

Hydrothermal modification of ceramic waste: Characterization, optical properties and low-cost adsorbent agent for removal organic dyes

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Abstract

In this research, the sanitary ware waste is modified by the addition of sodium hydroxide via the hydrothermal method. The obtained product was characterized utilizing different techniques such as X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), and Diffuse reflectance spectroscopy (DRS). XRD showed the synthesized samples before and after calcination containing the two phases Faujasite-NaY and Sodalite. The average crystallite sizes of S17R and S17C samples were recorded from the x-ray diffraction peaks to be 24.6 and 15.4 nm, respectively. Also, the optical properties were studied using diffuse reflectance spectroscopy. The band gap and color analysis of the synthesized products were studied. The values of light and solar reflectance were determined using JISA5759 2008, and JISK5602 2008 methods. In addition, the prepared products have an efficient adsorption performance to anionic dyes (sunset yellow, naphthol green B, and amaranth dyes). The maximum adsorption capacities of S17R and S17C samples toward sunset yellow, naphthol green B, and amaranth dyes are 65.38/39.1/51.66 mg/g and 82.7/46.97/58.33 mg/g, respectively.

Keywords: Ceramic waste; Adsorption; Anionic dyes; Optical properties; color analysis.

1. Introduction

Pollution is a major issue that damages the environment to varying degrees, especially in watercourses, and can endanger humans and other species. Water is a necessary component of life and one of the components required for the maintenance of all aspects of living, including food production, economic progress, and overall well-being [1, 2]. Water pollution has a substantial influence on water consumption and a variety of pollutants, including pharmaceuticals, organic and inorganic chemicals, colors, heavy metals, viruses, and radioactive materials [3]. Out of the organic pollutants, dyes are completely popular in industry like cosmetics, paper printing, food processing, rubber, textile, plastics and other that use significant amount of dyes in their products [4]. Yet, these industry pollutants include a large quantity of toxic and cancer-causing dyes [5-7]. As a result, it is critical to treat colored water for removal of pollutants such as dyes using a range of modern approaches [8] such as adsorption, photo catalysis, oxidation, flocculation, electrolysis, biodegradation, ozonation, membrane filtration, ion-exchange, coagulation, and etc. Among all these techniques, the adsorption technique since it is currently considered as the most favored approach because of its cost-effectiveness, simplicity, lack of secondary pollutants, and superior performance over other traditional treatment methods in the elimination of dyes from wastewater [9].

Several sorbents based on low-cost materials by acting as recycled waste for had been used for the water treatment, which included sago waste, banana pith, silk cotton hull, maize cob and coconut tree sawdust [10], coconut shell [11-13], banana peel and green coconut mesocarp [14-16], rice husk ash and aluminum cans wastes [17-21], clam shells [22, 23], Brazilian marble waste [24, 25]. One of the most low-cost materials, Sanitary ware waste, a type of ceramic ware, is typically found in bathtubs, urinals and sinks

[26] that contributes to the reduction of environmental problems for the low-cost adsorbent agent for elimination of anionic dyes from aqueous media using hydrothermal method.

The hydrothermal synthesis is a straightforward procedure that can produce fine, uniform, and non-agglomerated powder in a single step. Particle characteristics can be manipulated, and desired repeatability of particles can be attained, by modifying the reaction parameters and method. In the meantime, the cost of tools, energy and precursors has significantly decreased, which is better for the environment [27]. In this work, aqueous solutions of sodium hydroxide were utilized for preparation of the low-cost adsorbing agent for the elimination of anionic dyes from aqueous media from sanitary ware waste using hydrothermal route. The characteristics of the obtained nanosized samples are well studied using several instruments like XRD, FTIR, and DRS.

2. Experimental

2.1. Chemicals and reagents

The primary starting materials and chemicals were used in the construction and installation of the fabrication process and the elimination procedure for the organic dyes were hydrochloric acid (HCl; 30-34%), Naphthol Green B dye ($C_{30}H_{15}FeN_3Na_3O_{15}S_3$, 99%), Sunset Yellow dye ($C_{16}H_{10}N_2Na_2O_7S_2$, 99.9%), Amaranth dye ($C_{20}H_{11}N_2Na_3O_{10}S_3$, 85-95%), and sodium hydroxide (NaOH, 98%). The Sigma-Aldrich Chemical Company was the source of all starting chemicals and compounds which were used as provided.

2.2. Characterization tools

X-ray powder diffraction, (Rigaku MiniFlex 600 diffractometer with monochromatic Cu-K α radiation, 1.54178 (\AA) at room temperature in the angular range of 10° - 70° (2θ)), was used for identification of the size and phases of the as-prepared (S17R) sample and the calcinated (S17C) sample. Fourier Transform Infrared

Spectrometer (Thermo Scientific; model Nicolet iS10), with addition of KBr as the standard, and the IR spectral was used for the study of (4000 to 400 cm^{-1}) of the as-prepared (S17R) sample and the calcinated (S17C) sample. A pH meter was measured the pH values of solutions of the sunset yellow, naphthol green B and amaranth dyes using digital pH/mV and temperature meter using Adwa model (AD1030, Italy). Using an integrating sphere calibrated with barium sulfate as the white standard and a Jasco UV-Vis spectrophotometer (Jasco; model V 670), diffuse reflectance of the as-prepared (S17R) sample and the calcinated (S17C) sample were examined in the ultraviolet-visible NIR range (200-2500 nm). Color parameters were determined using the CIE-Lab colorimetric approach for the as-prepared (S17R) and the calcinated (S17C) samples. A second technique for calculating the color axes is CIE LCH. The chroma parameter (C^*) of the as-prepared (S17R) sample and the calcinated (S17C) sample calculated from $C^* = \sqrt{(a^*)^2 + (b^*)^2}$ and the hue angle h^* of the as-prepared (S17R) sample and the calcinated (S17C) sample is determined from $h^* = \tan^{-1}(b^*/a^*)$. Japanese Industrial Standards Association (JIS) standard JISK5602:2008 and JISA5759:2008 methods are installed for determination of the solar reflectance (SR) of the as-prepared (S17R) sample and the calcinated (S17C) sample and it is investigated by using the following formula (1).

$$SR = \int_{\lambda_1}^{\lambda_2} R(\lambda)I(\lambda)d\lambda / \int_{\lambda_1}^{\lambda_2} I(\lambda)d\lambda \quad (1)$$

Where, $I(\lambda)$ and $R(\lambda)$ are the standard solar spectrum and the experiment reflectance [28, 29].

2.3. Method

The samples were fabricated by mixing three grams of sanitary ware waste and 21 g of sodium hydroxide in bidistilled water. Then, the resulting mixture was hydrothermally treated at 150 °C for 30 h in an electric oven using Teflon-lined stainless-steel autoclave. After completion of the hydrothermal reaction, the autoclave was then allowed to naturally be cooled and the formed precipitates were filtered, washed thoroughly several

times with distilled water and dried at 100 °C overnight, then calcinated at 550 °C for 3 h and labeled the as-prepared (S17R) sample and the calcinated (S17C) sample.

3. Results and discussion

3.1. X-ray diffraction studies

The as-prepared (S17R sample) and the calcinated (S17C sample) were studied using XRD as displayed in Figure (1). XRD patterns show the presence of Faujasite-NaY and Sodalite structures according to cards no. 1545416 and 9003331, respectively. As result of hydrothermal treatment, the waste samples contain two phases (Faujasite-NaY and Sodalite). Debye-Scherrer equation are utilized for the calculation of the obtained crystal sizes of the as-prepared S17R and the calcinated S17C samples by using ($S = 0.9\lambda/G \cos \theta$) equation. Where θ is the Bragg diffraction angle, G is the X-ray full width of the diffraction peak at half-maximum height, and λ is the X-ray wavelength (1.5406 Å for Cu K) [30, 31]. The average crystallite size (S) of the synthesized (S17R and S17C samples) was detected to be 24.6 and 15.4 nm from the x-ray diffraction lines, respectively.

3.2. FT-IR studies

The S17R and S17C samples are characterized using FT-IR patterns as shown in Figure 2. The findings demonstrated that the peaks at about 3450–3510 cm^{-1} and 1648 cm^{-1} can be linked to the stretching and bending vibrational mode of the adsorbed molecule on the synthesized S17R and S17C samples such as water molecules and hydroxides groups. Asymmetric internal and exterior vibrations of S-O-S (where S = Si or/and Al) are also responsible for the peaks that were observed at 970-985 cm^{-1} and 1448-1477 cm^{-1} , respectively. Moreover, the peaks in the zone of 560-665 cm^{-1} and 730-734 cm^{-1} can be attributed to the internal and exterior symmetric vibrations of S-O-S in the synthesized S17R and S17C samples, respectively. Moreover, S-O-S bending vibration is responsible for the lines that developed in the 430-460 cm^{-1} region in the synthesized S17R and S17C samples [30-33].

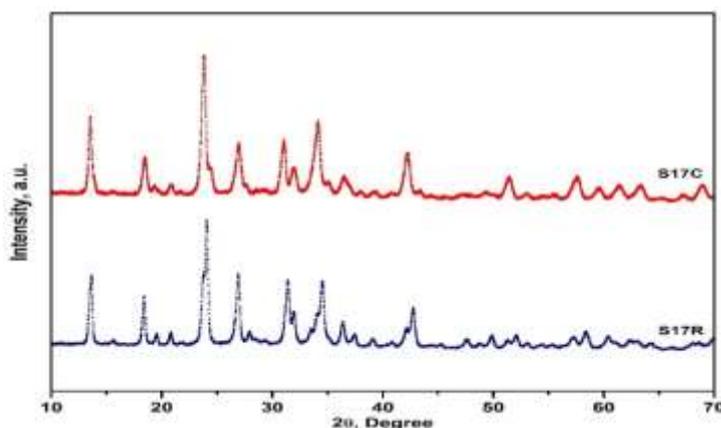


Fig. (1) XRD pattern of the as-prepared (S17R sample) and the calcinated (S17C sample).

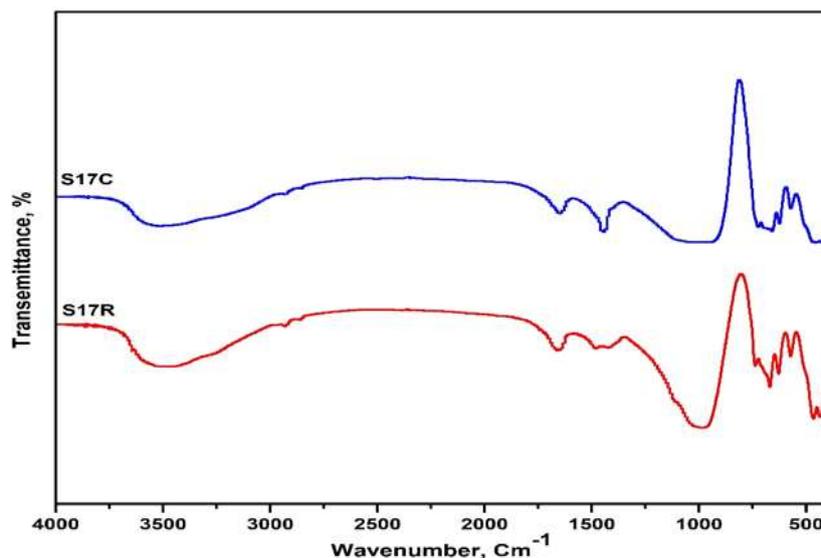


Fig. (2) FT-IR spectrum of the synthesized Faujasite-NaY and Sodalite composite (S17R and S17C samples).

3.3. Optical properties of the synthesized composite

The as-prepared and calcinated samples in the form of Faujasite-NaY and Sodalite composite (S17R and S17C samples) was studied using UV-VIS and NIR diffuse reflectance. Figure (3) displayed the measured reflectance between 200-2500 nm. The reflectance edge between 260-350 nm for (S17R and S17C samples). Figure (4) demonstrates the UV-vis-NIR absorption spectra of the as-prepared and calcinated sample in the form of Faujasite-NaY and Sodalite composite (S17R and S17C samples). Kubelka Munk function is used for deduction of the absorption coefficients (Y) using the experiment reflectance data as demonstrated in equation no (2)[28, 34, 35].

$$R(r) = Y = (K - M) = (1 - R)^2 / 2R \quad (2)$$

Where, R is the experiment reflectance, R(r) is K-M function, Y is absorption coefficients.

According to Figure (5), The absorption edge of the as-synthesized and calcinated (S17R and S17C samples) appeared in the range of 240-250 nm. The optical direct band gap of the S17R and S17C samples can be determined using equation No. (3):

$$(Yh\nu)^u = Z(h\nu - E_g) \quad (3)$$

Where Y is Kubelka Munk function, h and ν are constants, and the value of u is equal to 2 as indicated for the permitted direct electronic transitions. The direct band gap of sample in the form of Faujasite-NaY and Sodalite composite (S17R and S17C samples) calculated to be 4.1 eV and 4.2 eV for (S17R and S17C) samples, respectively as shown in Figure (5) [36].

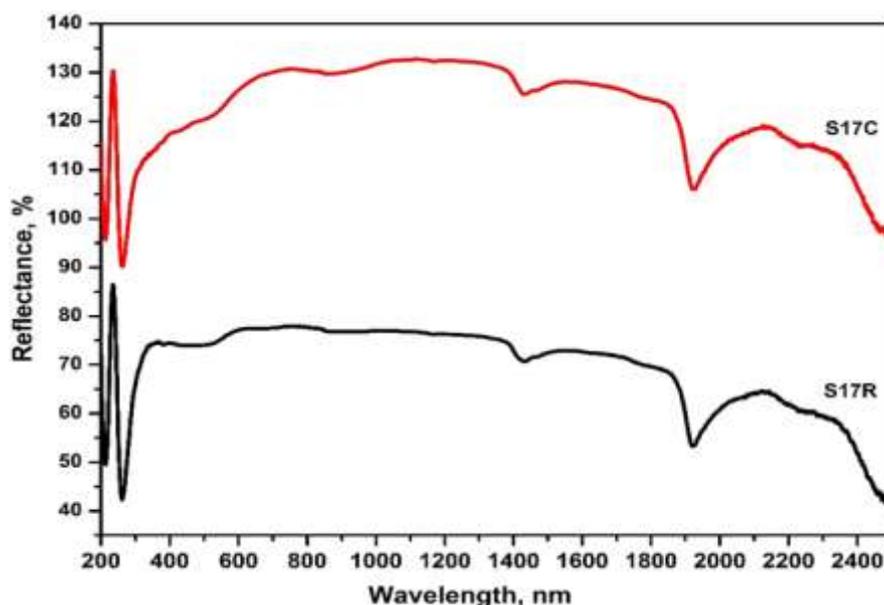


Fig. (3) UV-VIS and NIR diffuse reflectance of the Faujasite-NaY and Sodalite composite (S17R and S17C samples).

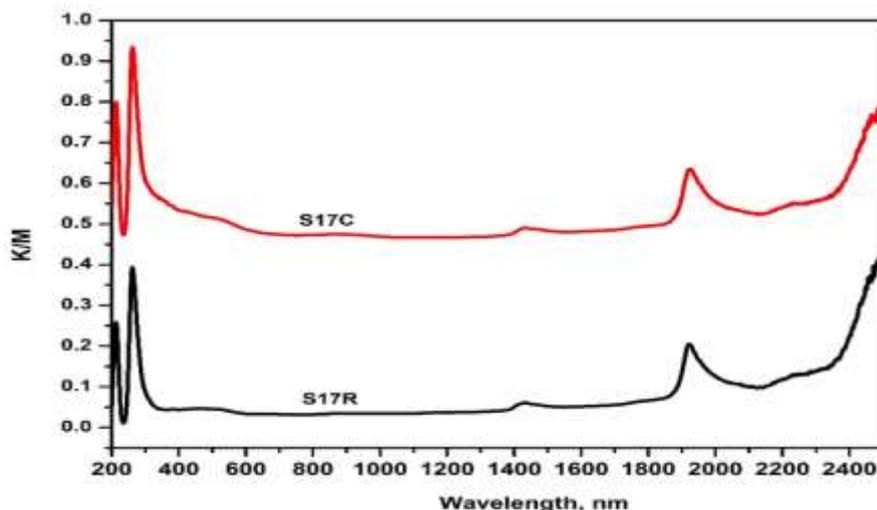


Fig. (4) UV-VIS and NIR absorption spectra of the Faujasite-NaY and Sodalite composite (S17R and S17C samples).

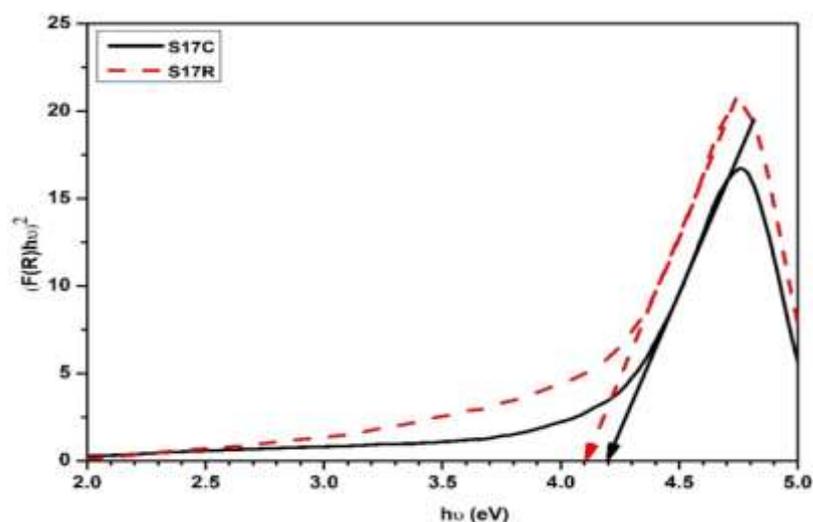


Fig. (5) The direct band gap of the Faujasite-NaY and Sodalite composite (S17R and S17C samples).

According to CIE-L*a*b* and CIE-L*C*H* methods for determination of the color analysis of the synthesized Faujasite-NaY and Sodalite composite (S17R and S17C samples), the values of L*/a*/b*/c*/h* calculated to be 89.63, 1.23, 1.23, 1.74 and 44.87 for S17R sample and 87.74, 2.25, 3.57, 4.21 and 57.78 for S17C sample [36]. The calculated data reflected the light-yellow color of the synthesized Faujasite-NaY and Sodalite composite (S17R and S17C samples). Light and solar reflectance of the synthesized Faujasite-NaY and Sodalite composite (S17R and S17C samples) determined by using JISK5602:2008 and JISA5759:2008 methods according to the eq. no. (1). The value of light and solar reflectance using JISA5759:2008 determine to be 75.62 % and 75.36% for (S17R sample) and 71.79 % and 74.53 % for (S17C sample). Also, the solar reflectance determined in the UV-VIS, NIR and total solar reflectance using JISK5602:2008 method to be 75.9 %, 74.64 %, 75.32 %, 72.53 %, 77.49 % and 74.64 % for Faujasite-NaY and Sodalite composite (S17R and S17C samples), respectively [34].

3.4. Effect of the obtained samples on the removal of anionic dyes

The obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) from the hydrothermal method were used as adsorbents for the elimination of sunset yellow, naphthol green B and amaranth dyes from the aqueous phase utilizing the batch technique. The elimination process of the anionic dyes using the synthesized Faujasite-NaY and Sodalite composite (S17R and S17C samples) was examined using different adsorption parameters: pH = 2, 0.05 g dose adsorbents, equilibrium time 60 min and (250 mg/L of sunset yellow dye), (200 mg/L of naphthol green B dye), and (150 mg/L of amaranth dye). The adsorption capacities of the synthesized Faujasite-NaY and Sodalite composite (S17R and S17C samples) were found to be 65.38, 82.7, 39.1, 46.97, 51.66, 58.33 mg/g for removal sunset yellow, naphthol green B and amaranth dyes, respectively. The spectra before and after adsorption processes of anionic dyes and removal capacities of the obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) represented in Figure (6).

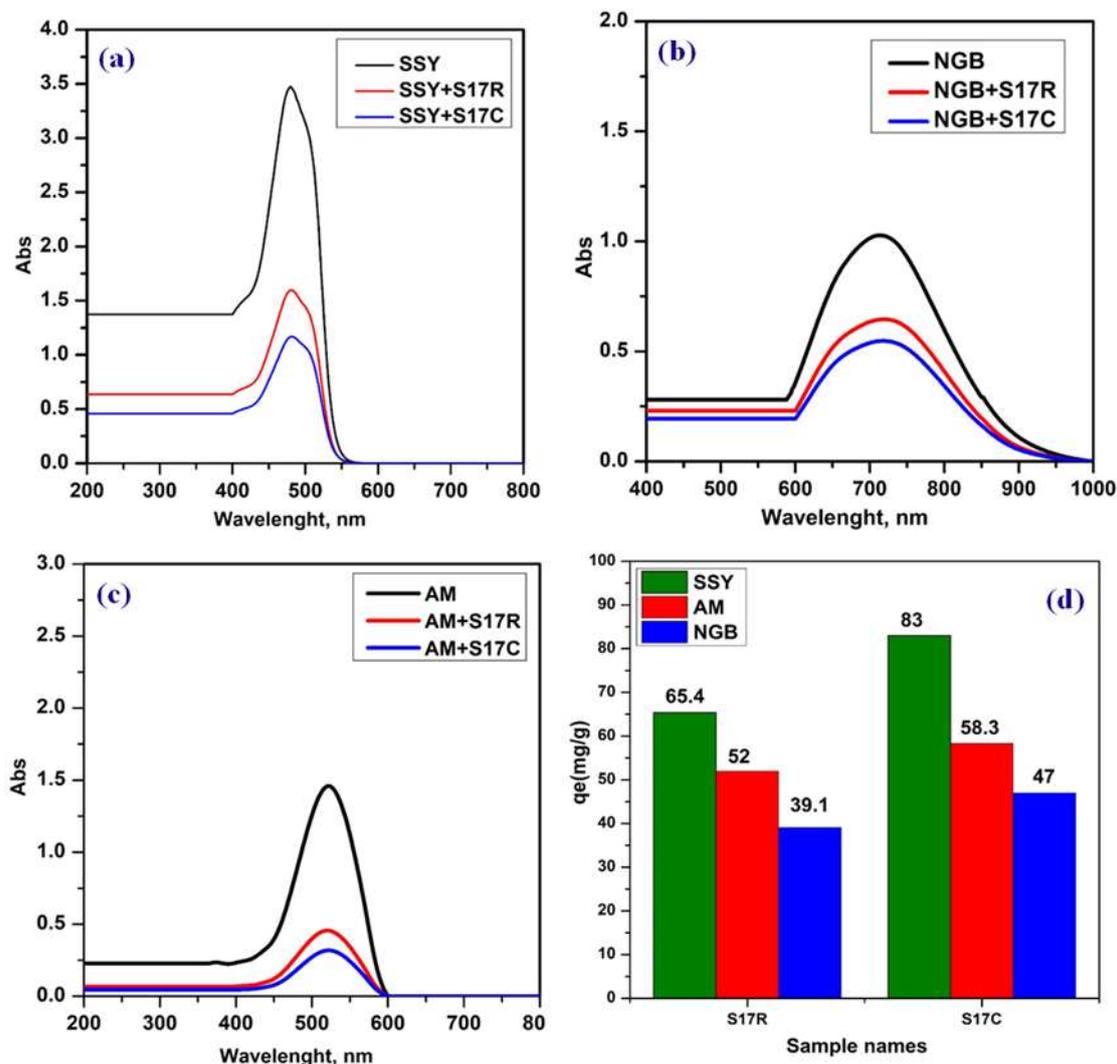


Fig. (6) The spectra before and after adsorption processes of sunset yellow (a), naphthol green B (b) and amaranth (c) dyes and removal capacities of S17R and S17C samples (d).

4. Conclusion

Ceramic waste was utilized as the low-cost starting materials or synthesis of Faujasite-NaY and Sodalite structures using a hydrothermal method. The phase structure, functional groups and optical properties of the obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) were successfully characterized. XRD showed the fabricated samples before and after calcination containing the two phases Faujasite-NaY and Sodalite. The average crystallite sizes of obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) were determined to be 24.6 and 15.4 nm, respectively. UV-VIS-NIR diffuse reflectance spectra of the synthesized samples displayed the reflectance edge between 260-350 nm. The absorption edge of obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) appeared in the range of 240-250 nm. The direct band gap calculated to be 4.1 eV and 4.2 eV for S17R and S17C samples, respectively. The values of color parameters ($L^*/a^*/b^*/c^*/h^*$) calculated to be 89.63, 1.23, 1.23,

1.74 and 44.87 for S17R sample and 87.74, 2.25, 3.57, 4.21 and 57.78 for S17C sample and it reflected the light-yellow color of the synthesized samples. The value of light and solar reflectance determined to be 75.62 % and 75.36% for S17R sample and 71.79 % and 74.53 % for S17C sample. Also, the solar reflectance determined in the UV-VIS, NIR and total solar reflectance to be 75.9 %, 74.64 % and 75.32 % for S17R sample and 72.53 %, 77.49 % and 74.64 % for S17C sample, respectively. Finally, obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) may be employed as highly effective and cost-effective nano-adsorbents for anionic dye removal from aqueous media. The adsorption capacities of obtained Faujasite-NaY and Sodalite composite (S17R and S17C samples) were resolved to be 65.38, 82.7, 39.1, 46.97, 51.66, 58.33 mg/g for removal sunset yellow, naphthol green B and amaranth dyes, respectively.

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