to one another [17] (Fig. 3 (a) (b) and (c)).

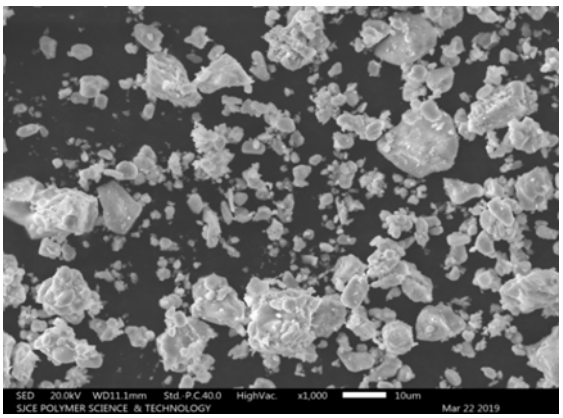

Fig. 3. Scanning Electron Micrographs of synthesized BaO Nanoparticles (a)

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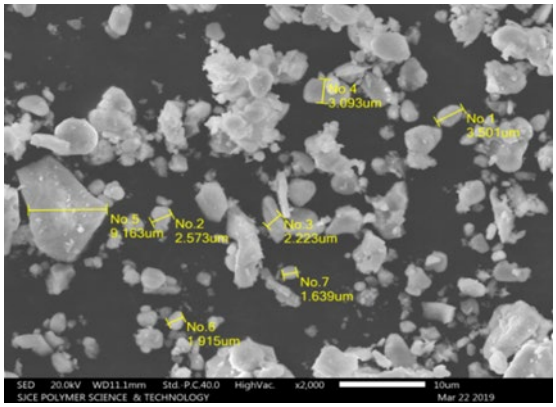

Fig. 3. Scanning Electron Micrographs of synthesized BaO Nanoparticles (b)

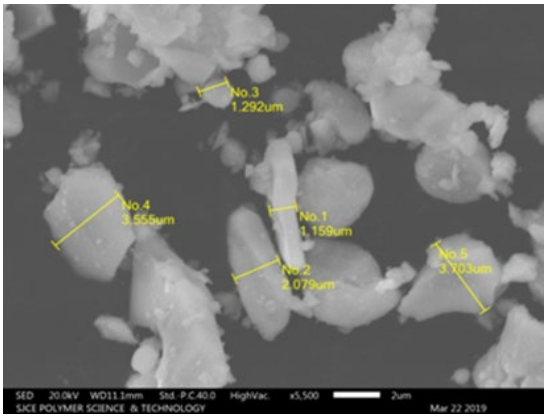

Fig. 3. Scanning Electron Micrographs of synthesized BaO Nanoparticles (c)

C. UV-Vis Spectroscopy

The use of optical absorption to determine the optical band gap is a useful tool (OBG) of the nanomaterials (Fig. 4). The elemental absorption of photons is to excite the electrons from valence band to the conductivity band. The spectrum reveals that, the BaO nanoparticles absorb radiations more in visible region beyond 350 nm [18]. The value of OBG is calculated from the TAUC's relation. OBG of the BaO nanoparticle is found to be 3.63eV.

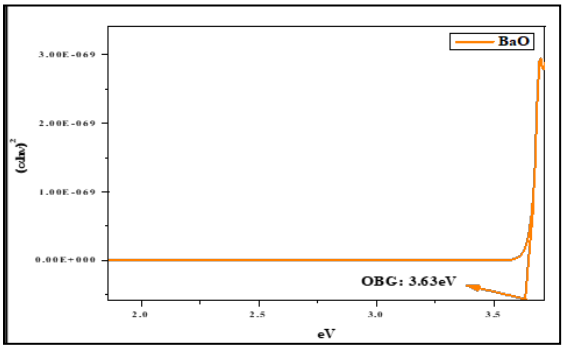

Fig. 4. UV-absorption spectra of synthesized Barium oxide Nanoparticles

D. EDAX

TABLE 2. ELEMENTAL ANALYSIS OF BAO

Element	Weight %	Atomic %
C K	13.26	37.11
O K	20.80	43.69
SiK	3.21	3.84
BaL	62.73	15.35

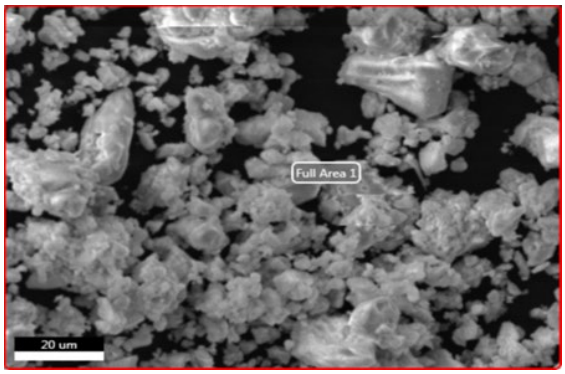

Fig. 5. SEM EDX image of BaO Nanoparticle

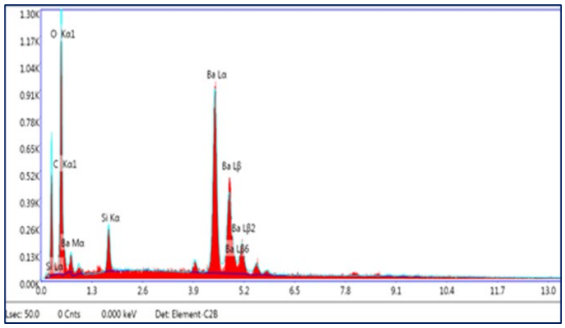

Fig. 6. Energy Dispersive X-ray of synthesized BaO nanoparticles

Elemental investigation confirms the presence of BaO in nanoparticle sample (Fig.6). The weight percentage of Carbon, Oxygen, Silicon and Barium was found to be 13.26, 20.80, 3.21 and 62.73 and atomic percentage is found to be 37.11, 43.69, 3.84, and 15.35 respectively.

V. CHARACTERIZATION OF BAO NANOPARTICLES

Irradiation is the main source for photocatalytic degradation experiments. The Brilliant green was selected for the degradation experiments against BaO nanoparticles. A standard (20mg/L) dye solution was prepared by mixing 20 mg of Brilliant green dye was prepared separately in 1 liter double distilled water and utilized for degradation studies against BaO nanoparticles. Different parameters such as BaO dosage, varied pH levels, dye concentrations and irradiations were used to enumerate the degradation studies and results were recorded. pH balance of the dye solutions was kept precisely by adding 1 N HCl and NaOH and the percentage of color degradation was calculated.

A. UV-Vis Spectroscopy

BaO dosage varied between 0.1g to 1.0g/100ml for Brilliant green dye solution. The concentration of dye solution was kept at 20ppm at pH 7 for the duration of 120 minutes. The degradation of dye has shown the following results. 96.62%

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degradation at 0.8g/100ml in 120 minutes (Fig.7). Further, these dosages were kept constant for pH, dye concentrations and irradiation studies. After a repeated studies optimum degradation was observed at 0.8g for Brilliant green dye and at this optimum dosage, release of OH• radicals are maximum and also due to the availability of optimum active sites on BaO nanoparticle. Further increasing the dosage level beyond the optimum dosage level will reduce the photo degradation due to overlying, overcrowding and collision with the ground state catalysts [19-20].

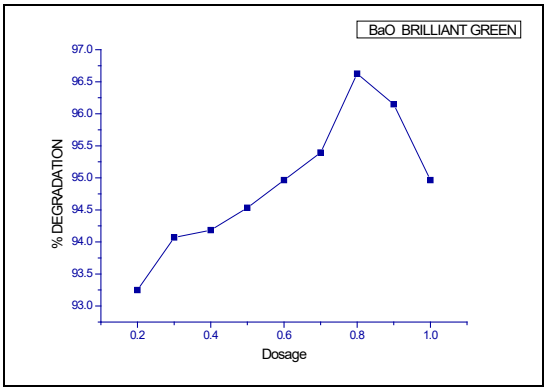

Fig. 7. Effect of catalyst concentration on Brilliant green at 120 minutes, (Brilliant green=20ppm at pH =7, BaO=0.8gm/100ml.)

B. Effect of pH

For pH experiments, the pH range was set to pH 2, pH 4, pH 6, pH 8 and pH 10 for dye solutions. The outcomes indicated that, pH fundamentally influenced the debasement effectiveness (Fig.8). The degradation rate for the Brilliant green dye solutions have shown a remarkable increase from 94.67% to 97.25% with a pH change from 2 - 4 and a reduction up to 82.87% at pH 10. An optimum degradation was achieved at pH 4. The time required to achieve the degradation was 120 minutes at an optimum dosage of 0.8g/ 100 ml dye solution.

As the dye is a cationic compound which is efficient in forming OH• radicals both in acidic and alkaline solution, OH• radicals are the main source of oxidation in carrying out photocatalytic degradation of Brilliant green [21]. In this reaction optimum amount of OH• radicals were generated at pH 4 in the solution [22-23]. Acidic condition of solution less than pH 4 has noted reduction in degradation due to excessive addition of chloride ions from hydrochloric acid. The basic condition has an inhibition effect on Brilliant green a cationic dye due to over production of OH• radicals and collision effect [24-25]. The presence of the quinone moiety in Brilliant green dye gives it a green hue [26]. The maximum absorbance was recorded at pH 4, and the pH of a solution is most likely to be in the acidic range in order to generate the optimum hydroxyl radicals [27]. With the loss of the quinone structure, an electrophilic attack of OH• radicals produced an aromatic ketone molecule [28]. At higher pH level above 4, formation of little turbidity was observed in the experimental sample. The formation of turbidity may be due to precipitation in the solution at higher pH [29]. This results in lesser absorption of photons by the dye solution and as a result less hydroxyl radicals are available in higher pH conditions [30].

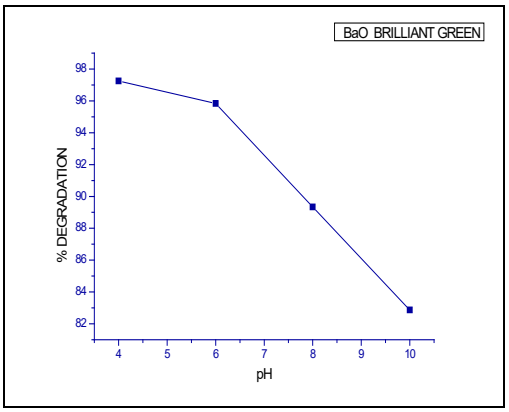

Fig. 8. Effect of pH on Brilliant green at 120 minutes (Brilliant green=20 ppm, BaO=0.8g/100ml)

C. Effect of Dye Concentration

Experiments were conducted by differing concentration of Brilliant green dye levels from 20 ppm - 50 ppm. The results for degradation of Brilliant green dye by BaO nanoparticles are 97.25% for 20ppm, 96.17% for 30ppm, 93.83% for 40ppm and 90.75% for 50ppm, respectively (Fig. 9). For the experimental dye, optimum degradation was observed at a dye concentration of 20ppm. This has proved that photo degradation capacity is directly depend upon the concentration of dye solution. An increased path length at lower dye concentration directly influences the increased photo degradation. At higher dye concentrations, the path length reduces and hence less absorption of photon by catalyst [31]. This results in reduced photo degradation rate.

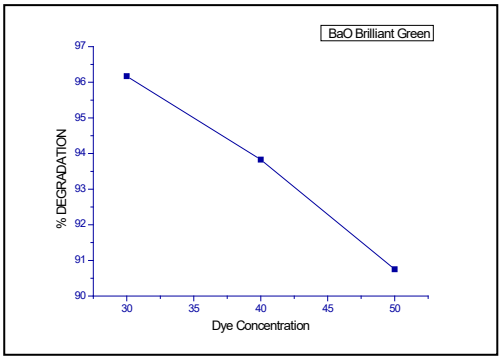

Fig. 9. ffect of dye concentration on the photo degradation of Brilliant green [BaO g/pH=0.8/4 and Brilliant green= (20+30+40+50) ppm]

D. Irradiation effect on degradation

Irradiation experiments were conducted for Brilliant green dye. Under four different conditions i.e., through sunlight alone, dye-dark-catalyst, dye-UV-catalyst and dye-sunlight-catalyst experiments were conducted in order to check the nanoparticle efficiency. The experiments were conducted at an optimum dosage of 0.8gm/100ml, optimum pH 4 and an optimum concentration of 20ppm. In sunlight alone without catalyst, the photo degradation of Brilliant green was noted almost nil. 97.25% of degradation achieved at dye-sunlight-BaO condition, 68.78% of degradation recorded at dye-UV light-BaO condition and 19.00% degradation observed at dye-dark-BaO condition (Fig.10 (a) and (b)).

This clearly emphasizes the importance of different light conditions in the degradation of experimental dye. The efficient photo degradation requires both sunlight as well photocatalyst.

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Without catalyst no degradation was observed. From this experiment, it is evident that photon energy is absorbed primarily by the BaO catalyst to induce the primary reaction to generate OH• radicals. The formation of electron hole on the catalyst surface requires excitation of semiconductors [32]. The sun light gives the excitation energy to the semiconductors and thus efficient break down of organic dye molecule is achieved [33]. The wavelength absorbed by the experimental dye is more than 400nm thereby maximum absorbance of photon is in visible range of spectrum and hence maximum dye degradation was observed in visible range [34]. The resonance between energy absorbing medium and source of energy leads to maximum degradation [35]. The UV light due to higher energy induces a different path of the reaction and creates ions, not the radicals. Visible light gives optimum energy to excite the electrons from valance band to conduction band [36].

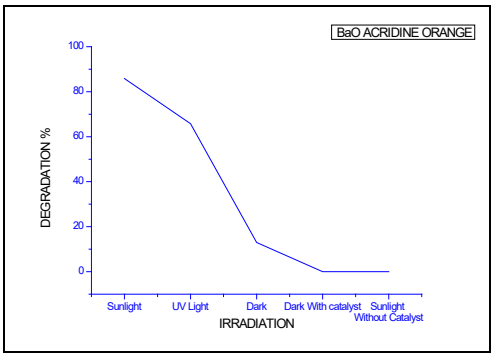

Fig. 10. (a) Effect of sunlight irradiation with respect to Dark condition and UV condition on photocatalytic degradation of Brilliant green in 120 minutes

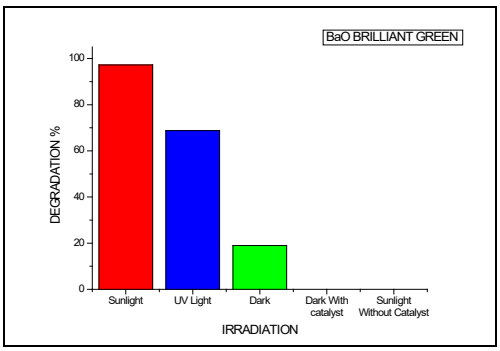

Fig. 10. (b)Effect of sunlight irradiation with respect to Dark condition and UV condition on photocatalytic degradation of Brilliant green in 120 minutes

VI. CONCLUSION

The current study used synthesized BaO nanoparticles as a photocatalyst to degrade Brilliant green dye under natural sunshine. In comparison to dark (19%) and UV (68.78%) conditions, BaO nanoparticles obtained maximal degradation (97.25%) at 0.8g/100ml of catalyst for pH 4 in a short interval period (120 min) in visible light. In the absence of sunlight, no degradation was seen. This demonstrates BaO nanoparticals' photocatalytic action in the presence of visible light. As a result, this technology can be used to industry for the efficient treatment of hazardous effluent, and it is a cost-effective solution.

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