



# Sorption of acid dyes from aqueous solution by using non-ground ash and slag

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## ABSTRACT

The sorption of two acid dyes (Acid Yellow 99 and Acid Red 183) into locally available industrial waste materials, namely, Granulated Blast Furnace Slag (GBFS) and Furnace Bottom Ash (FBA), has been investigated by performing batch equilibrium experiments with pH, ionic conductivity, initial dye concentration and temperature as variables. The kinetic sorption data indicated that the sorption capacity of GBFS for these dyes was almost zero. On the other hand, the color removal efficiency for FBA could reach 50% depending on the initial dye concentration. It was also found that the kinetics of sorption of Acid Yellow 99 and Acid Red 183 onto the surface of FBA at different operating conditions were best described by the Elovich kinetic model. In addition, the adsorption equilibrium data were analyzed using various adsorption isotherm models and the results have shown that the sorption behaviors of the studied dyes could be best described by the Langmuir model.

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## 1. Introduction

Dyes are coloring materials and are one of the major constituents of the wastewater discharged from many industries including textile, leather, cosmetics, paper, printing, plastic, food and pharmaceuticals. They are visible even at concentrations as low as 0.005 ppm and can therefore create aesthetic problems. However, more important than any potential aesthetic problem is that the dyes can cause health problems, may affect growth of microorganisms and can hinder photosynthesis in aquatic plants [1]. Moreover, the complex aromatic molecular structures of dyes make them more stable and more difficult to biodegrade in water.

Various methods for dye removal from wastewaters exist including adsorption, coagulation, ultrafiltration, chemical oxidation and photocatalytic oxidation [2]. Amongst these methods, adsorption techniques employing activated carbon have been found to be reasonably effective in the removal of dyes [3]. However, activated carbon is quite expensive, and its use results in considerable loss (10–15%) of the adsorbent during regeneration [4]. Economic considerations require the use of inexpensive and locally available materials as the adsorbent for the removal of dyes. Such adsorbents range from industrial waste products [5–7] such as waste rubber tires, blast furnace slag, bottom ash and lignin, to agricultural products [8,9] such as wool, rice straw, coconut husk, sawdust, and peat moss. In addition, the low cost and commercial availability of biosorbents have increased attention in their use in recent years [10].

The prime objective of the present study is to explore the use of Granulated Blast Furnace Slag (GBFS) and Furnace Bottom Ash (FBA), which are locally available, as a sorbent in color removal from wastewater and to investigate the kinetics and mechanisms involved in dye adsorptions on GBFS and FBA.

GBFS is a by-product obtained in the production of pig iron in blast furnaces and is formed by the combination of earthy constituents of iron ore with limestone flux. When the molten slag is swiftly quenched with water, it forms a fine, granular, almost fully non-crystalline and glassy form, which is known as granulated slag. Due to the presence of high contents of silica and alumina in a non-crystalline state, GBFS is used as a cementitious ingredients in mortars, as an additive in blending cements, as fine aggregates or as mineral admixtures in concrete [11]. FBA, on the other hand, is a by-product of coal-fired power plants, where it agglomerates and settles down to the bottom of the combustion furnace. Its applications include structural fills, road bases and sub-bases, asphalt fillers, roofing granules, sandblasting grit and aggregates in cement and concrete products.

## 2. Materials and methods

### 2.1. Physical and chemical properties of sorbents

GBFS was obtained from the Erdemir Iron and Steel Production Plant, which is located in the eastern Black Sea region of Turkey (Zonguldak). Çatalağzı thermal power plant, where bituminous coal is the main fuel, is also located in the same region and FBA was obtained from this plant. The physical properties of GBFS and FBA are reported in Table 1. Atomic absorption spectrophotometry techniques were used for the chemical analyses of GBFS and FBA and the results has

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**Table 1**  
Physical properties of GBFS and FBA.

Property	GBFS	FBA
Loose unit weight, kg/m <sup>3</sup>	1052	620
Compact unit weight, kg/m <sup>3</sup>	1236	660
Specific gravity, g/cm <sup>3</sup>	2.08	1.39
Absorption (%)	8.30	6.10
Amount of clay (%)	1	2.0
Loss on ignition (%)	1.8	2.4
Prop. of lightweight particles (%)	3.00	7.00

been presented in Table 2. Major components in both sorbents were SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, CaO and MgO.

In this study, it was aimed to investigate GBFS and FBA as sorbents in their original form, i.e., as they were taken from the plants and were not grinded. As a result, the cost of sorbents was kept at minimum. Because of not grinding, the smallest mesh opening has been selected and the GBFS/FBA samples were sieved by 250 μm opening. Then they were dried at 105 °C for 24 h and no extra treatment was applied to either sorbents.

## 2.2. Sorbates

Commercial quality acid dyes (Acid Red 183, C.I. 18,800, C<sub>16</sub>H<sub>11</sub>-ClN<sub>4</sub>Na<sub>2</sub>O<sub>8</sub>S<sub>2</sub>.xCr, MW: 584.86 g/gmol and Acid Yellow 99, C.I. 13,900, C<sub>16</sub>H<sub>13</sub>N<sub>4</sub>NaO<sub>8</sub>S.Cr, MW: 496.4 g/gmol) were obtained from Aldrich and used without any further purification. These dyes are water-soluble dyes, which are widely used in wool and leather dyeing. The chemical structures of Acid Red 183 and Acid Yellow 99 are presented in Fig. 1. Both dyes are characterized by the presence of a hydroxyl group ortho to the azo-group and are colored aromatic compounds which require the presence of chromium. However, it is known that incomplete reduction of dichromate could lead to release of the toxic chromium (VI) salt into the environment [12]. Therefore, wastewater containing these dyes has to be treated before discharge.

## 2.3. Batch equilibrium experiments

Adsorption studies were performed by batch equilibrium experiments. Aqueous solutions were prepared by dissolving dyes in distilled water at the concentration of 20, 40, 60, 80, 100 and 120 mg L<sup>-1</sup>. Solutions were prepared by using distilled water to minimize interferences in the study. The studied pH range was 4 to 8. The solution pH was set to desired value by using HCl before the addition of sorbents. After the adjustment of pH, 400 mL of the dye solution was replaced in a glass flask and 4 g sorbents was introduced. Sorbent to liquid ratios were kept constant at 10 g/L during the all adsorption experiments. Then, the glass flasks were submerged into a temperature controlled water bath and the temperature was set to 20 °C. The glass flasks were shaken for a day and the sorbents were separated from the dye solutions by centrifuging at 3600 rpm and filtered (whatman 42) at the end of mixing. The residual dye color in the solution was analyzed colorimetrically using a spectrophotometer (UV-VIS Spectrophotometer Pharo 300 Spectroquant). To evaluate dye removal, color concentrations were measured before and after the experiment by measuring absorbance. The maximum wavelengths for

Acid Red 183 and Acid Yellow 99 was determined experimentally and found as 483 nm and 464 nm, respectively. The amount of dye adsorbed by GBFS or FBA was calculated by:

$$q_e = \frac{(C_0 - C_e)V}{W} \quad (1)$$

where  $q_e$ ,  $C_0$ ,  $C_e$ ,  $V$  and  $W$  represent the amount of dye removed from solution per unit mass of sorbent (mg g<sup>-1</sup>) at equilibrium, the concentration of dye in the solution before mixing with sorbent (mg L<sup>-1</sup>), the equilibrium concentration of dye left in the solution (mg L<sup>-1</sup>), the solution volume (L) and the weight of dried GBFS or FBA (g), respectively.

## 2.4. Sorption dynamics

The adsorption mechanisms of Acid Red 183 and Acid Yellow 99 on GBFS or FBA was analyzed by using pseudo first order, pseudo second order, Elovich and Weber–Morris intraparticle rate equations.

According to pseudo first order equation model, dye adsorption on GBFS or FBA can be considered as a reversible process and equilibrium has been attained between the liquid and solid phases. It assumes that the rate of change of dye adsorption with time is a linear function of the difference between the dye uptakes at equilibrium and time  $t$  and adsorption can be described as the diffusion control process. It is generally written as [13]:

$$\frac{dq}{dt} = k_1(q_e - q) \quad (2)$$

where  $q$  is the amount of lead adsorbed per unit mass of adsorbent (dye uptake) at time  $t$ , and  $k_1$  is the pseudo first order rate constant.

While the pseudo first order kinetic model is used for interparticle and intraparticle mass transfer mechanisms, the reaction kinetics approximation is usually employed for systems where the reaction step at pore surfaces is the controlling step. In this case, the adsorption dynamics can be represented by a second order reaction rate constant as [14]:

$$\frac{dq}{dt} = k_2(q_e - q)^2 \quad (3)$$

where  $k_2$  is the pseudo second order rate constant.

Elovich's equation is another kinetic model, which is based on the adsorption capacity and can be derived from either a diffusion-controlled process or a reaction-controlled process [15]:

$$\frac{dq}{dt} = ae^{-bq} \quad (4)$$

where 'a' and 'b' are constants and represent the initial adsorption rate and the surface coverage, respectively.

Finally, the intraparticle diffusion model of Weber and Morris [16] has been tested. According to this model, the variation of dye uptake with time can be represented as:

$$q = k_i \sqrt{t} \quad (5)$$

where  $k_i$  is the intraparticle diffusion constant. Eq. (5) yields a parabola with the initial curved portion showing a surface adherence or boundary layer effect; and the following linear part identifies intraparticle or pore diffusion and the slope gives  $k_i$ . If the intercept is zero the surface adherence in the sorption rate-limiting step is negligible. The larger the intercept the greater is the contribution to the surface adherence in the rate-limiting step.

**Table 2**  
Chemical compositions of GBFS and FBA (%).

	SiO <sub>2</sub>	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	S	K <sub>2</sub> O	MnO	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	P <sub>2</sub> O <sub>3</sub>
GBFS	35.09	37.79	5.50	17.54	0.30	0.66	–	0.83	–	0.68	0.37
FBA	57.90	2.00	3.20	22.60	0.086	–	0.604	–	6.50	–	–

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