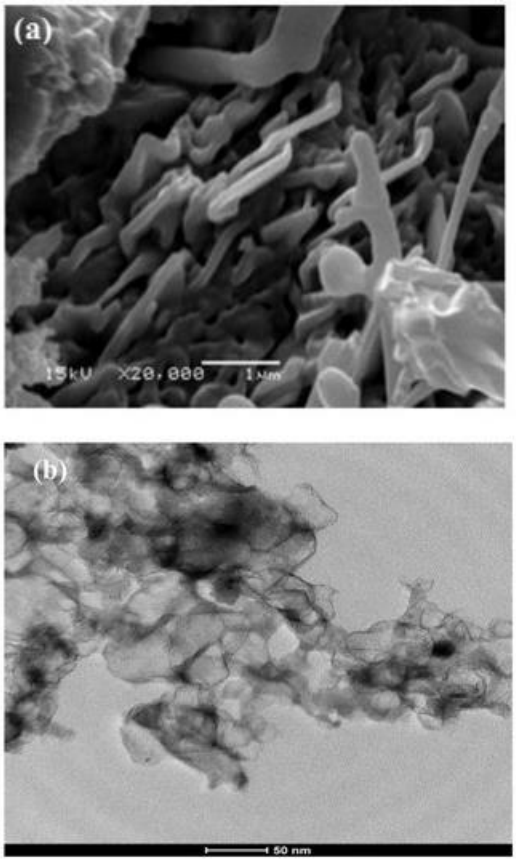

Figure S1. SEM and TEM of graphene [1].

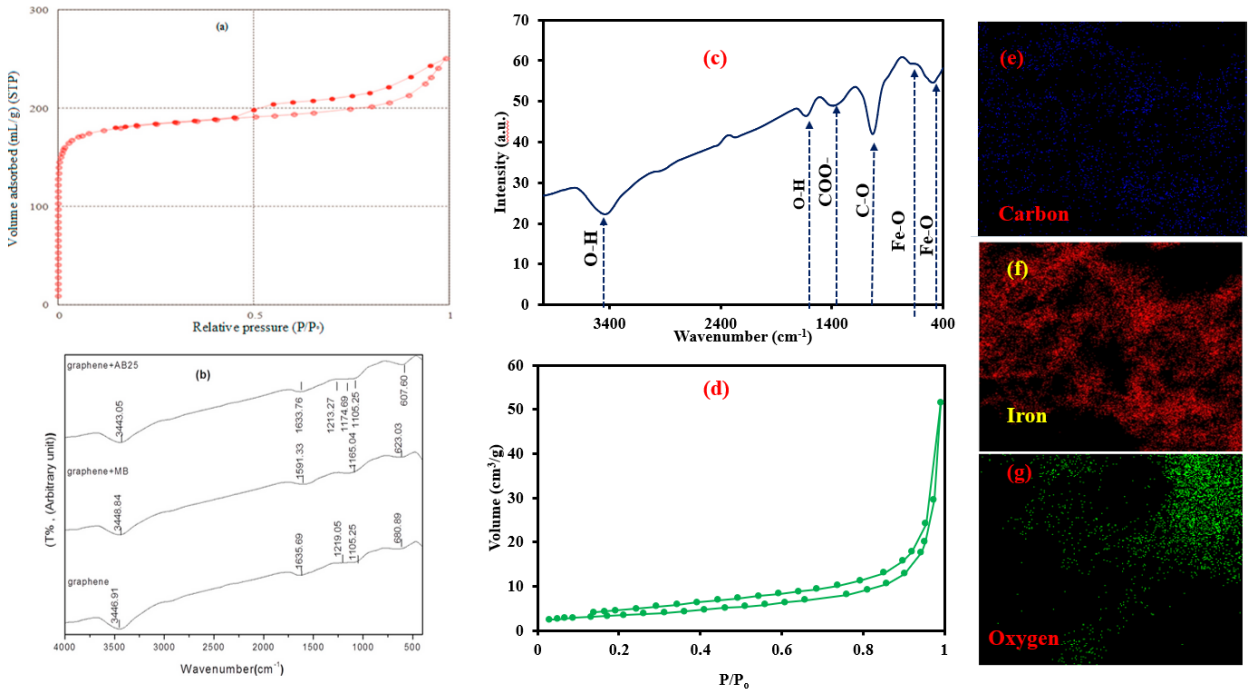

Figure S2. (**a**,**b**) adsorption-desorption isotherms and FTIR spectra of graphene, (**c**,**d**) FTIR spectra and adsorption-desorption isotherms of ZVIgraphene and (**e**–**g**) elemental mapping of graphene and ZVIgraphene [1].

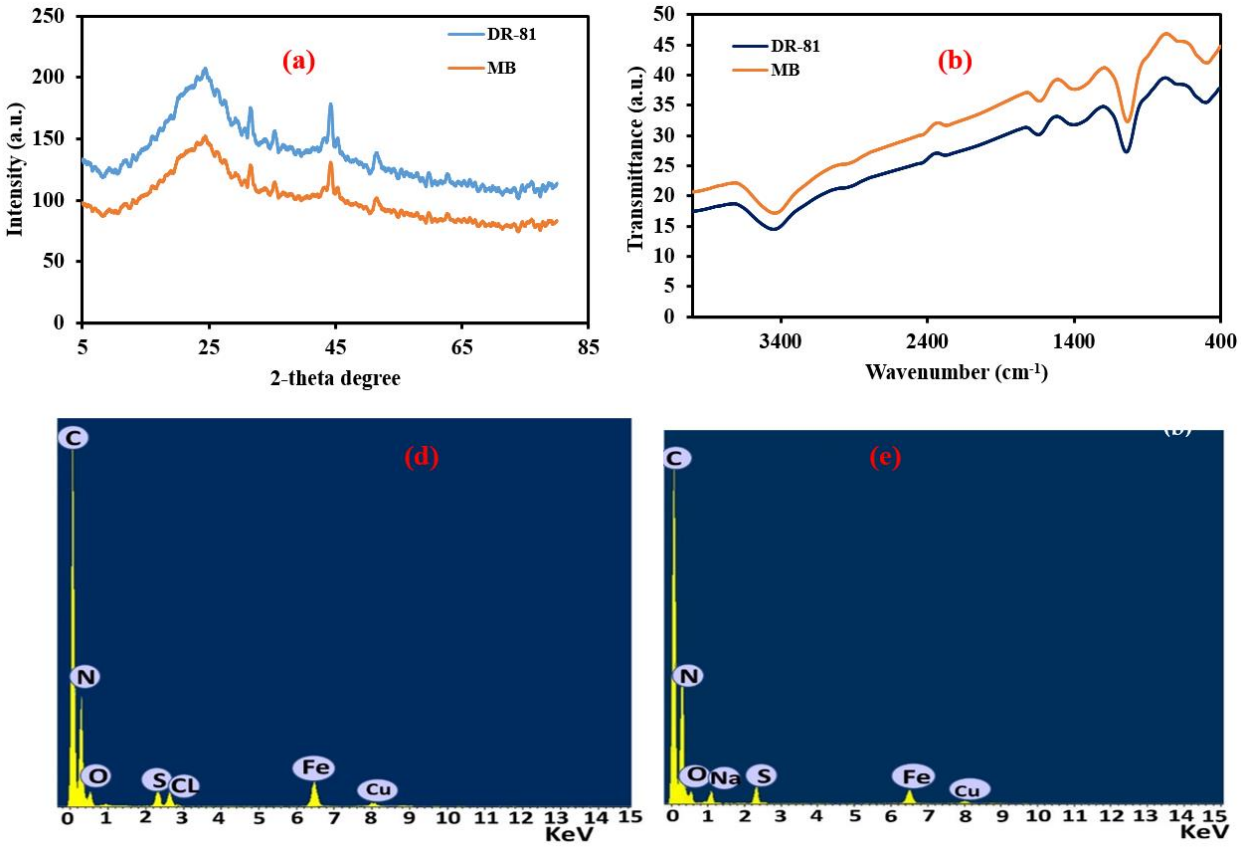

Figure S3. (**a**) XRD pattern, (**b**) FTIR spectra and EDXof ZVIgraphene after adsorption of (**d**) MB and (**e**) DR-81.

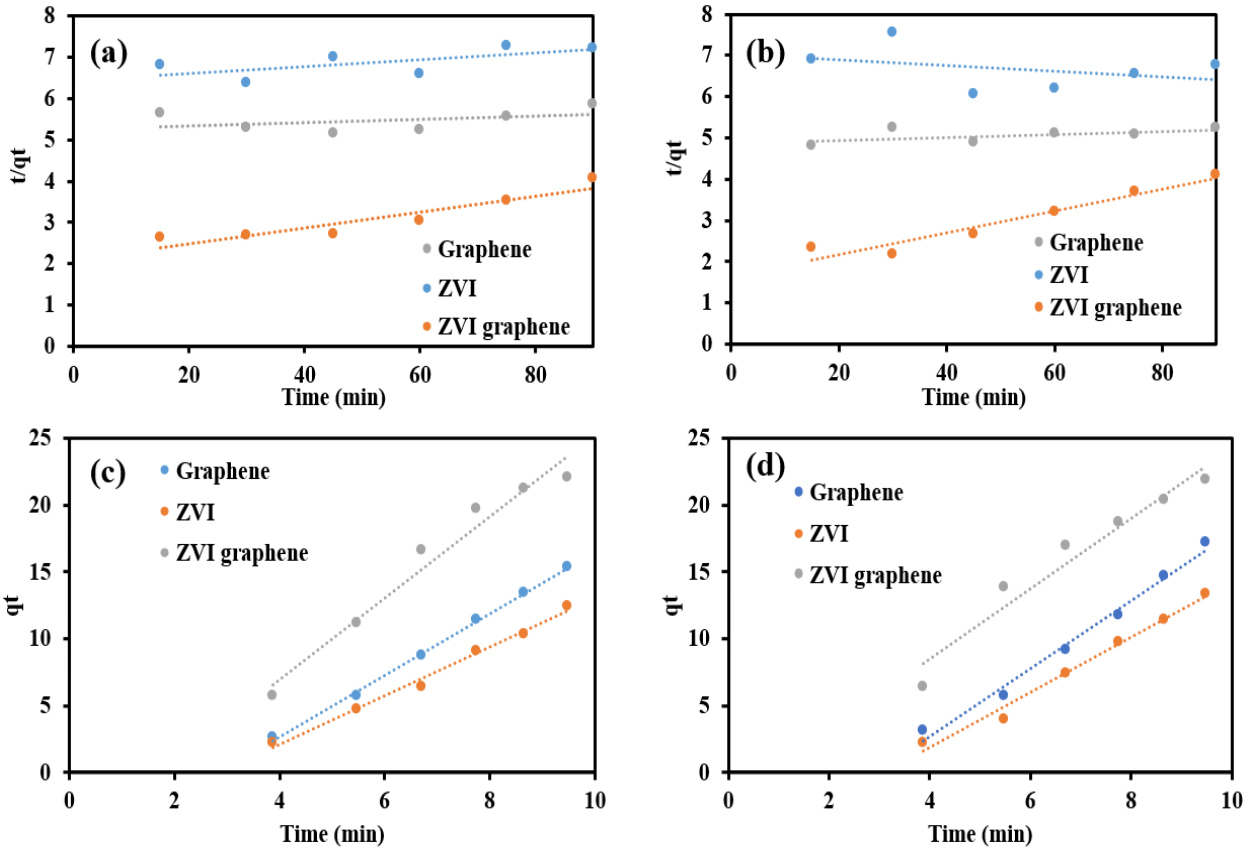

Figure S4. Adsorption kinetics (**a**) second-ordermodel of DR-81, (**b**) second-order model of MB, (**c**) intraparticle diffusion model of DR-81 and (**d**) intraparticle diffusion model of MB for adsorption on ZVI, graphene, and ZVIgraphene.

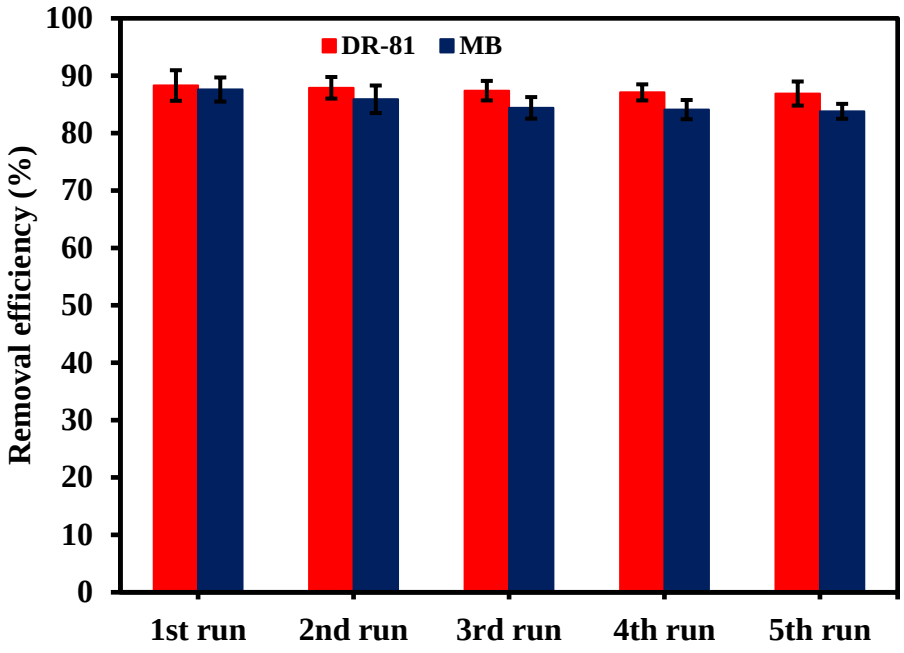

Figure S5. Stability of the prepared composite.

Table S1. Constant rates and coefficients of determination of first-order, second-order andintraparticle diffusionkinetic models.

Kinetic Model	DR-81	MB
First-order model Graphene	R^2 = 0.947 K_1 = 0.0273 min^{-1} q_e = 18.76 mg/g	R^2 = 0.929 K_1 = 0.026min^{-1} q_e = 20.6 mg/g
ZVI	R^2 = 0.96 K_1 = 0.0236 min^{-1} q_e = 14.33 mg/g	R^2 = 0.94 K_1 = 0.0245min^{-1} q_e = 16.36 mg/g
ZVIgraphene	R^2 = 0.95 K_1 = 0.0434 min^{-1} q_e = 30.17 mg/g	R^2 = 0.99 K_1 = 0.0352 min^{-1} q_e = 23.8 mg/g
Second-order model Graphene	R^2 = 0.1466 K_2 = 0.0000072 g/mg/min q_e = 263.2 mg/g	R^2 = 0.338 K_2 = 0.0000028 g/mg/min q_e = 270.27 mg/g
ZVI	R^2 = .42 K_2 = 0.0000107 g/mg/min q_e = 120.5 mg/g	R^2 = 0.126 K_2 = 0.0000065 g/mg/min q_e = 147 mg/g
ZVIgraphene	R^2 = 0.86 K_2 = 0.00017 g/mg/min q_e = 52.08 mg/g	R^2 = 0.93 K_2 = 0.00043 g/mg/min q_e = 37.6 mg/g
Intraparticle diffusion model Graphene	R^2 = 0.9975 k_d= 2.3 mg/g/$min^{0.5}$ c = 6.6 mg/g	R^2 = 0.98 k_d= 2.55 mg/g/$min^{0.5}$ c = 7.55 mg/g
ZVI	R^2 = 0.99 k_d= 1.8 mg/g/$min^{0.5}$ c = 5.14 mg/g	R^2 = 0.98 k_d= 2.06 mg/g/$min^{0.5}$ c = 6.4 mg/g
ZVIgraphene	R^2 = 0.96 k_d= 3.04 mg/g/$min^{0.5}$ c = 5.2 mg/g	R^2 = 0.94 k_d= 2.6 mg/g/$min^{0.5}$ c = 2 mg/g

K_1: Pseudo-first-order rate constant (min^{-1}), k_2: Pseudo-second-order rate constant (g/mg/min), K_d: Intraparticle diffusion rate constant (mg/g/$min^{0.5}$) and C: Boundary layer thickness (mg/g).

References

1. El Essawy, N.A.; Ali, S.M.; Farag, H.A.; Konsowa, A.H.; Elnouby, M.; Hamad, H.A. Green Synthesis of Graphene from Recycled PET Bottle Wastes for Use in the Adsorption of Dyes in Aqueous Solution. *Ecotoxicol. Environ. Saf.* **2017**, *145*, 57–68, doi:10.1016/j.ecoenv.2017.07.014.