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Adsorptive Removal of Bismarck Brown Dyes from Aqueous Media: A Comparative Review of Natural, Carbon-Based, Chitosan, Magnetic and Nanomaterial Adsorbents

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Abstract: *Bismarck Brown dyes (R, Y and G) are synthetic azo colorants used in the textile, leather, printing and paper industries, and their release in effluents colors natural waters, limits light penetration and harms aquatic life. Adsorption is widely preferred for dye removal because it is simple, efficient and inexpensive. This review compares 17 studies on the removal of Bismarck Brown dyes from aqueous media using natural and waste biomass, activated carbons, chitosan-based materials, magnetic materials, and other nanomaterials and polymers. The effects of pH, contact time, adsorbent dose, initial dye concentration and temperature are summarized together with the kinetic, isotherm and thermodynamic findings. Most systems followed pseudo-second-order kinetics and the Langmuir or Freundlich isotherms, and adsorption was generally spontaneous, although the sign of ΔH° depended on the adsorbent. Activated carbons, graphene oxide composites, magnetic carbon nanotubes and MCM-48 generally showed the highest adsorption efficiencies, whereas biosorbents offered economical and sustainable alternatives. Magnetic adsorbents eased separation and regeneration, and f-$CdWO_4$ combined adsorption with photocatalytic degradation.*

Keywords: Bismarck Brown, azo dye, adsorption, adsorbents, kinetics, isotherms, thermodynamics, wastewater treatment

I. INTRODUCTION

Bismarck Brown dyes (Bismarck Brown R, Y and G) are synthetic azo colorants that find wide use in the textile, leather, printing, paper and dye-manufacturing sectors. When effluents carrying these dyes reach natural waters, they impart strong color, limit the penetration of light, disturb aquatic life and add to environmental contamination [1].

Among the available treatment options, adsorption has drawn the greatest research interest for dye removal, since the process is straightforward, efficient, inexpensive and compatible with a broad range of adsorbent materials, including agricultural solid wastes and other industrial wastes [1], [2].

This paper consolidates the findings of 17 studies (Table I) in which the following adsorbents were examined: industrial biomass waste, Acacia nilotica, granular activated carbon (GAC), activated carbons from Ziziphus spinachristi and rubberwood sawdust, hen feathers, modified pea peels, magnetically modified biochar, magnetically modified multiwalled carbon nanotubes (MMWCNTs), mesoporous silica MCM-48, graphene oxide (GO), GO-gP(BMA-co-MAA), xanthan gum/silica nanocomposite, modified chitosan, chitosan Schiff base derivatives, chitosan–activated carbon composite, poly(thiourea-amide) and green-synthesized f-CdWO4 photocatalyst, and compares how these adsorbents perform.

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A. Aim of the Review

The aim is to survey and compare the adsorbent materials employed for taking Bismarck Brown dyes out of aqueous solutions, and to examine how operating conditions, kinetics, equilibrium behavior and thermodynamic parameters influence dye removal.

B. Objectives

- To examine the adsorption behavior of Bismarck Brown dyes;
- to compare the performance of the different adsorbents used for dye removal;
- to understand the mechanisms that govern adsorption;
- to assess how operating variables affect the process;
- to analyze adsorption kinetics and equilibrium;
- to evaluate the thermodynamic characteristics of the process;
- to identify the strengths and shortcomings of each adsorbent; and
- to judge how suitable low-cost and advanced materials are for wastewater treatment.

II. STUDIES REVIEWED AND CLASSIFICATION

Table I lists the 17 research papers reviewed together with the adsorbent examined in each. For comparison, each study was placed in one adsorbent category by the author of this review (Fig. 1): natural/waste biomass (4 studies), activated carbons (3), chitosan-based materials (3), magnetic materials (2) and other nanomaterials and polymers (5). The resulting classification of the adsorbents is shown in Fig. 2.

TABLE I: RESEARCH PAPERS REVIEWED

Sr. No.	Research Topic	Adsorbent
1	Use of industrial biomass waste to remove BBR	Industrial biomass waste
2	Uptake of BBY and RB5	Acacia nilotica
3	Simultaneous uptake of BBR and Cd(II)	Magnetic MWCNTs
4	BBR adsorption study	Mesoporous silica MCM-48
5	Color removal of BBR and Crystal Violet	Modified pea peels
6	Adsorption behavior of BBR	Graphene oxide and GO-g-P(BMA-co-MAA)
7	Elimination of organic xenobiotics	Magnetic biochar
8	Adsorption of cationic dyes	Xanthan gum/silica nanocomposite
9	Removal of BBR and Rhodamine B	Citric acid-modified chitosan Schiff base
10	Removal of BBR and Eosin Y	Modified chitosan Schiff base
11	BBY adsorption with DFT analysis	Poly(thiourea-amide)
12	Elimination of Bismarck Brown G	Chitosan–activated carbon composite
13	BBR adsorption	Ziziphus spina-christi activated carbon
14	BBR adsorption	Hen feather biosorbent
15	BBR adsorption	Rubberwood sawdust activated carbon
16	Dye degradation together with adsorption	Green-synthesized f-CdWO4
17	Dye uptake on activated carbon	Granular activated carbon

Abbreviations: BBR, BBY and BBG denote Bismarck Brown R, Y and G; RB5 is Reactive Black 5.

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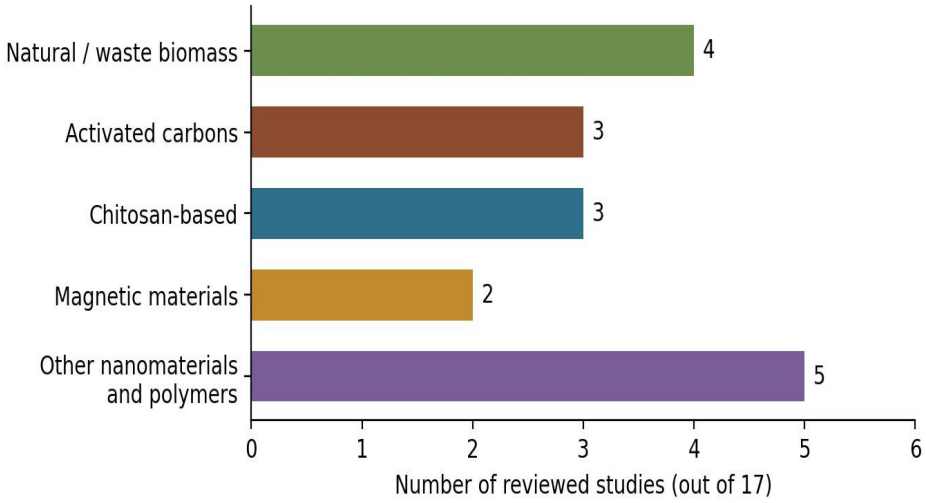


Fig. 1. Distribution of the 17 reviewed studies by adsorbent category (each study is placed in one category by the author of this review).

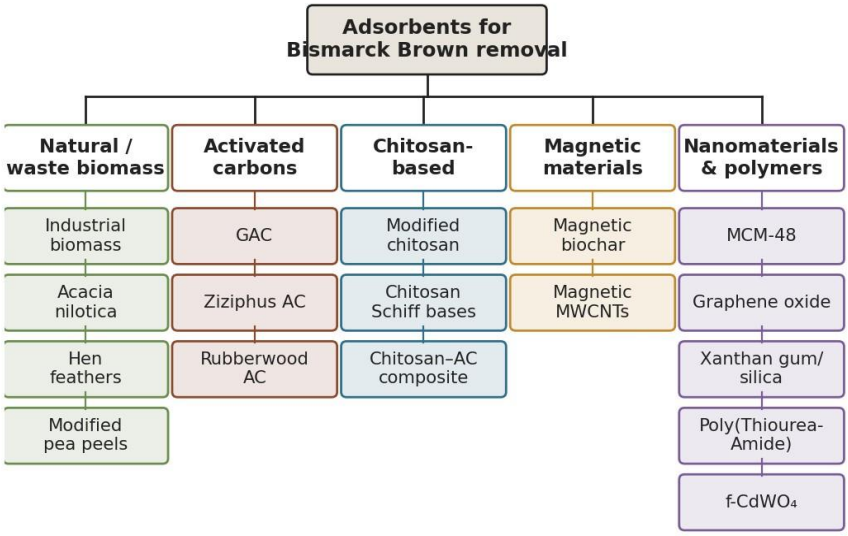


Fig. 2. Classification of the adsorbents covered in this review.

III. PRINCIPLE OF ADSORPTION AND EXPERIMENTAL APPROACH

The underlying principle is adsorption. A Bismarck Brown dye solution is contacted with the adsorbent; the dye binds to the surface or pores of the adsorbent, which is then separated to leave treated water. Dye molecules thus leave the liquid phase and attach themselves to the active sites on the adsorbent surface. The main binding interactions are electrostatic attraction, hydrogen bonding, π–π interaction and pore filling (Fig. 3).

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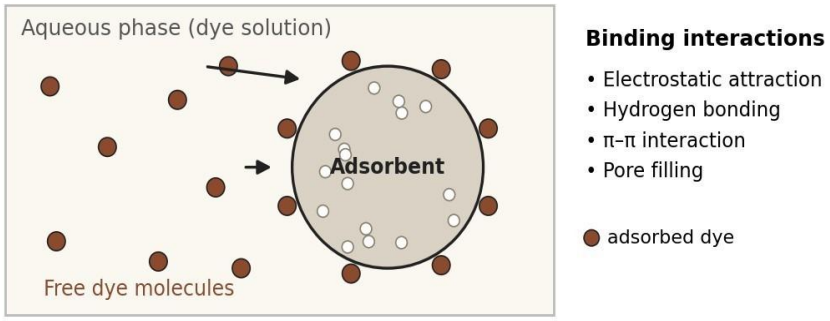


Fig. 3. Schematic of Bismarck Brown dye adsorption on a porous adsorbent, with the main binding interactions (not to scale).

The reviewed studies followed the typical batch workflow shown in Fig. 4: a dye stock solution is prepared, a set adsorbent dose is added, pH and temperature are adjusted, and the mixture is agitated for the contact time. The adsorbent is then separated by filtration, centrifugation or a magnet, the residual dye is measured by UV–Vis spectrophotometry, removal percentage and uptake are calculated, kinetic and isotherm models are fitted, and ΔG°, ΔH° and ΔS° are derived.

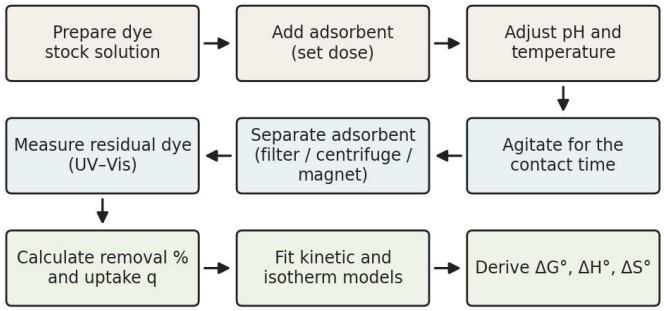


Fig. 4. Typical batch adsorption workflow used in the reviewed studies, from solution preparation to model fitting.

IV. SUMMARY OF OBSERVATIONS

The main observations drawn from the reviewed studies are summarized in Table II.

TABLE II: SUMMARY OF OBSERVATIONS FROM THE REVIEWED STUDIES

Parameter	Observation
Target pollutant	Bismarck Brown R, Bismarck Brown Y and Bismarck Brown G
Treatment method	Adsorption
Adsorbent sources	Biomass, polymers, activated carbons, composites, nanomaterials
Natural adsorbents	Acacia nilotica, hen feathers, industrial biomass, pea peels [3]
Activated carbon adsorbents	GAC, Ziziphus AC, rubberwood sawdust AC
Composite adsorbents	Chitosan–AC, xanthan gum/silica, GO-grafted polymer
Magnetic adsorbents	Magnetic biochar, magnetic MWCNTs
Nanomaterial adsorbents	MCM-48, graphene oxide, f-CdWO4
Key adsorption factors	pH, contact time, adsorbent dose, initial concentration, temperature

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Equilibrium models	Langmuir, Freundlich, Temkin [4]–[6]
Kinetic models	Pseudo-first-order, pseudo-second-order, intraparticle diffusion [7]–[9]
Thermodynamic studies	ΔG°, ΔH°, ΔS°
Main advantages	High efficiency, low cost, reusability
Sustainability aspect	Reuse of waste biomass and renewable raw materials

V. DETAILED OBSERVATIONS ON THE ADSORBENTS

A. Industrial Biomass Waste

Industrial biomass waste proved capable of removing Bismarck Brown R dye as well as Zn2+ ions. Advantages: inexpensive, puts waste to use and is environmentally friendly.

B. Acacia nilotica

Acacia nilotica removed Bismarck Brown Y and Reactive Black 5 successfully. Advantages: a natural, widely available biosorbent with good adsorption performance.

C. Magnetic MWCNTs

Magnetic MWCNTs removed BBR and Cd(II) efficiently. Magnetic MWCNT derivatives have also been applied to Bismarck Brown Y, with very high Langmuir capacities (about 1,724 to 1,961 mg/g) and chemisorption as the proposed mechanism [10]. Advantages: high adsorption capacity and magnetic recovery.

D. Mesoporous Silica MCM-48

Adsorption was fast, which is attributed to the highly ordered mesoporous framework. A separate BBR study reported pseudo-second-order kinetics, an optimum contact time of about 120 min and a Langmuir capacity near 159 mg/g [11]. Advantages: large surface area, uniform pore structure and excellent adsorption efficiency.

E. Modified Pea Peels

Modified pea peels gave good color removal for BBR and Crystal Violet. For citric-acid-modified pea peels, BBR uptake fitted the Freundlich and Sips isotherms and pseudo-second-order kinetics (Langmuir capacity about 5.4 mg/g), and the process was endothermic [12]. Advantages: reuses agricultural waste; inexpensive and renewable.

F. Graphene Oxide and GO-Grafted Polymer

These materials showed very high adsorption capacity owing to their many oxygen-containing groups. Bismarck Brown uptake on GO and GO-g-pBCM followed the Langmuir isotherm and pseudo-second-order kinetics and was endothermic and spontaneous [13]. Advantages: large surface area, strong π–π interactions and excellent adsorption performance.

G. Magnetic Biochar

Magnetic biochar removed Bismarck Brown Y effectively while allowing simple separation with a magnet. The preparation and water-treatment uses of magnetic biochar are reviewed elsewhere [14]. Advantages: reusable, easy to recover and made from a low-cost biomass precursor.

H. Xanthan Gum/Silica Nanocomposite

The nanocomposite proved highly effective for the adsorption of cationic dyes; a sol-gel xanthan gum/silica nanocomposite reached a Langmuir capacity of roughly 448 mg/g for Bismarck Brown Y [15]. Advantages: biodegradable, with many binding sites and good stability.

I. Modified Chitosan Adsorbents

Chemical modification markedly improved both adsorption efficiency and stability. A modified chitosan Schiff base has been reported for removing Bismarck Brown R and Eosin Y, with added antibacterial activity [16]. Advantages: biocompatible, renewable and rich in functional groups.

J. Poly(Thiourea-Amide)

Poly(thiourea-amide) delivered strong adsorption of BBY, credited to its sulfur- and nitrogen-containing functional groups. Advantages: high adsorption capability, strong dye interaction and support from DFT studies.

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K. Green-Synthesized f-CdWO4

This material carried out both adsorption and photocatalytic degradation of the dye. Advantages: dual-function treatment, eco-friendly synthesis and potential for complete dye degradation.

VI. EFFECT OF OPERATING PARAMETERS

A. pH

pH influences the surface charge of the adsorbent, the degree of dye ionization and the electrostatic interactions. Removal efficiency changed markedly with pH, as also reported for basic dyes [17].

B. Contact Time

Adsorption was fast in the initial stage, slowed gradually in the intermediate stage and reached equilibrium in the final stage.

C. Adsorbent Dosage

Raising the dosage generally improved dye removal, since more adsorption sites were available.

D. Initial Dye Concentration

A higher initial concentration raised the amount adsorbed per unit mass until the adsorbent became saturated.

E. Temperature

Temperature affected both dye diffusion and the position of the adsorption equilibrium.

Fig. 5 illustrates these trends for BBR on citric-acid-modified pea peels [12]. Removal rose from 58% to 88% as the dose increased from 2 to 10 g/L, from 56% to 88% as contact time increased from 10 to 60 min, from 75% to 92% as pH increased from 2 to 10, and from 83% to 89% as temperature increased from 298 to 308 K, whereas it fell from 96% to 87% as the initial concentration increased from 5 to 30 mg/L.

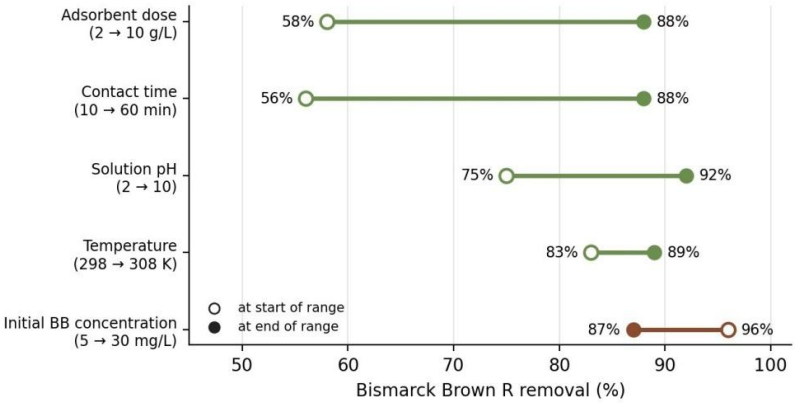


Fig. 5. Reported change in Bismarck Brown R removal by citric-acid-modified pea peels across the tested range of each operating parameter [12].

Open circles mark the start of each range and filled circles the end.

VII. ADSORPTION EFFICIENCY, KINETICS AND ISOTHERMS

A. Adsorption Efficiency

Removal efficiency is calculated as

$$\text{Removal Efficiency (\%)} = [(C0 - Ce) / C0] \times 100 \quad (1)$$

where C0 is the initial dye concentration and Ce is the equilibrium dye concentration. Under optimized conditions, most adsorbents achieved considerable dye removal.

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B. Kinetics

Kinetic analysis describes how quickly adsorption proceeds. The most commonly applied models are the pseudofirst-order [7], pseudo-second-order [8] and intraparticle diffusion [9] models. The pseudo-second-order model gave the best fit in most of the reviewed studies, pointing to strong interactions between adsorbent and dye [11], [13]. For BBR on citric-acid-modified pea peels, both liquid-film and intraparticle diffusion were found to influence the rate [12]. A typical kinetic curve with its three stages is sketched in Fig. 6.

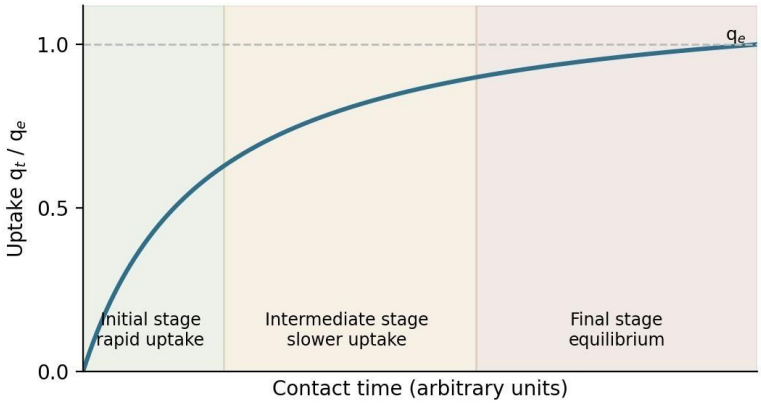


Fig. 6. Schematic of a typical adsorption kinetic curve showing the three stages described in Section VI-B (illustrative curve, arbitrary units).

C. Isotherms

Isotherms describe the equilibrium of the adsorption process; the main isotherm models are compared in [18]. The Langmuir isotherm [4] was frequently suited to activated carbons, MCM-48 and graphene-based materials [11], [13] and indicates monolayer adsorption on uniform surface sites. The Freundlich isotherm [5] was often suited to natural biosorbents and agricultural wastes and indicates a heterogeneous surface and multilayer adsorption. The Temkin isotherm [6] was applied to evaluate adsorption energy and surface interactions. The two basic shapes are compared in Fig. 7.

Three-parameter models, namely Redlich–Peterson [19], Sips [20] and Radke–Prausnitz [21], are used when twoparameter models describe the data poorly. For BBR on citric-acid-modified pea peels, the Freundlich and Sips models fitted best, the Langmuir capacity was 5.37 mg/g and the separation factor indicated favorable adsorption [12].

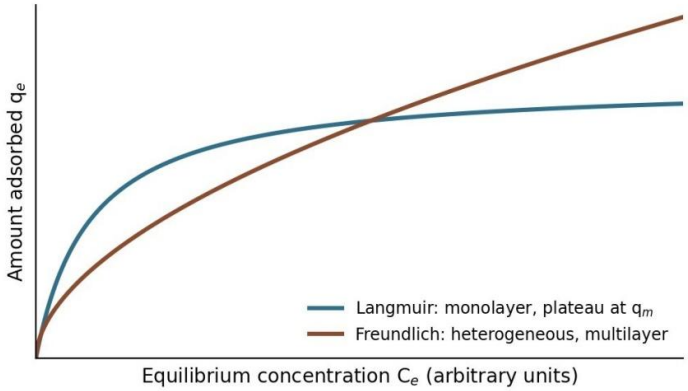


Fig. 7. Schematic comparison of Langmuir and Freundlich isotherm shapes (illustrative curves, arbitrary units).

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VIII. THERMODYNAMIC ANALYSIS

The thermodynamic parameters observed in the reviewed studies are summarized in Table III.

TABLE III: THERMODYNAMIC PARAMETERS OBSERVED

Parameter	Observation
ΔG°	Mostly negative, indicating spontaneous adsorption
ΔH°	Often positive, indicating endothermic adsorption
ΔS°	Generally positive, showing greater randomness at the solid–solution interface

Most adsorption systems were thermodynamically favorable and spontaneous [13]. For example, for BBR on citricacid-modified pea peels, ΔG° ranged from −3.57 to −5.40 kJ/mol between 298 and 308 K, ΔH° was 38.60 kJ/mol and ΔS° was 0.142 kJ/(mol·K), indicating spontaneous, endothermic, physical adsorption [12]. In contrast, Bismarck Brown adsorption on Ziziphus spina-christi carbon and on rubberwood sawdust was reported as exothermic [22], [23], so the sign of ΔH° depends on the adsorbent.

IX. COMPARATIVE EVALUATION OF THE ADSORBENTS

Table IV compares the main advantage reported for each adsorbent, and Fig. 8 compares the maximum Langmuir capacities reported for selected adsorbents. These capacities come from different dyes, conditions and methods and are therefore not directly comparable.

TABLE IV: COMPARISON OF THE ADSORBENTS

Adsorbent	Main Advantage
Industrial biomass waste	Low-cost reuse of waste
Acacia nilotica	Natural biosorbent
Hen feathers	Reuse of poultry waste; BBR uptake reported to follow pseudo-second-order kinetics [3]
Modified pea peels	Valorization of agricultural waste [12]
Activated carbon (GAC)	Very high adsorption capacity [24]
Ziziphus activated carbon	Effective Bismarck Brown uptake reported [22]
Rubberwood activated carbon	Renewable precursor; monolayer capacity of about 35 mg/g reported for Bismarck Brown on rubberwood sawdust [23]
Chitosan-based adsorbents	Functional groups and biocompatibility [16]
Graphene oxide composites	Very high adsorption efficiency
Magnetic biochar	Easy recovery with a magnet
Magnetic MWCNTs	Removes dye and metal ions together [10]
MCM-48	Ordered mesoporous structure
Xanthan gum/silica	Eco-friendly nanocomposite [15]
Poly(thiourea-amide)	Active nitrogen and sulfur sites
f-CdWO4	Adsorption combined with photocatalysis

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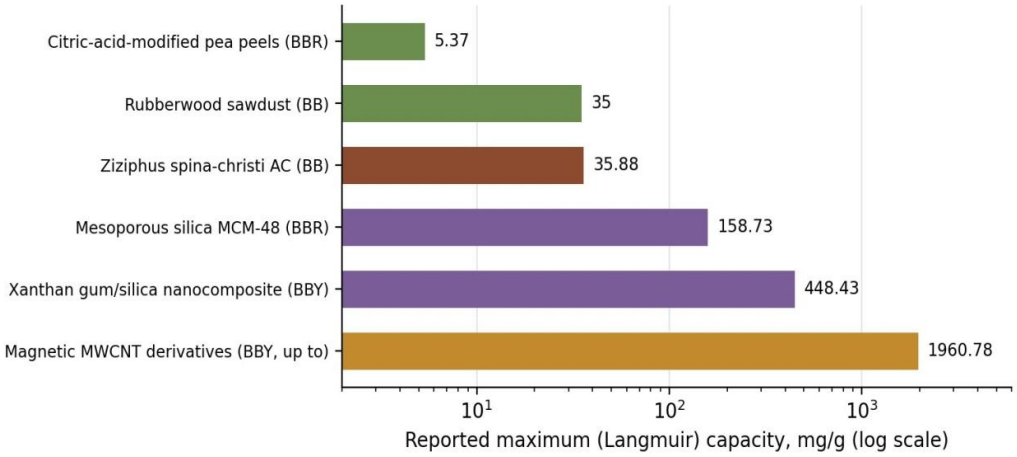


Fig. 8. Reported maximum Langmuir capacities for Bismarck Brown dyes on selected adsorbents [10], [11], [12], [15], [22], [23]. BB, BBR and BBY denote the dye form given in each study; capacities come from different dyes, conditions and methods and are not directly comparable.

X. OVERALL OBSERVATIONS

Having reviewed all the studies, the following points emerged:

Adsorption is a highly effective way to remove Bismarck Brown dyes.

Activated carbons, graphene oxide composites, magnetic carbon nanotubes and MCM-48 generally showed the highest adsorption efficiencies [24].

Biosorbents such as Acacia nilotica, hen feathers, industrial biomass waste and modified pea peels offered economical and sustainable alternatives [23], and fruit peels such as orange peel have also been used to decolorize Bismarck Brown [25].

Modified chitosan materials performed excellently and also showed antibacterial properties [16].

Magnetic adsorbents made separation and regeneration easier.

Photocatalytic materials such as f-CdWO4 provided both adsorption and degradation of dyes.

XI. CONCLUSION

The reviewed studies show that a wide range of adsorbents can remove Bismarck Brown dyes from aqueous solutions successfully. Natural biomass materials supply economical, eco-friendly options, whereas activated carbons, mesoporous silica, graphene-based materials, magnetic nanocomposites and modified polymers deliver higher adsorption efficiency. Most systems conformed to Langmuir or Freundlich isotherms, pseudo-second-order kinetics and spontaneous thermodynamic behavior. Taken together, these adsorbents hold strong promise for the sustainable treatment of dye-laden industrial wastewater.

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